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2010 J. Phys. B: At. Mol. Opt. Phys. 43 074002

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Fundamental constants and tests of theory in Rydberg states of one-electron ions

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Received 10 September 2009, in final form 22 October 2009

Published 19 March 2010

Online at stacks.iop.org/JPhysB/43/074002

Abstract

The nature of the theory of circular Rydberg states of hydrogen-like ions allows highly accurate predictions to be made for energy levels. In particular, uncertainties arising from the problematic nuclear size correction which beset low angular-momentum states are negligibly small for the high angular-momentum states. The largest remaining source of uncertainty can be addressed with the help of quantum electrodynamics calculations, including a new nonperturbative result reported here. More stringent tests of theory and an improved determination of the Rydberg constant may be possible if predictions can be compared with precision frequency measurements in this regime. The diversity of information can be increased by utilizing a variety of combinations of ions and Rydberg states to determine fundamental constants and test theory.

1. Introduction

Quantum electrodynamics (QED), the quantum field theory of electrons and photons, is the first satisfactory quantum description of the interaction of charged particles (and antiparticles) via the exchange of photons and of the creation and annihilation of elementary particles. QED makes precise predictions of various physical quantities which have been tested across a vast array of phenomena. Spectroscopic measurements in atoms, however, have played a crucial role in spurring the development of QED to be the most accurate physical theory yet invented. With the quantization of the electromagnetic field, QED does not remove the divergences well known in Maxwell's classical theory of electromagnetism; in contrast, new infinities are found in QED, associated with virtual processes of the vacuum. Sensible, finite results are obtainable only after renormalization; this is an art inextricably tied to the introduction of fundamental constants, such as the electron mass and charge, into the theory.

The first tests of the emerging formalism of QED came shortly after World War II. Kusch and Foley (1947) reported the first measurement of the anomalous magnetic moment of the electron; in the same year, Lamb and Retherford (1947) presented the first measurement of the 'Lamb shift' in the 2s level of hydrogen (Lamb 1951), another departure from

the Dirac theory of the hydrogen atom. These discoveries have led to more stringent tests of QED, with remarkable progress over six decades. On the one hand, with control and minimization of cavity effects that limited early geonium $|g|-2$ experiments (Van Dyck *et al* 1987, Brown and Gabrielse 1986), the magnetic moment of the electron $|g| = 2(1 + a_e)$ has been measured recently at Harvard University with a relative uncertainty of 2.8×10^{-13} using a single electron isolated in a cylindrical Penning trap (Hanneke *et al* 2008); by comparison, the calculated magnetic moment of the electron has a relative uncertainty of 5.2×10^{-12} coming entirely from the uncertainty of the best independent determination of the fine-structure constant. On the other hand, the hydrogen 1s–2s transition has been measured with a relative uncertainty of 1.4×10^{-14} (Hänsch *et al* 2005); here a test of theory is hampered by uncertainties from nuclear size corrections, which currently limit the accuracy in the determination of the Rydberg constant to a relative uncertainty of 6.6×10^{-12} (Mohr *et al* 2008). It seems astonishing that QED should have attained such high accuracy in the abstractions employed for representing physical objects and measurements, particularly when one of its pioneers has noted the 'mathematical inconsistencies and renormalized infinities swept under the rug' (Dyson 2006).

Looking to the future, in this paper we consider possible determinations of fundamental constants and tests of theory in circular Rydberg states of one-electron ions. This has been

discussed by Jentschura *et al* (2008), where it was pointed out that the problems that limit the theoretical predictions in low angular-momentum states are strongly suppressed in circular Rydberg states, because the electron has a very small probability of being near the nucleus in such states. However, even though some problematic aspects of the theory are absent in these states, there is still a contribution from QED that needs to be accounted for in order to make accurate predictions for the energy levels. That contribution was estimated by calculating the largest unknown term in the expansion of the electron self-energy in powers of $Z\alpha$. It was then argued that, based on the general trends of calculated terms in the series, the value of the unknown term A_{60} should be sufficient to provide an accurate result, and the value of this coefficient for various states relevant to the experiments of interest was calculated. Here we revisit the theory of the energy levels and give results of an all-orders calculation that confirms the expectation that the A_{60} term provides an accurate result.

2. Theory

In principle, it is known that the $Z\alpha$ expansion converges very well for Rydberg states. However, if one of the most important physical constants is to be determined on the basis of spectroscopic measurements—the Rydberg constant—then one would like to have very reliable predictions. The recent proposal by Jentschura *et al* (2008) considered, in particular, precise measurements for transitions among Rydberg states of hydrogen-like ions with medium charge numbers $Z = 10, \dots, 20$ and in the range of principal quantum numbers $n = 10, \dots, 20$, with unit change in the principal quantum number n . These transitions, according to figure 1 of Jentschura *et al* (2008), are in the optical (THz) domain.

