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Rydberg states of helium: Relativistic and second-order corrections

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In this paper the corrections of relative order $(Z\alpha)^2$ to all multipole polarizabilities of a hydrogenic ion of nuclear charge Z are computed, with use of third-order perturbation theory with nonrelativistic wave functions. To accomplish this, the spin-independent part of the Pauli Hamiltonian of order α^2 is treated as a perturbation along with the usual Coulomb interaction expanded in a multipole series. The leading (dipole) correction is in agreement with the previous results of Zon *et al.* In addition, the previously discussed second-order energy shift has now been calculated analytically. Both of these corrections are applied to the high-lying Rydberg states of helium, yielding small but important corrections to the fine-structure splittings.

I. INTRODUCTION

The optical-potential method for calculating perturbations of highly excited state of two-electron atoms was first applied in the adiabatic approximation by Bethe,[1] who assumed that the outer electron (NL) could be considered at rest while the distortion of the inner $(1s)$ electron was being evaluated. This picture has been considerably developed in recent years,[2–4] especially in Ref. 4 which uses the Feshbach operator technique[5] to systematize the perturbation bookkeeping. With care even quite small effects like the nuclear-recoil (mass-polarization) corrections of order m_e/M_{nuc} can be derived analytically, at least for those large values of N and L for which only the long-range parts of the potential are significant.

At the same time there has been a large increase in the precision of fine-structure measurements in excited states of helium.[6,7] In Ref. 7, for example, the fine-structure splittings in the $N=10$ manifold have been measured to within about 10 kHz. Precision of this magnitude is potentially capable of observing retardation corrections[8,9] to the fine structure, but only if all the "ordinary" effects of comparable size have been properly accounted for. For H-I transitions the asymptotic expansion of the potential[4] accounts for an estimated error of about 10 kHz, and more sophisticated methods are needed, but for I-J transitions and higher the error is not more than about 1 kHz. It is thus necessary that all corrections of this order of magnitude be reliably calculated, in order to reveal the interesting retardation effects. In this paper two types of corrections will be discussed: those due to the relativistic modification of the polarizability of the $1s$ He$^+$ core and those coming from the second-order perturbation of the Rydberg electron. The results will be presented in tabular form and compared with the best experimental data.[7]

II. RELATIVISTIC POLARIZABILITY CORRECTIONS

The effect of relativity on the value of the dipole polarizability of systems in the hydrogen isoelectronic series was first investigated by Zon *et al.*[10] They used a certain representation of the relativistic Green's function and were able to write the corrections to the ordinary polarizability as a power series in $(Z\alpha)^2$, retaining the first two terms. In this section I will outline a new derivation, capable of obtaining these corrections for all multipoles correct to order $(Z\alpha)^2$, which is quite adequate for the present application. The derivation uses the optical-potential formalism of Ref. 4, applied to the case of the helium isoelectronic series, but includes in the perturbing potential the relativistic Hamiltonian of order α^2:

$$V=V_e+V_{\text{MP}}+V_{\text{rel}}\ , \tag{1}$$

where V_e is the usual multipole expansion of the electrostatic interaction of the two electrons $(r<x)$, the second term is the nuclear-recoil [mass-polarization (MP)] term, and V_{rel} is the spin-independent part of the Breit Hamiltonian in the Pauli approximation.[11] (I have omitted certain terms involving correlation between the two electrons from the relativistic part, and these should be investigated later; they are not involved in the polarizability correction.) The recoil term $V_{\text{MP}}=-K\nabla_{\mathbf{r}}\cdot\nabla_{\mathbf{x}}$ was examined previously;[4,12] its leading effect is to increase the effective dipole polarizability by the factor $(1+K)$, but it also can produce other corrections. The relativistic term involving x comes from the variation of mass with velocity of the outer electron and gives a sizable correction in first order; it has also been discussed previously.[13] Thus, for the present calculation it is sufficient to retain only the following terms[11] in the perturbation in reduced Rydberg units

$$[\mathscr{R}=M_{\text{nuc}}\mathscr{R}_\infty/(m_e+M_{\text{nuc}})$$

$$=3.289\,391\,08\times10^9\ \text{MHz (for }^4\text{He)}]\ :$$

$$V\simeq V_e+V_{\text{rel}}(\mathbf{r})$$

$$=\frac{2}{x}\sum_{l=1}^{\infty}\left[\frac{r}{x}\right]^l P_l(\hat{\mathbf{x}}\cdot\hat{\mathbf{r}})+\alpha^2[\pi Z\delta(\mathbf{r})-\tfrac{1}{4}\nabla_r^4]\ . \tag{1'}$$

