Comparisons of the QED and relativistic parts of the triplet-state energies in the heliumlike sequence

H. G. Berry and R. W. Dunford
Physics Division, Argonne National Laboratory, Argonne, Illinois 60439

A. E. Livingston
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556
(Received 10 June 1992)

We have made a careful comparison of all available high-precision data for the $1s2s\,^3S_1$–$1s2p\,^3P_{0,1,2}$ transition energies for all nuclear charges ranging from helium ($Z=2$) to uranium ($Z=92$). Systematic discrepancies appear between theory and experiment for the $J=1\rightarrow0$ transitions, but not for the $J=1\rightarrow1$ and $J=1\rightarrow2$ transitions. Analysis of laser-based high-resolution data ($Z=2$, 3, and 5), beam-foil spectroscopy data ($Z=10$–17), and hyperfine-quenched decay data ($Z>27$) suggests an additional contribution to the $^3P_0$ energy equal to 2.3 $(Z/10)^4$ cm^{-1}.

PACS number(s): 31.30.Jv, 32.30.Jc

The heliumlike two-electron atomic system has been a valuable testing ground for *ab initio* calculations of the fully relativistic electromagnetic interaction between charged particles. It is one of the simplest systems where the understanding of the quantum field-theoretical treatment of a bound system of many particles still poses interesting unsolved problems. In this paper we shall focus on the energies of the $1s2p\,^3P$ states relative to the $1s2s\,^3S$ level.

At low nuclear charge Z, the energy separation is dominated by the nonrelativistic electron-electron Coulomb interaction that removes the degeneracy occurring in the one-electron hydrogenic system. At higher Z, relativistic interactions become rapidly more important through the increased nuclear potential, with terms proportional to Z^2, Z^4, and higher. Note that the nonrelativistic interactions scale as Z, and can be expanded in powers of $1/Z$. Precise variational calculations of the nonrelativistic Hamiltonian have yielded steadily more precise results, and the work of Drake [1] gives these energy values to higher precision than any of the uncertainties being raised here for the relativistic many-body interactions.

The relativistic electromagnetic interactions of the heliumlike system include both the standard Lamb shift corrections, virtual photons and e^+e^- pair production, of the Lamb shift of the hydrogen atom, and the relativistic corrections to the Coulomb force between the two charged electrons. Some of these interactions have only been calculated to lowest order [1]. The complications can be explained by the two interaction diagrams shown in Fig. 1. Neither diagram occurs in hydrogen and neither has been calculated completely. The vertex graph in Fig. 1(a) represents a photon exchange between the two electrons at the same time as the production of a virtual photon (Lamb shift) by one of the electrons. In first order, this represents a screening of the effective Lamb shift and leads to a $Z^4(1/Z)=Z^3$ correction in the two-electron Lamb shift. This was noted following the first accurate (to our knowledge) high-Z measurements of the two-electron Lamb shift (see Table I below). Higher-order relativistic corrections will lead to both Z^4 and Z^2 corrections to the level energies. The second diagram, the "box graph" of Fig. 1(b), represents the fully relativistic two-photon exchange between the two electrons. Z^4 and higher-order corrections remain to be calculated for this diagram.

The benchmark unified calculations of Drake [1] incorporate precise variational wave functions into a calculation of each specified relativistic and QED interaction. The partial absence of the above terms (those of Fig. 1) in his calculations led Drake [1] to project an estimated error in his energies of a term proportional to Z^4, with a magnitude of $1.2(Z/10)^4$ cm^{-1}.

In this paper, we show that the benchmark theory for the $1s2s\,^3S_1-1s2p\,^3P_J$ transition energies agrees with experiment to well within the estimated errors for the $J=1\rightarrow2$ transitions, but not for the $J=1\rightarrow0$ transitions.

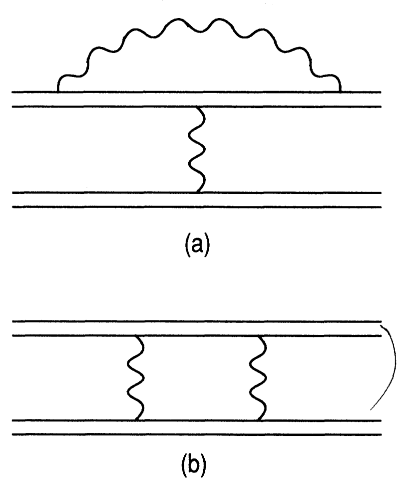

FIG. 1. Second-order relativistic interactions in a two-electron atom. The double line represents an electron in the field of a nucleus.

