Quantum electrodynamic effects in atomic structure

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Summary. In high-Z atoms, quantum electrodynamic (QED) corrections are an important component in the theoretical prediction of atomic energy levels. The main QED effects in electronic atoms are the one-electron self-energy and vacuum-polarization corrections which are well known. At the next level of precision, estimates of the effect of electron interactions on the self energy and higher-order effects in two exchanged photon corrections are necessary. These corrections can be evaluated within the framework of QED in the bound interaction picture. For high-Z few-electron atoms, this approach provides a rapidly converging series in $1/Z$ for the corrections, which is the generalization of the well-known relativistic $1/Z$ expansion methods. This paper describes recent work on the effect of electron interactions on the self energy. The QED effects are particularly important for the theory for lithiumlike uranium where an accurate measurement of the Lamb shift has been made, as well as for numerous other cases where systematic differences appear between theory that does not include these QED effects and experiment.

Key words: High-Z atoms – QED effects – Lithiumlike uranium – Self energy – Lamb shift

1. Introduction

The basis for atomic structure theory presently appears to be bound-state quantum electrodynamics (QED). With suitable approximations to the perturbation expansion and infinite summations of subsets of Feynman diagrams, one can recover the usual starting points for calculations such as the Schrödinger equation or the no-pair relativistic Hamiltonian for many-electron systems. This workshop is a testimony to the level of sophistication to which calculations based on such equations have been developed.

One might ask the complementary question: If no approximations whatsoever are made to the equations of bound-state QED, how far can the calculations

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of atomic structure be carried out? The immediate answer is not very far at all, but progress is being made in this approach. This paper describes current work along these lines.

High-Z few-electron atoms are systems that are highly suitable for such a study for several reasons. First, in high-Z systems the QED effects, i.e., effects that require infinite renormalization and effects that go beyond the no-pair relativistic Hamiltonian, are enhanced relative to binding and correlation effects. Second, atomic structure perturbation theory, which is closely coupled to QED perturbation theory, converges rapidly in these systems, and correlation effects are small. Third, and not least, experiments are being done on such systems, so the validity of the calculations can be checked. An example of an experiment on an extreme system of this kind is a recent measurement of the $2^2S_{1/2}-2^2P_{1/2}$ splitting in lithiumlike uranium done at the Bevalac at Lawrence Berkeley Laboratory [1]. For this system, the relativistic many-body perturbation theory (RMBPT) prediction for the splitting is approximately 322 eV and the QED correction to the splitting is of order -40 eV.

2. Basic theory

The theory, without approximations, is based on the bound-interaction (or Furry) picture in QED [2]. This approach is described elsewhere, so the details will not be repeated here [3, 4]. The starting point for uranium ions is the Dirac equation for electrons in the field of a bare $Z = 92$ nucleus. Interactions including QED effects are then perturbations, and level shifts are obtained by applying the Gell-Mann, Low, and Sucher prescription to the perturbation expansion [5, 6]. The result is expressions for the level shift that can be loosely associated with bound-state Feynman diagrams.

Enhancement of QED effects at high-Z is illustrated by comparing the approximate scaling of the QED effects:

$$E_{QED}^{(2)} \sim \alpha(Z\alpha)^4 mc^2 \tag{1}$$

to the order-of-magnitude of the second-order Coulomb splitting:

$$E_C^{(2)} \sim \left\langle \frac{\alpha}{r_{12}} \right\rangle \sim \alpha(Z\alpha)mc^2, \tag{2}$$

which gives

$$\frac{E_{QED}^{(2)}}{E_C^{(2)}} \sim (Z\alpha)^3. \tag{3}$$

This ratio is consistent with the order-of-magnitude of the calculated numbers for the RMBPT result and the QED correction mentioned above.