In order to isolate just those terms of second order in V_e and first order in $V_{\rm rel}$ which contribute to the polarizability correction, all third-order terms in the expansion of the optical potential must be examined. Only the adiabatic type of term is considered, since the polarizability is an effect of the static field of the outer electron acting on the inner one. (In the notation of Ref. 4 this means $j=1$.) The third-order optical potential is

$$\bar{U}_{\rm rel}(x)=\sum_{n,m}{}' \frac{\langle 1s \mid V \mid n\rangle\langle n \mid V \mid m\rangle\langle m \mid V \mid 1s\rangle}{d_n d_m} -\langle 1s \mid V \mid 1s\rangle \sum_{n}{}' \frac{\langle 1s \mid V \mid n\rangle\langle n \mid V \mid 1s\rangle}{d_n^2}\ , \qquad (2)$$

where the summations run over all states of the core except $1s$. Inserting the two different terms in V explicitly, and noting that $\langle 1s \mid V_e \mid 1s\rangle=0$, we can write

$$\bar{U}_{\rm rel}(x)=\sum_{n,m}{}'(\langle 1s \mid V_e \mid n\rangle\langle n \mid V_{\rm rel} \mid m\rangle\langle m \mid V_e \mid 1s\rangle+2\langle 1s \mid V_{\rm rel} \mid n\rangle\langle n \mid V_e \mid m\rangle\langle m \mid V_e \mid 1s\rangle)\frac{1}{d_n d_m} - \sum_{n}{}'\langle 1s \mid V_{\rm rel} \mid 1s\rangle\langle 1s \mid V_e \mid n\rangle\langle n \mid V_e \mid 1s\rangle\frac{1}{d_n^2}\ . \qquad (3)$$

The method of Dalgarno and Lewis[14] can be used to carry out the sums in Eq. (3). In the first step of this procedure the following equation must be solved for the function G:

$$V_e \mid 1s\rangle=[H_0(\mathbf{r}),G] \mid 1s\rangle\ , \qquad (4)$$

where

$$H_0(\mathbf{r})=-\nabla_r^2-\frac{2Z}{r}\ .$$

Then, since the energy denominator $d_n=E_{1s}-E_n$ the matrix elements of the potential appearing in Eq. (3) can be written as

$$\langle n \mid V_e \mid 1s\rangle=-d_n\langle n \mid G \mid 1s\rangle\ , \qquad (5)$$

and Eq. (3) can be partially summed:

$$\bar{U}_{\rm rel}(x)=\langle 1s \mid GV_{\rm rel}G \mid 1s\rangle -\sum_{n}{}'\frac{2}{d_n}\langle 1s \mid V_{\rm rel} \mid n\rangle\langle n \mid V_e G \mid 1s\rangle -\langle 1s \mid V_{\rm rel} \mid 1s\rangle\langle 1s \mid G^2 \mid 1s\rangle\ . \qquad (6)$$

It is easy to show that the solution of Eq. (4) is

$$G(\mathbf{x},\mathbf{r})=\frac{1}{Z^2x}\sum_{l=1}^{\infty}\left[\frac{r}{x}\right]^l\left[\frac{1}{l}+\frac{Zr}{l+1}\right]P_l(\hat{\mathbf{x}}\cdot\hat{\mathbf{r}})\ , \qquad (7)$$

and I have used the fact that $\langle 1s \mid G \mid 1s\rangle=0$ to simplify Eq. (6) slightly.

The first and last terms in Eq. (6) can now be explicitly evaluated; one should notice that the δ-function part of $V_{\rm rel}$ does not contribute to the first term because G vanishes at $r=0$ [Eq. (7)]. As emphasized in Ref. 11, it is not correct to allow ∇^4 to operate to the right in these matrix elements, but it should always be replaced by ∇^2 operating to the left and right. With this in mind one finds that $\langle 1s \mid V_{\rm rel} \mid 1s\rangle=-\alpha^2Z^4/4$, and the sum of these two terms [the first and the third of Eq. (6)] is

$$\langle 1s \mid GV_{\rm rel}G \mid 1s\rangle-\langle 1s \mid V_{\rm rel} \mid 1s\rangle\langle 1s \mid G^2 \mid 1s\rangle=\frac{4(Z\alpha)^2[(2l-1)!](4l^4+20l^3+17l^2-20l-14)}{(2Zx)^{2l+2}l(l+1)(2l+1)}\ . \qquad (8)$$

The remaining term, which still contains a sum over n, is the most difficult to evaluate. It would be simple if one could use the Dalgarno and Lewis technique based on the operator $V_{\rm rel}$, and this is indeed possible for the δ-function part.[15] Because of the special meaning of the ∇^4 operator mentioned previously, it does not seem to be possible to treat it in the usual way. Instead the rather complicated expression V_eG, which is a product of two sums, must be used.