After considering the most precise measurements, we find a systematic deviation for $J=1\rightarrow 0$ transitions of approximately $2.3(Z/10)^4$ cm^{-1}. This deviation becomes apparent only when considering three recent and different types of measurements of the $^3S_1-{}^3P_0$ transitions over a large range of Z values. The purpose of this paper is to make this clear and to emphasize that there is a need for a conclusive measurement to verify these observed differences. Recent theoretical work of Johnson and Sapirstein [2] confirms the Z^4 dependent discrepancy for nuclear charges Z between 10 and 36. The three techniques are introduced below.

At low nuclear charge $Z=2,3,5$, precise measurements of the absolute transition energies have been made by laser-induced fluorescence techniques [3–5]. These techniques provide wavelength precisions in the parts per billion range. However, the relativistic energy corrections are small, and the missing Z^4 dependent corrections are tested with a precision of about 2% in lithium [4] and 10% in boron [5].

At intermediate Z, $Z=5$ to 36, direct wavelength measurements by beam-foil spectroscopy and high-energy discharges [6–21] give measured wavelength precisions as low as 10 parts per million. At $Z=16$ and 17 the missing Z^4 dependent discrepancy is tested to a precision of about 20%.

At higher Z, lifetime measurements [22–25] give less direct measurements of the $^3P_0$ energy. For all ^3P_J levels, the electric dipole decay mode to the $1s2s\ ^3S$ state dominates at low Z, whereas at high Z, relativistic contributions produce relatively strong decay rates to the ground state $1s^2\ ^1S_0$ for the $J=2$ state through magnetic quadrupole radiation and for the $J=1$ state through mixing with the $1s2p\ ^1P_1$ state. Hyperfine mixing among the different P states dependent on their energy differences produces changes in their decay mean lives. Lifetime measurements at $Z=28$[22], $Z=47$[23], and $Z=64$[24] have been used to measure $^3P_0$-$^3P_1$ energy splittings. At $Z=92$, Munger and Gould [25] measured the $^3P_0$ lifetime. The lifetime depends on the inverse third power of the transition energy, and assuming the matrix elements are calculated correctly, this energy difference is deduced

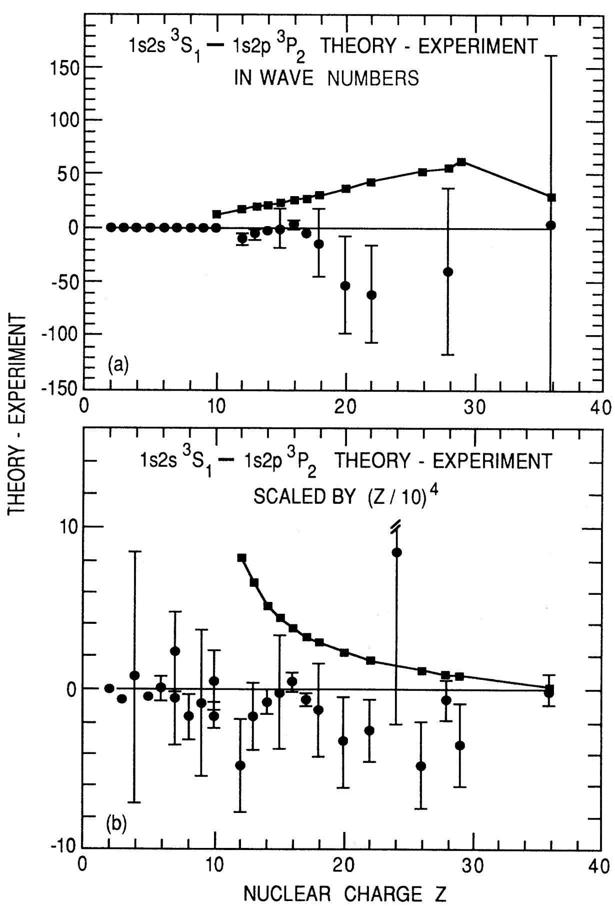

FIG. 2. Comparison of theory (Drake, Ref. [1]) and experiment for the $1s2s\ ^3S_1-1s2p\ ^3P_2$ transitions: (a) the differences in cm^{-1}. The circles are experimental values (see References, Table I); solid squares, Indelicato, Ref. [26]. (b) The same differences scaled by $(Z/10)^4$. Some data points in (a) with large error bars have been omitted in (b).

