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Energy levels of triply ionized carbon (C IV): Polarization method

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In a previous publication [Can. J. Phys. **75**, 11 (1997)] we calculated the generalized polarizabilities up to multipole order 3 as well as certain higher-order hyperpolarizabilities for two-electron atoms and ions of $Z=2-6$ and 10. In this paper we apply some of these results to calculate excited-state energies in three times ionized (lithiumlike) carbon. For states with angular momentum $L>3$ accurate results are obtained using an asymptotic polarizability expansion that includes nonadiabatic effects. Comparison is made with recent optical measurements, and a critical discussion of the correct form of the expansion is given. In addition, the possibility of very accurate measurements of the fine-structure splitting encourages us to present a table of such splittings to very high accuracy. An appendix contains similar results for lithiumlike oxygen and neon ions. [S1050-2947(99)07610-6]

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I. INTRODUCTION

In two recent papers [1] term energies for high Rydberg states of lithiumlike carbon (C IV) have been given and analyzed using both the quantum defect method and a polarization model following Edlén [2]. These are semiempirical techniques that describe deviations from the simple approximate hydrogenic level scheme. We have previously treated the high-*NL* Rydberg states of lithium itself by an *ab initio* polarization method [3]. This uses very accurately calculated values of the generalized core polarizabilities [4] appearing in the effective potential that acts between the two-electron core and the excited electron. After including effects of relativity and retardation we found excellent agreement with recent high-precision measurements of the fine structure splittings [5].

In more recent work, we extended the calculations of Ref. [4] to other systems, including carbon, and in the present paper we will use those results to compute theoretical values of Rydberg energy levels in C IV. An important point, which will be emphasized later, is the fact that the correct form of the effective potential differs from the form that has been traditionally used.

The familiar, basic idea of the method is to reduce the three-electron system to an effective one-electron system by eliminating the coordinates of the two-electron core. To do this we must find an effective interaction between the core and the outer electron and then calculate the energy shift of the outer electron by perturbation theory. The traditional way to do this is to use the adiabatic approximation for the outer electron. That is, one holds the outer electron fixed and computes the change in energy of the core due to polarization. This energy shift is obviously dependent on the position of the outer electron, so it gives rise to an adiabatic effective potential whose long-range leading term has the following form:

$$V(x)=-\sum_{l=1}\frac{\alpha_l}{x^{2l+2}}. \tag{1}$$

Here x is the position of the outer electron relative to the nucleus and α_l is the multipole polarizability of order l. In this approximation one calculates the expectation value of $V(x)$ for the hydrogenic state of interest; these expectation values are analytic and well known [6]. For a state of angular momentum L all terms up to $l=L$ give finite results; this is obviously not a convergent series and it must be truncated at its smallest term. In Ref. [2] this method is described, and the first two terms are used to calculate energy levels; the same method is also used in the more recent work on C IV [1].

But the adiabatic theory is not the whole story. Beginning with the term in $1/x^6$ the complete theory introduces additional terms due to the inability of the core to follow the motion of the outer electron exactly. These are usually called ‘‘nonadiabatic corrections,’’ and they serve to reduce the effect of the adiabatic polarization terms, which are all attractive. (This was emphasized by Dalgarno and Shorer a long time ago [7].) In the next section we will show the explicit form of the complete effective potential up to terms of order $1/x^8$.

II. THE EFFECTIVE POTENTIAL

Consider the C IV ion, with two electrons in a $1s^2\ {}^1S_0$ core state and the third electron in a highly excited state with quantum numbers (N,L). If we take the mass of the nucleus as M in electron mass units, the system is described by the following nonrelativistic Hamiltonian ($Z=6$ for carbon):