The rapid convergence of the perturbation expansion is illustrated by comparing the order of magnitude of the fourth-order Coulomb energy correction:

$$E_C^{(4)} \sim \left\langle \frac{\alpha}{r_{12}} \frac{1}{E_0 - H} \frac{\alpha}{r_{12}} \right\rangle \sim \frac{[\alpha(Z\alpha)mc^2]^2}{(Z\alpha)^2 mc^2} = \alpha^2 mc^2 \tag{4}$$

to the second-order Coulomb splitting given above by writing the ratio:

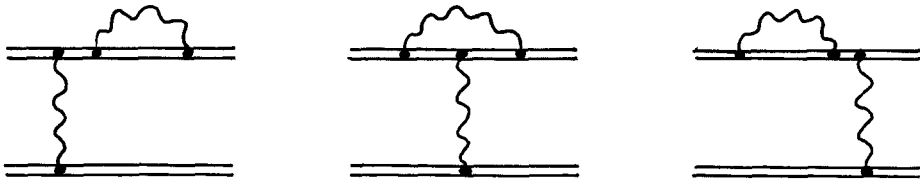$$\frac{E_C^{(4)}}{E_C^{(2)}} \sim \frac{1}{Z}. \tag{5}$$

Fig. 1. Feynman diagrams for the bound-state vertex correction. The *double lines* represent propagation of an electron in the nuclear Coulomb field. The *wavy lines* represent virtual photons
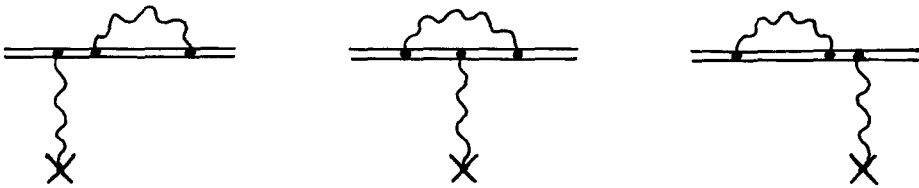

Fig. 2. Feynman diagrams for the first-order correction to the self energy of a bound electron by an external potential. The symbol × denotes the charge distribution that is the source of the external potential

For few-electron systems, where these estimates are valid, and for high Z, where the ratio is small, perturbation theory converges rapidly. In such a case, the difficulty of doing the exact calculations is offset by the fact that only a few terms are numerically important, so concrete predictions are possible.

This expansion is the generalization of the $1/Z$ expansion methods of Layzer and Bahcall [7] and of Dalgarno and Stewart [8]. This general approach has been discussed briefly by Bethe and Salpeter [9] and by Sucher [6], and more recent studies of non-QED electron-interaction corrections in this framework have been made by Ivanov, Ivanova, and Safronova [10].

In this approach the dominant corrections are given by the zero-order Dirac eigenvalues. The next corrections, second order in e, consist of the self-energy, vacuum polarization, and exchanged-photon correction. These corrections have been discussed in detail elsewhere [3].

The leading fourth-order corrections are the two-exchanged photon correction and the one-exchanged-photon vertex and vacuum polarization corrections. The non-QED exchanged-photon corrections, to a good approximation, are included in the RMBPT result of Blundell, Johnson, and Sapirstein [11]. We expect that the main contribution to the vertex correction, corresponding to the Feynman diagram in Fig. 1, will be of the form of a screening correction, corresponding to the diagram in Fig. 2, where the other electron provides an effective potential correction to the one-electron self-energy diagram. Such a calculation is described in the following sections.