In Jentschura *et al* (2008), the self-energy remainder function is represented as

$$G(Z\alpha) = A_{60} + A_{81}(Z\alpha)^2 \ln(Z\alpha)^{-2} + A_{80}(Z\alpha)^2 + \dots + \frac{\alpha}{\pi} B_{60} + \dots + \left(\frac{\alpha}{\pi}\right)^2 C_{60} + \dots \quad (1)$$

where the A , B , and C coefficients refer to one-loop, two-loop and three-loop effects, respectively. This expansion is valid for states with angular momentum $l \geq 2$. The main contribution to G comes from the one-loop self-energy, and indeed, for states with angular momentum $l \geq 3$, the coefficients A_{81} and A_{80} are determined exclusively by the self-energy because the vacuum polarization contribution to these coefficients vanishes. For even higher angular momenta ($l \geq 10$), we can estimate vacuum polarization effects to be entirely negligible at a relative accuracy level of 10^{-15} for the transitions under consideration. We therefore define the one-loop self-energy (SE) remainder function as

$$G_{\text{SE}}(Z\alpha) = A_{60} + A_{81}(Z\alpha)^2 \ln(Z\alpha)^{-2} + A_{80}(Z\alpha)^2 + \dots \quad (2)$$

where the coefficients are understood to be entirely due to self-energy effects. In Jentschura *et al* (2008), the following approximation was made:

$$G_{\text{SE}}(Z\alpha) \approx A_{60}, \quad (3)$$

Table 1. Values of A_{60} used by Jentschura *et al* (2008) for states with principal quantum numbers $n = 13, 14, 15$.

n	κ	A_{60}
13	11	0.000 006 795 75(5)
13	−12	0.000 043 189 98(5)
13	12	0.000 004 699 73(5)
13	−13	0.000 027 294 75(5)
14	12	0.000 004 108 25(5)
14	−13	0.000 029 799 37(5)
14	13	0.000 002 966 41(5)
14	−14	0.000 019 452 79(5)
15	13	0.000 002 521 08(5)
15	−14	0.000 021 160 50(5)
15	14	0.000 001 893 09(5)
15	−15	0.000 014 206 31(5)

and A_{60} was calculated using a semi-analytic method. The A_{60} coefficient depends only on the principal quantum number n of the state under investigation, and on the Dirac angular momentum quantum number κ . Because the A_{81} and A_{80} coefficients can be expected to be numerically small (this is in line with the trend exhibited by lower order coefficients as a function of n and κ), the approximation in equation (3) can be used with good effect even for the range $10 \leq Z \leq 20$, but on the other hand, it may appear questionable that all the Z -dependent terms from equation (2) are ignored in formulating the approximation (3). Values of A_{60} used by Jentschura *et al* (2008) are summarized in table 1, for states with principal quantum numbers $n = 13, 14, 15$.

Recently, we have carried out a fully nonperturbative calculation of QED self-energy shifts for Rydberg states. This calculation incorporates the entire Z -dependence of the right-hand side of (3) and takes into account all higher order (in $Z\alpha$) effects. The basic method of calculation is described by Jentschura *et al* (2001), but significant refinements have been made for the case at hand. Here, we discuss nonperturbative results in the range $n = 13, 14, 15$ of principal quantum numbers. Special emphasis will be placed on the state with $n = 13, \kappa = 12$ and nuclear charge numbers $Z = 14$ and $Z = 16$ (see also equations (4)–(6) below). In order to appreciate the difficulty associated with this nonperturbative calculation, we must recall a few facts about the bound-electron self-energy. In general, the radiative energy shift is complex quantity whose imaginary part corresponds to the one-photon decay width of the reference state. Here, we are interested only in the real part of the energy shift, which is the relevant physical quantity for spectroscopic measurements, but this means that the pole terms which are otherwise responsible for the nonvanishing imaginary part, have to be subtracted.