$$H_{\mathrm{NR}}=\left[-\nabla_1^2-\nabla_2^2-K\boldsymbol{\nabla}_1\cdot\boldsymbol{\nabla}_2-\frac{2Z}{r_1}-\frac{2Z}{r_2}+\frac{2}{r_{12}}\right]+\left[-\nabla_x^2-\frac{2(Z-2)}{x}\right]+\left\{-\frac{4}{x}+\frac{2}{|\vec{x}-\vec{r}_1|}+\frac{2}{|\vec{x}-\vec{r}_2|}\right\}, \tag{2}$$

where $K=2/(M+1)$ and the energy is in reduced Rydberg units $R=(1-K/2)$ Ry. The first bracket is the Hamiltonian of the core $H_0(12)$, the second of the Rydberg electron $H_0(x)$, and the braces contain the interaction between the two parts H_{int}, which decreases rapidly with increasing distance of the third electron x. (We neglect the difference between the reduced mass of an electron relative to the nucleus

TABLE I. Illustration of the contributions of the various correction terms in the case of the $N=8$ manifold in C IV. Results are given in units of cm^{-1}. (E_N is the nonrelativistic, hydrogenic binding energy of the Rydberg electron in the shielded Coulomb potential; it is independent of L.) An approximate value of the energy shift due to penetration and exchange is also given to indicate that it is of the same order as the estimated error in convergence for $L=3$ and is negligible for $L>3$.

	$L=3$	$L=4$	$L=5$	$L=6$	$L=7$
E_N	-27433.0745	-27433.0745	-27433.0745	-27433.0745	-27433.0745
Δ_2	-1.4208	-0.3777	-0.1284	-0.0511	-0.0225
D_2	-0.0011	-0.0001			
Δ_{rel}	-0.5609	-0.3754	-0.2573	-0.1756	-0.1157
D_{rel}	0.0021	0.0005	0.0002	0.0001	
Δ_{ret}	0.0009	0.0002	0.0001		
Δ_{Lamb}	-0.0002	-0.0001			
E_{total}	-27435.0545 ± 0.0435	-27433.8271 ± 0.0020	-27433.4599 ± 0.0001	-27433.3011	-27433.2127
Δ_{pen}	-0.037	-0.0002	-8×10^{-7}		
Δ_{exch}	-0.064	-0.0004	-1.3×10^{-6}		

and that of an electron relative to the two-electron core.) To a pretty good approximation, the energy of this system is $\epsilon_0-(Z-2)^2/N^2$, where ϵ_0 is the energy of the core, and the third electron moves in an almost pure Coulomb potential, partially shielded by the two electrons in the core. Our problem is to find the small corrections to this zero-order energy.

We do this by first expanding the interaction in a multipole series, with $x \gg r_{1,2}$:

$$H_{\text{int}}=\sum_{l=1}^{\infty}\sum_{j=1}^{2}\frac{2r_j^l}{x^{l+1}}P_l(\hat{r}_j\cdot\hat{x}) \tag{3}$$

and then calculate perturbations in the usual way, beginning with second order

$$\Delta_2=\sum\frac{\langle NL|\nu_n(x)|N'L'\rangle\langle N'L'|\nu_n(x)|NL\rangle}{(\epsilon_0-\epsilon_n)+(E_{NL}-E_{N'L'})}, \tag{4}$$

where $\nu_n(x)=\langle 0|H_{\text{int}}|n\rangle$. In principle, we could evaluate this sum, including excited states of both the core (n) and the outer electron ($N'L'$). Not only would this be somewhat difficult but it would have to be repeated for every value of N and L in which we were interested. Instead, we make the assumption (justified after the fact) that the excitation energies of the core are larger than those of the outer electronic system. Then we expand the denominator of Eq. (3) in a binomial series:

$$\Delta_2\approx\sum_n\frac{\langle NL|\nu_n(x)|N'L'\rangle\langle N'L'|\nu_n(x')|NL\rangle}{\epsilon_0-\epsilon_n}\times\left[1+\frac{E_{N'L'}-E_{NL}}{\epsilon_0-\epsilon_n}+\cdots\right]. \tag{5}$$

In the first term above it is easy to apply closure over the intermediate states $N'L'$ to obtain an effective ''adiabatic'' potential

$$V_{\text{ad}}(x)=\sum_n\frac{\nu_n(x)\nu_n(x)}{\epsilon_0-\epsilon_n}=-\sum_{l=1}\frac{\alpha_l}{x^{2(l+1)}} \tag{6}$$

whose expectation value $\langle NL|V_{\text{ad}}(x)|NL\rangle$ gives the leading correction to the energy. The second sum in Eq. (6) cannot be carried out to ∞ because the higher terms would diverge; the series is probably asymptotic and should be terminated at its smallest term.