3. Screening effect

The starting point for this calculation is the well-known expression for the self-energy level shift E_{SE}, valid for an arbitrary fixed external potential, that can be written as the sum $E_{SE} = E_L + E_H$ of a low-energy part E_L and a high-energy

part E_H [12], where (in units in which $\hbar = c = m_e = 1$)

$$E_L = \frac{\alpha}{\pi} E_n - \frac{\alpha}{\pi} P \int_0^{E_n} dz \int d\vec{x}_2 \int d\vec{x}_1 \, \phi_n^\dagger(\vec{x}_2) \alpha^j G(\vec{x}_2, \vec{x}_1, z) \alpha^l \phi_n(\vec{x}_1)$$

$$\times (\delta_{jl} \vec{\nabla}_2 \cdot \vec{\nabla}_1 - \nabla_2^j \nabla_1^l) \frac{\sin[(E_n - z)x_{21}]}{(E_n - z)^2 x_{21}} \tag{6}$$

and

$$E_H = -\frac{i\alpha}{2\pi} \int_{C_H} dz \int d\vec{x}_2 \int d\vec{x}_1 \phi_n^\dagger(\vec{x}_2) \alpha_\mu G(\vec{x}_2, \vec{x}_1, z) \alpha^\mu \phi_n(\vec{x}_1) \frac{e^{-bx_{21}}}{x_{21}}$$

$$- \delta m \int d\vec{x} \phi_n^\dagger(\vec{x}) \beta \phi_n(\vec{x}) \tag{7}$$

and where $b = -i[(E_n - z)^2 + i\delta]^{1/2}$, $\mathrm{Re}(b) > 0$. In these expressions, ϕ_n and E_n are the eigenfunction and eigenvalue of the Dirac equation for the bound state n, and G is the Green's function for the Dirac equation corresponding to the operator $G = (H - z)^{-1}$, where H is the Dirac Hamiltonian. In the Dirac equation, the potential V is an arbitrary external potential that could be the Coulomb potential, or a more complicated potential that includes a screening correction δV. The contour C_H extends from $-i\infty$ to $0 - i\epsilon$ and from $0 + i\epsilon$ to $+i\infty$, with the appropriate branch of b chosen in each case.

In this calculation, the screening effect is regarded as a small change in the external potential with respect to the background Coulomb potential:

$$V(x) = V_C(x) + \delta V(x) \tag{8}$$

and an expression for the correction is obtained by expanding the constituents of the exact Eqs. (6) and (7) to first order in powers of δV. The zeroth-order term in each case is just the self-energy correction corresponding to the Coulomb potential V_C. The first-order correction $\delta E_n^{(1)}$ can be written as the sum of three terms that correspond to the dependence of the level shift on δV through the bound-state energy E_n:

$$E_n \to E_n + \langle \phi_n | \delta V | \phi_n \rangle + \cdots \tag{9}$$

the bound-state wave function ϕ_n:

$$\phi_n \to \phi_n - \sum_{i \neq n} \frac{\langle \phi_i | \delta V | \phi_n \rangle}{E_i - E_n} \phi_i + \cdots \tag{10}$$

and the Green's function G:

$$G \to G - G\delta V G + \cdots . \tag{11}$$

These three terms give comparable numerical contributions to the total correction.

4. Singularities

Here, we outline a road map of potential divergences that would create problems in the numerical work if no provision were made to take them into account. First, there are the usual ultraviolet divergences associated with the self-energy calculation and mass renormalization. These divergences cancel for an arbitrary

potential, so the starting expression is finite. However, the individual terms in the perturbation expansion can contain both infrared and ultraviolet divergences that ultimately cancel, but still require attention.

In the high-energy part, the energy, wave function, and Green's function corrections all have ultraviolet divergences. The wave-function divergence in the main term cancels a similar divergence in the mass renormalization term that arises from the wave-function correction to that term. The divergence in the energy correction is canceled by an equal and opposite divergence in the Green's function correction. In each of these cases, we eliminated the divergence by the introduction of suitable subtraction terms.

If the low-energy part were calculated in the Feynman gauge, there would be equal and opposite infrared divergences in the corresponding energy correction and Green's function correction. However, in the Coulomb-gauge-like form for the low-energy part employed in this calculation, the integrand in the low-frequency photon limit is suppressed, and there is no infrared singularity.