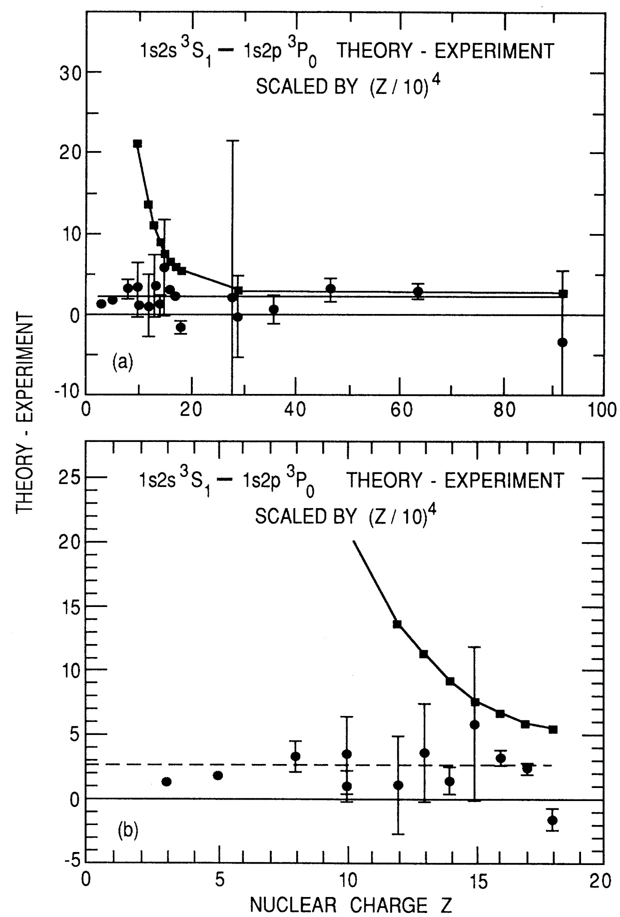

FIG. 3. Comparison of theory (Drake, Ref. [1]) and experiment for the $1s2s\ ^3S_1-1s2p\ ^3P_0$ transitions: (a) The differences in cm^{-1} between theory and experiment are scaled by $(Z/10)^4$. The circles are experimental values (Table I); solid squares, Indelicato, Ref. [26]. (b) Enlarged view for nuclear charges $3<Z<180$. The dashed horizontal line at $2.3(Z/10)^4$ in (a) and (b) is our estimated correction.

TABLE I. Theory and experiment for the heliumlike $1s2s\ ^3S_1-1s2p\ ^3P_0$ transition energies.

Nuclear charge	Reference	Transition Energy (cm^{-1}) Expt.	Theor. (Drake [1])	Difference: (Theor.)−(Expt.)
2	3	9 231.856 50(9)	9231.856 669	0.000 17
3	4	18 231.301 88(19)	18 231.312	0.010
4	6	26 864.6(2)	26 864.675	0.1
5	5	35 393.627(13)	35 393.736	0.109
6	7	43 899.12(96)	43 898.96	0.005
7	8	52 413.9(1.4)	52 420.97	6.1
7	9	52 420.0(1.1)	52 420.97	1.0
8	9	60 978.4(0.5)	60 979.65	1.3
9	10	69 586.0(3.0)	69 592.5	6.5
10	9	78 265.0(1.2)	78 265.9	0.9
10	11	78 262.6(3.0)	78 265.9	3.3
10	21	78 263.2(3.5)	78 265.9	2.7
12	12	95 851(8)	95 853	2
13	12	104 778(11)	104 787	10
14	13	113 815(4)	113 820	5
15	14	122 941(30)	122 970	29
16	15	132 219(4)	132 238	19
17	16	141 621(4)	141 640	19
18	17	151 204(9)	151 186	−18
28	22	256 125(1200)	256 240	115
29	18	267 950(360)	267 920	−30
36	20	357 400(300)	357 330	70
47	23	530 889(400)	532 253	1364
64	24	920 700(1500)	924 805	4105
92	25	$2097(64).10^3$	2 069 600	−27 400

from the lifetime measurement.