The second term in Eq. (5) seems harder to convert into effective potential form because of the energy-dependent factor, but a simple trick solves this problem:

$$\sum_n\frac{\langle NL|\nu_n(x)|N'L'\rangle\langle N'L'|\nu_n(x)|NL\rangle(E_{N'L'}-E_{NL})}{(\epsilon_0-\epsilon_n)^2}=\left\langle NL\left|\sum_n\frac{\nu_n(x)[H_0(x),\nu_n(x)]}{(\epsilon_0-\epsilon_n)^2}\right|NL\right\rangle. \tag{7}$$

By converting the energy dependence to a commutator we are able to carry out the closure sum; the first time this trick was used may have been by Mittleman and Watson [8]. After commutation and integration by parts, this ''first nonadiabatic term'' takes the following form:

$$V_{\text{nad}}(x)=\frac{1}{2}\sum_n\frac{\nabla_x^2\nu_n^2(x)}{(\epsilon_0-\epsilon_n)^2}=\sum_l\frac{(l+1)(2l+1)\beta_l}{x^{2l+4}}, \tag{8}$$

where β_l is just like α_l but with one more power of the energy difference in the denominator, and higher terms can be computed in a similar way: γ_l has three powers of the energy difference in the denominator, δ is a third-order term, and ϵ is of fourth order.

In this way the effective potential can be written out to the term of order x^{-8} as follows:

$$U(x)=-\frac{\alpha_1}{x^4}+\frac{6\beta_1-\alpha_2}{x^6}+\frac{\delta+16\gamma_1/5}{x^7}+\frac{15\beta_2-\alpha_3-\epsilon+\alpha_1\beta_1-72\gamma_1[1+L(L+1)/10]}{x^8}. \tag{9}$$

In a series of papers [9], which are summarized in perhaps excessive detail in Ref. [10], this method was applied to the Rydberg states of helium, including terms up to order x^{-10},

TABLE II. Level energies of C IV in cm^{-1}, compared with those of Quinet [1]. Errors are estimated from the convergence rate of the effective potential $U(x)$, as described in Sec. IV.

$L=3$	ΔE	Level $E(NL)$	$E(NL)$ (Quinet)
$N=4$	−11.48	−109743.78 ± 0.18	−109744.4
5	−6.86	−70235.53 ± 0.13	−70234.6
6	−4.310	−48774.220 ± 0.090	−48773.5
7	−2.856	−35833.810 ± 0.062	−35834.0
8	−1.980	−27435.055 ± 0.043	−27435.1
9	−1.425	−21676.941 ± 0.032	−21677.1
10	−1.140	−17558.308 ± 0.024	−17558.3
11	−0.869	−14510.925 ± 0.018	−14510.9
12	−0.677	−12193.155 ± 0.014	−12193.2
$L=4$			
$N=5$	−2.133	−70230.804 ± 0.004	−70230.7
6	−1.487	−48771.397 ± 0.003	−48770.1
7	−1.045	−35831.999 ± 0.003	−35830.3
8	−0.752	−27433.827 ± 0.002	−27432.1
9	−0.556	−21676.072 ± 0.002	−21676.1
10	−0.421	−17557.589 ± 0.001	−17557.6
11	−0.330	−14510.386 ± 0.001	−14510.4
12	−0.260	−12192.738 ± 0.001	−12192.7
$L=5$			
$N=6$	−0.654	−48770.564 ± ⩽0.0001	−48769.9
7	−0.506	−35831.460	−35830.1
8	−0.385	−27433.460	−27432.1
9	−0.296	−21675.812	−21674.5
10	−0.230	−17557.398	−17557.4
11	−0.181	−14510.237	−14510.2
12	−0.145	−12192.623	−12192.6

TABLE III. Theoretical fine structure intervals of C IV in MHz, including all the effects discussed above. The errors shown are of the two levels combined in quadrature.