In the fully relativistic formalism, there are no selection rules that would allow only dipole transitions to occur, and all possible electric as well as magnetic multipoles have to be taken into account. If the reference state were the 2S state, then, e.g. the integrand of the 2S self-energy would have a pole due to an one-photon M1 transition to the ground state. This would be the only pole to subtract for this calculation. For the state with quantum numbers $n = 13, \kappa = 12$, a total of 166 lower lying bound-state poles have to be subtracted. For

the highest state considered here, which is the state with $n = 15$, $\kappa = -14$, this number increases to 224. The calculation of the pole terms proceeds by a highly accurate integration of angular variables, with a Green function that is restricted to the virtual state in question. This calculation yields the residue at the bound-state energy and is subtracted before the final integration over the virtual photon energy, with the full bound-electron Green function that corresponds to a sum over all possible virtual bound states. Extreme care must be taken in this subtraction operation, and both the pole terms as well as the Green function term have to be calculated to sufficient accuracy in order to retain numerical significance in the results. These subtractions affect the so-called low-energy part of the calculation, where the virtual photon energy is of the same order-of-magnitude as a virtual atomic transition.

There is a further ‘trap’ to be avoided in the low-energy part. Normally, for lower lying states, one observes the apparent convergence of terms in the Green function as more and more partial waves are added and terminates the summation once apparent convergence to a specified accuracy is reached. This method would have disastrous consequences for Rydberg states. The reason is that for low-energy virtual photons, dipole transitions from the reference state give rise to the numerically dominant contribution to the self-energy integrand. These correspond to virtual states with $|\kappa|$ being displaced from the reference state κ Dirac quantum number by no more than two. Therefore, the convergence criterion has to be modified, and the summation over angular momenta of the virtual states cannot be terminated before the reference state κ is reached.

Further numerical problems plague the highly excited Rydberg states in the so-called high-energy part, where virtual photons of the order of the rest mass energy of the electron are considered. In this region, it is customary to expand the self-energy in powers of Coulomb interactions, i.e. in the number of Coulomb photon exchanges of the electron with the atomic nucleus. The so-called mass renormalization is associated with the zero-Coulomb-vertex term, and the vertex renormalization is associated with the one-Coulomb-vertex diagram. It is especially problematic to evaluate the subtraction terms associated with the zero-Coulomb-vertex and the one-Coulomb-vertex diagrams for the Rydberg states. When these calculations are performed in momentum space, numerical cancellations occur because the Rydberg states are weakly bound as compared to lower lying states, and slow convergence of the momentum space integrals is observed. Surprisingly, there are also quite severe numerical cancellations in the evaluation of the fully relativistic momentum space wavefunctions themselves. Although their well-known expressions entail only a finite number of terms and although these can be calculated recursively (Mohr and Kim 1992), there are numerical cancellations among the terms which cannot easily be avoided, and enhanced precision arithmetic has to be used. In the evaluation of the high-energy remainder terms, the weak binding of the Rydberg electron implies that many partial waves have to be summed before convergence is reached. In a typical case, one has to sum about one to two million κ for the ground-state electron self-energy

in atomic hydrogen, and much less for higher nuclear charge numbers. Here, an enhanced accuracy relativistic Green function is used, and about five million κ are summed before convergence is reached (see also Jentschura *et al* (1999)).

We therefore use octuple (roughly 64 digit) precision for the fully relativistic Green function of a hydrogen-like bound system in order to describe the virtual excitations of the hydrogen-like ion. The implementation of octuple-precision arithmetic is based on a separation of the eightfold-precision number into double-precision parts (Korobov 2005). The enhanced accuracy is used in order to overcome the convergence problems in angular momentum expansions and the severe numerical losses due to renormalization for weakly bound Rydberg states. The calculations were carried out on a cluster of IBM POWER5 64 bit workstations at Missouri University of Science and Technology³.

After the subtraction of the lower-order analytic terms, including the Bethe logarithm (Jentschura and Mohr 2005),

$$\ln k_0(n = 13, l = 12) = -0.315\,815\,189(1) \times 10^{-4}, \quad (4)$$

we obtain

$$G_{SE}(n = 13, \kappa = 12, Z = 14) = 4.69(9) \times 10^{-6}, \quad (5a)$$

$$G_{SE}(n = 13, \kappa = 12, Z = 16) = 4.68(7) \times 10^{-6}. \quad (5b)$$

These nonperturbative results have to be compared to the known analytic result for the A_{60} coefficient (Jentschura *et al* 2008), which is equivalent to the self-energy remainder at vanishing nuclear charge $Z = 0$:

$$A_{60} = G_{SE}(n = 13, \kappa = 12, Z = 0) = 4.699\,73(5) \times 10^{-6}. \quad (6)$$