We first consider the energies of the transition $1s2s\ ^3S_1-1s2p\ ^3P_2$, shown in Fig. 2. The measurements agree with the calculations of Drake to well within his estimated uncertainty of $1.2(Z/10)^4$. Indelicato [26] has made a multiconfiguration relativistic Hartree-Fock calculation of these energies. At the low-Z limit, for $Z<20$, these calculations become inaccurate, and deviate significantly from experiment. At higher Z the calculations agree with Drake and experiment.

Accurate measurements of the transition energies $1s2s\ ^3S_1-1s2p\ ^3P_1$ are limited to the nuclear charges $Z=2$, 3, and 5 (Refs. [3–5]). The results are in good agreement with Drake's calculations, well within his estimated precision. An accurate measurement of the $1s2p\ ^3P$, $J=1\rightarrow2$ fine structure in fluorine [27], $957.88\pm0.03\ cm^{-1}$, also agrees with Drake's value of $957.38\ cm^{-1}$.

Measurements for the $1s2s\ ^3S_1-1s2p\ ^3P_0$ transition cover a much broader range of nuclear charge due to the combination of the different measurement techniques described above. In Table I, we list the most accurate measurements available for this transition. It must be noted that the higher Z measurements are indirect. The hyperfine-quenched lifetime measurements at $Z=28$, 47, and 64, all provide derived values of the energy splitting of the $1s2p\ ^3P$ $J=0$ to $J=1$ levels. To obtain the values in the Table, we have used the theoretical energies of Drake [1] for the $1s2p\ ^3P_1$ level to derive the tabulated values for the $1s2s\ ^3S_1-1s2p\ ^3P_0$ energies from the referenced lifetime measurements. The justification for this procedure is based on our observation that for the very precise laser-based measurements at $Z=3$, $Z=5$, and $Z=9$, no significant deviation from the Drake theory is found for the $J=1$ or $J=2$ components of the $1s2s\ ^3S-1s2p\ ^3P$ transitions, leaving only the $^3P_0$ component with a significant discrepancy.

Figures 3(a) and 3(b) show the results given in Table I. Only the results that are of sufficient precision to tests of a discrepancy at the order of $(Z/10)^4\ cm^{-1}$ are shown on the figures. We make the following conclusions from these data for the $^3P_0$ transition.

(i) The most accurate data, the laser-based measurements at $Z=3$ and 5 (Ref. [4] and [5]), the beam-foil data at $Z=14$, 16, and 17 (Refs. [13–16]), and the lifetime data at $Z=64$ (Ref. [23]), are consistent with a term added to the Drake theory of greater than $1.2(Z/10)^4\ cm^{-1}$. For $Z>9$, the correction is about $2.3(Z/10)^4$, more than twice Drake's estimated uncertainty.

(ii) The $Z=3,5$ laser measurements are significantly less than the dashed line of Fig. 3(b), when considering their very small error bars. This may be due to higher-order correlation effects proportional to powers of $Z^{-n}(\alpha Z)^m$, where the resulting Z power dependence is 2 or less.

(iii) The other measurements are consistent with this interpretation, except the result at $Z=18$. However, this measurement [17] used fast beam excitation of an argon gas target, and the transition was weak compared to neighboring spectral lines. A new measurement at $Z=18$ would be very helpful in resolving this difference.

(iv) The calculations of Indelicato [26], although inaccurate at lower Z, confirm at higher Z the magnitude and sign of the Z^4 dependent correction proposed.

(v) New relativistic many-body perturbation-theory calculations of Johnson and Sapirstein [2] also show a deviation from the work of Drake. From their calculations of $Z=10$ to 36, they project a needed correction of $2.6(Z/10)^4$ for $Z=10$ and $2.9(Z/10)^4$ for $Z>12$. This is in excellent agreement with the more extensive set of data presented here.

(vi) The energies of the $1s2p\ ^3P$, $J=1$, and $J=2$ levels agree with the calculations of Drake to within his estimated uncertainty, and to within $0.5(Z/10)^4$. For the case of $J=1$, there are no experimental data to directly justify this conclusion above $Z=9$, and no accurate $J=2$ data above $Z=36$.