Transition	Interval (MHz)	Transition	Interval (MHz)
5F-5G	$(141.7\pm4.0)\times10^{3}$	8I-8K	2 652.15 ± 0.20
6F-6G	$(84.6\pm2.7)\times10^{3}$	9I-9K	1 884.71 ± 0.18
7F-7G	$(54.3\pm1.8)\times10^{3}$	10I-10K	1 385.43 ± 0.15
8F-8G	$(36.8\pm1.3)\times10^{3}$	11I-11K	1 047.28 ± 0.13
9F-9G	$(26.05\pm0.95)\times10^{3}$	12I-12K	810.41 ± 0.11
10F-10G	$(21.54\pm0.71)\times10^{3}$		
11F-11G	$(16.16\pm0.54)\times10^{3}$	9K-9L	1228.623 ± 0.022
12F-12G	$(12.5\pm0.42)\times10^{3}$	10K-10L	902.351 ± 0.020
		11K-11L	681.665 ± 0.018
6G-6H	24 984 ± 98	12K-12L	527.232 ± 0.015
7G-7H	16 150 ± 78		
8G-8H	11 001 ± 60	10L-10M	648.6395 ± 0.0034
9G-9H	7 814 ± 47	11L-11M	489.6397 ± 0.0031
10G-10H	5 742 ± 36	12L-12M	378.4989 ± 0.0028
11G-11H	4 454 ± 29		
12G-12H	3 438 ± 23	11M-11N	374.5652 ± 0.0007
		12M-12N	289.3936 ± 0.0006
7H-7I	6 983.1 ± 3.0		
8H-8I	4 763.6 ± 2.6	12N-12O	230.77032 ± 0.00016
9H-9I	3 386.7 ± 2.1		
10H-10I	2 490.4 ± 1.7		
11H-11I	1883.0 ± 1.4		
12H-12I	1466.9 ± 1.1		

with considerable success. Of course, in that case all the coefficients that appear in the effective potential can be evaluated exactly and analytically, since the core is simply a hydrogenic ion. In the case of lithiumlike systems, the core contains two electrons, and the sums giving the various coefficients must be performed numerically. To calculate them, we used the method of pseudostate summation; that is, we replaced the complete set of states n by a finite and discrete set of pseudostates. These are in the form of prediagonalized Hylleraas expansions, and the eigenvalues ϵ_n are replaced by the discrete expectation values of the core Hamiltonian $H_0(12)$.

At this point it is worthwhile to examine the difference between the form of the effective potential shown in Eq. (9) and the adiabatic potential. In the particular case of C IV the coefficient of the term in x^{-6} in the present form, $6\beta_1-\alpha_2$, is almost exactly equal to $+\alpha_2$, just the negative of the adiabatic value. (In fact [11], the ratio of these two quantities is 0.9988, for the case of ^{12}C IV, the most abundant isotope.) So it is clear that for any level of C IV for which this term is significant we will find exactly the opposite effect from the adiabatic model prediction.

III. OTHER ENERGY CORRECTIONS

There are several other corrections to the energy levels in addition to the expectation value of the effective potential $U(x)$ that must be included. First, we should include second-order effects of $U(x)$. Since we have included terms of order x^{-8} in the effective potential, it is reasonable to include its leading term (x^{-4}) in second-order perturbation theory as well. That is,

$$D_2=\sum_{N'\neq N}\frac{|\langle NL|\alpha_1/x^4|N'L\rangle|^2}{E_N-E_{N'}}. \tag{10}$$

An analytic expression for this second-order energy shift has been derived [12], and we will use it here.