Within the numerical uncertainty, the results in equation (5a) are equal to those in equation (6), and therefore, the approximation indicated by equation (3) in estimating the higher order remainder function for the Rydberg states can be justified *a posteriori*. We re-emphasize that the results for G_{SE} from equation (5a) contain the entire higher-order nonperturbative remainder beyond A_{60} , and thus the entire series starting with A_{60} , A_{81} , *etc.* An analogous pattern can be observed for other transitions (see table 2). Within the quoted numerical uncertainty of the fully nonperturbative results for the remainder function, which we estimated based on the apparent convergence of the multi-dimensional integrals, the nonperturbative result for G_{SE} is equal to the perturbative result for A_{60} . This highlights the internal consistency of the theoretical approach to the transitions in question. Both the analytic as well as the numerical calculations are highly nontrivial, and mistakes therefore cannot be excluded. However, the consistency of two completely independent approaches is reassuring.

To summarize, we conclude that (a) the higher-order remainder beyond A_{61} is described to excellent accuracy by

³ Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by the Missouri University of Science and Technology nor by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Table 2. Values of the nonperturbative self-energy remainder function $G_{SE}(Z\alpha)$ for near-circular Rydberg states with principal quantum numbers $n = 13, 14, 15$, and for nuclear charge numbers $Z = 14, 16$.

n	κ	Z	$G_{SE}(Z\alpha)$	n	κ	Z	$G_{SE}(Z\alpha)$
13	11	14	0.000 006 76(9)	13	11	16	0.000 006 82(4)
13	-12	14	0.000 043 17(5)	13	-12	16	0.000 043 21(2)
13	12	14	0.000 004 69(9)	13	12	16	0.000 004 68(7)
13	-13	14	0.000 027 28(9)	13	-13	16	0.000 027 28(5)
14	12	14	0.000 004 03(8)	14	12	16	0.000 004 08(6)
14	-13	14	0.000 029 74(5)	14	-13	16	0.000 029 78(3)
14	13	14	0.000 002 96(9)	14	13	16	0.000 002 96(9)
14	-14	14	0.000 019 44(9)	14	-14	16	0.000 019 45(9)
15	13	14	0.000 002 43(9)	15	13	16	0.000 002 49(3)
15	-14	14	0.000 021 07(9)	15	-14	16	0.000 021 14(2)
15	14	14	0.000 001 84(7)	15	14	16	0.000 001 91(9)
15	-15	14	0.000 014 15(7)	15	-15	16	0.000 014 23(9)

the A_{60} coefficient, even at medium nuclear charge numbers $Z \geq 10$, and (b) the analytic coefficients multiplying the higher order corrections beyond A_{60} are small, being consistent with a general trend observed qualitatively by Jentschura *et al* (2003) and quantified by Jentschura *et al* (2008). We also observe that (c) there is mutual consistency of the numerical and analytic approaches, which enhances the reliability of the theoretical predictions.

3. Experimental considerations

Rydberg states of hydrogen-like ions with $l \geq 2$ essentially avoid a number of problems associated with either higher order binding corrections to QED interactions or the nuclear size effect in Lamb shift predictions. Circular states have other features which may be useful in experiments: they have the longest lifetime in a given shell n and a suppressed Stark effect. In the cases being considered here, the higher-order QED binding corrections for Rydberg states are smaller by a factor of about 10^7 compared to S states, providing significant advantages from a theoretical point of view. Such simplifications come with experimental trade-offs associated with a large spontaneous emission rate, for example. Natural decay linewidths tend to be small for states from which electric dipole (E1) decay is forbidden (as in the case of the 2S level). In contrast, the spontaneous decay rate for a circular Rydberg state is dominated by an electric dipole E1 transition from the highest- l value of the state n to the highest- l value of the state $n - 1$. The nonrelativistic expression for this decay rate has been examined in Bethe and Salpeter (1957) and also by Jentschura *et al* (2005) as the nonrelativistic limit of the imaginary part of the level shift. It is possible that this decay process can introduce asymmetries into the line shape for the transition frequencies. Fortunately, these effects are small and of the order $\alpha(Z\alpha)^2 E_{QED}$ as has been shown by Low (1952). These corrections, should they become necessary, can be calculated and determined for the particular systems chosen for the experiments, taking into account details of the experimental set-up. Precision experiments with one-electron ions in Rydberg states require considerable effort to develop. It is encouraging to note that circular Rydberg states of hydrogen have been studied with high precision in

atomic beams for transitions in the millimeter region; as a result, a determination of the Rydberg constant with a relative uncertainty of 2.1×10^{-11} has been made (De Vries 2001). The advent of optical frequency combs (Hänsch 2006) has opened the possibility of making precise measurements of optical transitions between Rydberg states in one-electron ions, with potential for higher precision (Ma *et al* 2004) and broader range of applications (Marian *et al* 2004). Concurrently, an array of tools and techniques have emerged for realizing ‘engineered atoms’ that are built in traps and tailored into states of interest. Cooling techniques (Itano 1995) developed for low- Z ions, for instance, can be extended to high- Z ions extracted from sources like an electron beam ion source/trap (EBIS/T) (see the review by Donets (1998)). Some such synthetic atoms or artificial quantum systems—for example, antihydrogen (Gabrielse 2005) or a single electron in a Penning trap (Brown and Gabrielse 1986)—have the potential to extend the range of precision measurements that probe nature.