We conclude that the major theoretical discrepancy lies in the energy of the $^3P_0$ level, whereas the energy of the $^3P_2$ level relative to the $^3S_1$ level is in good agreement with experiment. Further progress in this high-precision field may best be made through fine-structure measurements at medium and at very high nuclear charge Z. The former range is where significant correlation effects are not dominated by the relativistic central field nuclear interaction. The latter range is important in verifying the high-field dependence of the electromagnetic interaction between two charged particles in a bound state.

We thank W. R. Johnson and J. Sapirstein for communication of their results in advance of publication. This was supported in part (A.N.L.) by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No W-31-109-ENG-38.

[1] G. W. F. Drake, Can. J. Phys. **66**, 586 (1988).
[2] W. R. Johnson and J. Sapirstein, Phys. Rev. A **46**, 2197 (1992).
[3] P. Zhao, J. R. Lawall, A. W. Kam, M. D. Lindsay, and F. M. Pipkin, Phys. Rev. Lett. **63**, 1593 (1989); C. J. Sansonetti, J. D. Gillaspy, and C. L. Cromer, *ibid.* **20**, 2539 (1990).
[4] E. Riis, H. G. Berry, O. Poulsen, S. A. Lee, and S. Y. Tang, Phys. Rev. A **33**, 3023 (1986).
[5] T. Dinneen, N. Berrah-Mansour, H. G. Berry, L. Young, and R. Pardo, Phys. Rev. Lett. **66**, 2859 (1991).
[6] M. Eidelsberg, J. Phys. B **5**, 1031 (1972); B. Löfstrand, Phys. Scr. **8**, 57 (1973).
[7] B. Edlén and B. Löfstrand, J. Phys. B **3**, 1380 (1970).
[8] S. C. Baker, J. Phys. B **6**, 709 (1973).
[9] M. F. Stamp, D. Phil. thesis, Oxford University, 1983; N. J. Peacock, M. F. Stamp, and J. D. Silver, Phys. Scr. **T8**, 10 (1984).
[10] W. Engelhardt and J. Sommer, Astrophys. J. **167**, 201 (1971).
[11] H. G. Berry and J. E. Hardis, Phys. Rev. A **33**, 2778 (1986).
[12] H. A. Klein, F. Moscatelli, E. G. Meyers, E. H. Pinnington, J. D. Silver, and E. Träbert, J. Phys. B **18**, 1483 (1985).
[13] R. DeSerio, H. G. Berry, R. L. Brooks, J. Hardis, A. E. Livingston, and S. J. Hinterlong, Phys. Rev. A **22**, 1109 (1981).
[14] A. E. Livingston and S. J. Hinterlong, Nucl. Instrum. Methods **202**, 103 (1982); A. E. Livingston *et al.*, J. Phys. (Paris) Colloq. **50**, C1-255 (1989), and references therein.
[15] E. J. Galvez, Ph.D. thesis, University of Notre Dame, 1986; A. E. Livingston *et al.* (unpublished).
[16] A. S. Zacarias, Ph.D. thesis, University of Notre Dame, 1990; A. E. Livingston *et al.* (unpublished).
[17] H. F. Beyer *et al.*, Z. Phys. D **1**, 65 (1986).
[18] J. P. Buchet *et al.*, Phys. Rev. A **23**, 3354 (1981).
[19] J. Désesquelles, Nucl. Instrum. Methods **B31**, 30 (1988).
[20] S. Martin, A. Denis, M. C. Buchet-Poulizac, J. P. Buchet, and J. Désesquelles Phys. Rev. A **42**, 6570 (1990).
[21] W. A. Hallett, D. D. Dietrich, and J. D. Silver (private communication).
[22] R. W. Dunford *et al.*, Phys. Rev. A **44**, 764 (1991).
[23] R. Marrus *et al.*, Phys. Rev. Lett. **63**, 502 (1989). For the numbers, quoted with permission of the authors see Bull. Am. Phys. Soc. **37**, 1144 (1992).
[24] P. Indelicato *et al.*, Phys. Rev. Lett. **68**, 1307 (1992).
[25] C. T. Munger and H. Gould, Phys. Rev. Lett. **57**, 2927 (1986).
[26] P. Indelicato, Nucl. Instrum. Methods **B31**, 14 (1988).
[27] E. Myers *et al.*, Phys. Rev. Lett. **47**, 87 (1981).