The remaining corrections are relativistic or quantum electrodynamic. The first of these simply adds the Sommerfeld-Dirac correction of order α^2 to the nonrelativistic energy of the outer electron:

$$\Delta_{\text{rel}}=\frac{(Z-2)^4\alpha^2}{N^3}\left[\frac{3}{4N}-\frac{1}{J+1/2}\right], \tag{11}$$

where $J=L\pm1/2$. The difference between these two values of J should account well for the fine-structure splitting in those cases, with high enough values of L, for which the hydrogenic approximation is good. Otherwise, an appropriate J-weighted average over the two fine-structure levels can be used, which corresponds to replacing J by L.

The final three small corrections have been discussed previously in detail for the case of lithium [3], and the same methods are used here. The first is the shift D_{rel} due to relativistic changes in the motion of the inner electrons which is reflected in modification of the polarizability of the core. It is

TABLE IV. Theoretical fine-structure intervals of O VI in MHz, including all the effects discussed above.

Transition	Interval (MHz)	Transition	Interval (MHz)
5F-5G	$(292.0\pm 7.4)\times 10^3$	8I-8K	10 374.114±0.048
6F-6G	$(172.9\pm 5.0)\times 10^3$	9I-9K	7 318.806±0.064
7F-7G	$(110.3\pm 3.4)\times 10^3$	10I-10K	5 352.473±0.063
8F-8G	$(74.6\pm 2.4)\times 10^3$	11I-11K	4 030.875±0.056
9F-9G	$(52.7\pm 1.8)\times 10^3$	12I-12K	3 110.355±0.048
10F-10G	$(42.8\pm 1.3)\times 10^3$		
11F-11G	$(32.1\pm 1.0)\times 10^3$	9K-9L	5 279.4023±0.0044
12F-12G	$(24.84\pm 0.78)\times 10^3$	10K-10L	3 858.6529±0.0071
		11K-11L	2 904.6034±0.0075
6G-6H	67 015±24	12K-12L	2 240.5296±0.0071
7G-7H	42 785±22		
8G-8H	28 915±18	10L-10M	2 951.0675±0.0005
9G-9H	20 430±14	11L-11M	2 220.6258±0.0010
10G-10H	14 956±11	12L-12M	1 712.4654±0.0012
11G-11H	11 463.8±8.5		
12G-12H	8 840.0±6.8	11M-11N	1 765.04803±0.000 09
		12M-12N	1 360.85578±0.000 18
7H-7I	23 475.94±0.78		
8H-8I	15 852.85±0.86	12N-12O	1111.78432±0.000 02
9H-9I	11 194.49±0.77		
10H-10I	8 192.33±0.64		
11H-11I	6 172.55±0.53		
12H-12I	4 779.46±0.43		

TABLE V. Theoretical fine-structure intervals of Ne VIII in MHz, including all the effects discussed above.

Transition	Interval (MHz)	Transition	Interval (MHz)
5F-5G	$(58.2\pm 1.0)\times 10^4$	8I-8K	30 335.72±0.76
6F-6G	$(34.16\pm 0.70)\times 10^4$	9I-9K	21 345.46±0.70
7F-7G	$(21.69\pm 0.48)\times 10^4$	10I-10K	15 582.80±0.61
8F-8G	$(14.60\pm 0.34)\times 10^4$	11I-11K	11 719.39±0.52
9F-9G	$(10.29\pm 0.25)\times 10^4$	12I-12K	9 033.83±0.44
10F-10G	$(8.09\pm 0.18)\times 10^4$		
11F-11G	$(6.07\pm 0.14)\times 10^4$	9K-9L	15 929.536±0.082
12F-12G	$(4.69\pm 0.11)\times 10^4$	10K-10L	11 625.013±0.078
		11K-11L	8 740.927±0.071
6G-6H	$(1.6500\pm 0.0036)\times 10^5$	12K-12L	6 736.771±0.062
7G-7H	$(1.0469\pm 0.0029)\times 10^5$		
8G-8H	$(0.7048\pm 0.0022)\times 10^5$	10L-10M	9 059.354±0.012
9G-9H	$(0.4966\pm 0.0017)\times 10^5$	11L-11M	6 810.704±0.012
10G-10H	$(0.3629\pm 0.0014)\times 10^5$	12L-12M	5 248.483±0.011
11G-11H	$(0.2756\pm 0.0011)\times 10^5$		
12G-12H	21 241±85	11M-11N	5 472.9347±0.0024
		12M-12N	4 217.1917±0.0023
7H-7I	64 684±11		
8H-8I	43 491.1±9.8	12N-12O	3 468.36372±0.00054
9H-9I	30 621.3±8.2		
10H-10I	22 362.6±6.6		
11H-11I	16 823.4±5.4		
12H-12I	12 988.2±4.4		