Experiments with cold hydrogen-like ions in high- l Rydberg states may be possible for a wide range of nuclear charge Z and angular momentum l and would be useful for consistency checks and optimization. An effort is underway at NIST to make hydrogen-like ions starting from bare nuclei, with focus on low- Z to mid- Z ions ($1 < Z < 11$). Ion traps, such as a compact Penning trap (Brewer and Tan 2009), are being developed for experiments to utilize bare nuclei extracted from the NIST EBIT in its current configuration (see Ratliff and Roberts 2001, Pikin *et al* 1996, Gillaspay *et al* 2002). Low- Z hydrogen-like ions, with their narrower line widths for optical transitions among Rydberg states, seem the most favourable for frequency comb-based measurements that could lead to better determinations of fundamental constants—the Rydberg constant in particular. On the other hand, perturbations in Rydberg states (e.g. Stark mixing) tend to be significantly attenuated in heavier hydrogen-like ions because a higher nuclear charge Z produces a larger fine-structure separation, which scales as Z^4 . This wide range of available (n, Z) combinations could be useful for extending diversity of experiments used to determine fundamental constants and test theory. Apart from this long-term goal, experimental techniques developed in this effort may directly enhance research in other areas, such as laboratory astrophysics, plasma

diagnostics, new regimes/techniques in atomic spectroscopy and standards development.

4. Conclusion

In this paper (section 2), we report on a nonperturbative (in $Z\alpha$) calculation of the self-energy remainder function for highly excited, nearly circular Rydberg states in hydrogen-like ions with charge numbers $Z = 14$ and $Z = 16$ (see table 2). These numerical values eliminate the uncertainty due to uncalculated higher-order terms in the expansion of the bound-electron self energy in $Z\alpha$, where the growth of the number of terms in this expansion inhibits analytic calculations. The results of the nonperturbative calculations of the self-energy for circular Rydberg states reported here show that the approximation made by Jentschura *et al* (2008) in using the A_{60} term in the perturbation expansion to evaluate the energy levels is fully justified. The situation is even better: namely the values for the nonperturbative self-energy remainder listed in table 2 agree with the perturbative A_{60} coefficients listed in table 1 on the level of at least 10^{-7} for A_{60} , even though the coupling constant $Z\alpha$ is no longer negligible at the indicated values of the nuclear charge number (we recall that it is the absolute, not the relative, accuracy of the self-energy remainder function that determines the predictive limits of the calculations). This confirms the expectation that the remainder terms drastically decrease for Rydberg states in comparison to lower lying ionic states. We also confirm that accurate evaluations of the energy levels of these states can be made, with the potential to provide an independent value for the Rydberg constant and an additional test of theory.

The aforementioned agreement provides additional evidence for the Rydberg states being extremely ‘favourable’ states with regard to the theoretical analysis. Not only does the self-energy function converge much more rapidly than for lower lying states (see section 2 and Jentschura *et al* (2008)), but also, other problematic effects (such as the nuclear-size correction to the energy levels) are so vanishingly small for these states so as to be entirely negligible at current and projected levels of accuracy. In particular, the nuclear-size correction does not need to be taken into account in the prospective determination of the Rydberg constant from measurements which are currently being planned (see section 3).

We conclude with a few more general remarks. On the one hand, the electron in a Rydberg state is bound sufficiently loosely to avoid complications due to the shape of the nuclear charge distribution and other problematic issues. On the other hand, it is bound sufficiently strongly so that its energy levels provide additional insight into the nature of one of the most precisely understood interactions in nature and allow one to derive the Rydberg constant from a precise measurement using ionic Rydberg states, which are appropriately named for this purpose. Advances both in experimental techniques (see

section 3) as well as in the theoretical analysis (section 2) are indispensable along the way.

Acknowledgments

UDJ has been supported by the National Science Foundation (grant PHY-8555454) as well as by a Precision Measurement grant from the National Institute of Standards and Technology.

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