In the low-energy part there is an additional spurious singularity in the principal-value integration. The energy correction to this term involves a derivative with respect to energy, with the result that the poles in the principal-value integral become second-order poles. This case can be dealt with by numerical techniques analogous to the method applied to evaluate the principal-value integral in earlier work [13]. In the present case, it is sufficient to replace the principal-value integration by the original definition of the integral as a contour integral in order to identify the correct result.

5. Numerical methods

The numerical methods employed here are similar to those that have been applied in a Coulomb self-energy calculation [14]. A few points of departure are outlined here.

The starting point for both the low-energy part and the high-energy part is to carry out an expansion of the electron Green's function in angular momentum eigenfunctions. This leads to an expression that consists of a contour integral over z, four radial coordinate integrals, and an infinite summation over κ, the angular momentum quantum number.

Each term in the sum over κ contains radial Green's functions for the Coulomb field. Methods for calculating these to high precision are known [14]. In the case of wave-function corrections, reduced radial Green's functions are needed. There are methods described in the literature for calculating these functions [15]. However, we took advantage of the availability of the full Green's functions and calculated them by numerical interpolation. In particular, the reduced Green's function, defined by:

$$G^R(\vec{x}_2, \vec{x}_1, E_n) = \sum_{E_i \neq E_n} \frac{\phi_i(\vec{x}_2)\phi_i^\dagger(\vec{x}_1)}{E_i - E_n}, \tag{12}$$

can be obtained from the full Green's function, defined by:

$$G(\vec{x}_2, \vec{x}_1, z) = \sum_i \frac{\phi_i(\vec{x}_2)\phi_i^\dagger(\vec{x}_1)}{E_i - z}, \tag{13}$$

with the aid of the limit:

$$G^R(\vec{x}_2, \vec{x}_1, E_n) = \lim_{\epsilon \to 0} \tfrac{1}{2}[G(\vec{x}_2, \vec{x}_1, E_n + \epsilon) + G(\vec{x}_2, \vec{x}_1, E_n - \epsilon)]. \tag{14}$$

In taking this limit, the terms with $E_i = E_n$ vanish identically because of the symmetry of the expression. We calculate the limit numerically for the radial Green's functions with the analogous four point symmetric interpolation formula.

The summation over κ is a time-consuming part of the calculation; many terms (of order 10^3–10^4) are needed due to slow convergence of the sum. We dealt with this by analytically calculating a four-term asymptotic expansion of the Green's functions for large $|\kappa|$, and summing these terms analytically in the large $|\kappa|$ region in order to accelerate the convergence.

An additional computer time saving measure was to evaluate the potential shift δV and the wave-function correction $\delta\phi$ on a fixed grid and then employ a cubic spline interpolation to evaluate these functions for the numerical integration.

The numerical integration was carried out by Gaussian quadrature.

It is useful in developing the computer code to have a known case on which to test the calculation, eliminate bugs, and check precision. Fortunately, there exists a natural test case for this calculation. In particular, if the value of Z in the Coulomb self energy is shifted by a small amount, the first-order correction is just the result corresponding to a perturbing potential proportional to the Coulomb potential. We took advantage of this fact to check the calculation extensively by comparing the effect of a Coulomb potential perturbation to the derivative with respect to Z of the known Coulomb self energy. In fact, this test was applied to each separate component of the calculation, such as the wave-function correction, or the correction to the Green's function.

6. Model calculation

For a demonstration calculation, we employ a model in which the screening potential for each electron is the sum over the spherically averaged potentials that arise from the charge distributions of the other electrons in the atom:

$$\delta V_n(x) = \sum_j \delta V^{(j)}(x) \tag{15}$$

where

$$\delta V^{(j)}(x_2) = \frac{1}{4\pi} \int d\Omega_2 \int d\vec{x}_1 \frac{\alpha}{|\vec{x}_2 - \vec{x}_1|} |\phi_j(\vec{x}_1)|^2 \tag{16}$$

and where ϕ_j is the unscreened Dirac hydrogenic wave function. Since the level shift is linear in the perturbing potential, the total energy shift δE_n for state n is just the sum over j of the energy shift $\delta E_n^{(j)}$ of level n due to screening by the electron in state j:

$$\delta E_n = \sum_j \delta E_n^{(j)}. \tag{17}$$

For lithiumlike uranium, examples of level shifts from preliminary results of this calculation are:

$$1s^2 2p \; 2^2P_{1/2}: \quad 2\delta E_{1s}^{(1s)} + 2\delta E_{1s}^{(2p)} + 2\delta E_{2p}^{(1s)} = -8.90 \, \text{eV}$$

$$1s^2 2s \; 2^2S_{1/2}: \quad 2\delta E_{1s}^{(1s)} + 2\delta E_{1s}^{(2s)} + 2\delta E_{2s}^{(1s)} = -11.41 \, \text{eV}.$$

Table 1. Transition energy

Contribution	Value
RMBPT	322.4 eV
One-electron QED	−42.8 eV
Self-energy screening	2.5 eV
Total theory	282.1 eV
Experiment	280.41(.13) eV

In order to compare theory to experiment for the $2^2S_{1/2}-2^2P_{1/2}$ transition energy in lithiumlike uranium, we list the relativistic many-body perturbation-theory (RMBPT) calculation of the non-QED contributions done by Blundell, Johnson, and Sapirstein [11], the one-electron QED corrections (excluding the gross effect of the finite nuclear size which is contained in the RMBPT contribution) [16], and the self-energy screening correction from the present work. The sum of these terms is compared to the experimental result of Schweppe et al. [1] in Table 1.

The difference between theory and experiment is of the order of uncalculated theoretical contributions. These include the screening correction to the vacuum polarization that is expected to be of order -1 ± 1 eV, higher-order retardation contributions to the two-exchanged-photon corrections [17], and contributions to the vertex diagram that are not included in the screening approximation.

Other estimates for the QED screening effects in this system include an empirical extrapolation from low Z [11], and models in which the self energy is represented by an effective potential in a multiconfiguration Dirac–Fock calculation [18, 19].

7. Outlook

Now that a demonstration calculation of screening corrections to the Coulomb field self energy has been carried out for excited states, a number of natural extensions of this work lie ahead. Results of similar calculations for lower Z are needed in order to check the corrections along isoelectronic sequences. Ultimately, one would hope to attack the problem of highly-correlated systems such as neutral helium. The techniques described here might be of use for this purpose.

It would be useful for a broad range of applications, if the calculation of the effects of deviations from a Coulomb potential on the self-energy correction were formulated in such a way that a simple evaluation for a given deviation could be made. In particular, since the main correction is linear in the correction to the potential, it should be possible to write it as an integral of a function of the radial coordinate multiplied by the correction to the potential. In this way, it would be necessary to tabulate only one function for a given state and nuclear charge that would readily yield the level-shift correction to the self energy for any of a broad range of corrections to the Coulomb potential. It should be possible to extract such a function in the context of the calculation described here.
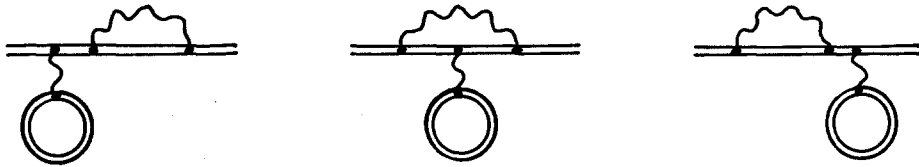
Fig. 3. Feynman diagrams for a hydrogenic QED correction of order α^2

Another by-product of this work is the possibility of evaluating other higher-order QED effects. For example, the diagram in which the potential correction considered above is replaced by a vacuum polarization bubble, as shown in Fig. 3, can be evaluated with essentially no additional development of techniques.

It requires only a relatively mild generalization of the approach described here to evaluate the full bound-state vertex correction, which is a necessary step in a rigorous evaluation of atomic level shifts in QED.

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