calculated using third-order perturbation theory including the Breit-Pauli operator. The second is the retardation correction $\Delta_{\rm ret}$, due to the finite light propagation time between the core and a highly excited valence electron. We have used the expression for this correction given by Au *et al.* [13], carefully evaluated for the case of a two-electron core. Finally, we have included the two-electron Lamb shift in the presence of the perturbing valence electron, using the expression given by Goldman and Drake [14]:

$$\Delta^{NL}_{\rm Lamb}=\frac{8}{3}Z\alpha^3\left[-2\ln\alpha+\frac{19}{30}-\ln K\right]\langle\delta(\vec{r}_1)+\delta(\vec{r}_2)\rangle, \tag{12}$$

where $\ln K=2.984129+2\ln(Z-0.00615)$. Again, the expectation value of the delta functions is evaluated using third-order perturbation theory.

IV. THEORETICAL ENERGY LEVELS

We can now calculate theoretical term values for any L sufficiently high that penetration of the valence electron into the core and exchange between core and valence electrons give negligible contributions. As an example, in Table I we show each of the energy contributions discussed above for the $N=8$ manifold and the total energy of each L sublevel, including the norelativistic hydrogenic main term, relative to the ionization limit. Since the contribution of the effective potential Eq. (9) is only asymptotic, we terminate the series of expectation values at its smallest term keeping $\frac{1}{2}$ of that term and estimating the error also as $\frac{1}{2}$ the smallest term. This is a fairly conservative procedure, and it has been very successful in previous cases. We also show some typical values for the penetration and exchange contributions, calculated using the open-shell approximation for the two-electron core wave function; it is clear that these contributions are insignificant for $L>3$, and we do not include them in the total energy even for $L=3$ since they are of the same order of magnitude as our estimate of the asymptotic convergence of the polarization energy.

In Table II we compute many term energies and compare them with those of Quinet [1]. In this reference the lower term values are obtained from experiment, while the higher ones are derived by an effective potential technique like ours, but with what we have described above as the incorrect term $-\alpha_2/x$ instead of $(6\beta_1-\alpha_2)/x$ in Eq. (9). Note that for these upper levels the difference is very small: for $N=10$ and $L=5$ this term contributes only 4×10^{-4} cm^{-1}, a completely negligible amount in comparison with the errors in the optical measurements.

Nevertheless, there is a good chance that the details of the calculation, including the small relativistic corrections and others, can be tested. The technique of Lundeen and his collaborators has been used to measure the fine structure of excited states of helium [15], lithium [5], and the hydrogen molecule [16] to extremely high precision, and it appears that the same methods could be applied to C IV. For that reason, in Table III we give the fine-structure differences for many C IV levels.

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APPENDIX

Although we were originally motivated to carry out the computation of energy levels for C IV because of the work in Ref. [1], the possibility of making very accurate measurements of the fine structure by Lundeen's methods has encouraged us to present additional data for two more lithium-like ions O VI and Ne VIII. In Tables IV and V we show theoretical fine structure intervals for these two ions, in analogy with Table III for C IV.

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