This is somewhat simplified by the fact that the sum over n involves only s states, since $V_{\rm rel}$ is rotationally invariant. Hence only the P_0 part of the product $P_lP_{l'}$ contributes to the angular integral, and that product can be replaced by the expression $\delta_{ll'}/(2l+1)$. Then one must solve the equation

$$[H_0,F] \mid 1s\rangle=[V_eG-\langle 1s \mid V_eG \mid 1s\rangle] \mid 1s\rangle \qquad (9)$$

for the function F which takes the following form:

$$y_l''(u)+2\left[\frac{1}{u}-1\right]y_l'(u)+\frac{u^{2l}}{l} +\frac{u^{2l+1}}{l+1}-\frac{[(2l+1)!](2l+1)(l+2)}{l2^{2l+1}}=0\ , \qquad (10)$$

where $u=Zr$, and $F=\sum y_l/Z^{2l+4}$. Equation (10) can be solved by the use of an integrating factor, and the solution is

$$y_l(u)=\frac{u^{2l+2}}{4(l+1)^2} +\frac{(l+2)(2l+1)[(2l+1)!]}{2^{2l+1}l}\sum_{k=2}^{2l+1}\frac{(2u)^k}{k(k+1)!}-K_l\ , \qquad (11)$$

where

$$K_l=\frac{(l+2)[(2l+1)!]}{2^{2l+3}} \times\left[\frac{4l^2+8l+5}{l+1}+\frac{2(2l+1)}{l}\sum_{k=2}^{2l+1}\frac{1}{k}\right]\ .$$

(The constant K_l is added to make the integral $\langle 1s \mid F \mid 1s\rangle=0$.) Then the second term in Eq. (6) is

$$2\langle 1s \,|\, V_{\mathrm{rel}}F \,|\, 1s\rangle=\frac{(Z\alpha)^2[(2l)!]}{2^{2l}(Zx)^{2l+2}}\left[\left[\frac{2l+3}{2l+2}\right]+\frac{(l+2)(2l+1)}{l}\sum_{k=2}^{2l+1}\frac{1}{k}\right] . \tag{12}$$

Recalling that the ordinary nonrelativistic polarizability is[14]

$$\alpha_l=\frac{[(2l-1)!](2l+1)(l+2)}{2^{2l-1}Z^{2l+2}} , \tag{13}$$

one can write the corrected relativistic polarization potential as

$$\overline{U}_l^{\mathrm{rel}}=-\frac{\alpha_l}{x^{2l+2}}[1-(Z\alpha)^2 g_l] ,$$

where (14)

$$g_l=\sum_{k=2}^{2l+1}\frac{1}{k}+\frac{(4l^4+14l^3+10l^2-10l-7)}{l(l+1)(l+2)(2l+1)^2} .$$

For the dipole case, $l=1$, this correction is $g_1=\frac{28}{27}$ in agreement with the previous results,[10] and for the quadrupole case $g_2=\frac{959}{600}$, a new result and one that may be significant. The higher terms are interesting at present only for mathematical completeness.

III. SECOND-ORDER CORRECTIONS

In order to keep terms in the optical potential up to order x^{-8}, it is necessary for consistency also to include the second-order effect of the x^{-4} potential. The problem was discussed briefly in Ref. 4, to which the reader is referred. The important point is that, when one is interested in corrections a few kHz as I am here, it is essential to include these second-order corrections. I will discuss the method used to calculate them in some detail.

The second-order energy shift of the hydrogenic level with quantum numbers (N,L) due to the x^{-4} dipole polarization potential is

$$\Delta_2^{N,L}=\alpha_1\int d^3x\,\Psi_{NL}^{(1)}W_{NL}\Psi_{NL}^{(0)} ,$$

where

$$W_{NL}(x)=\left[F_{NL}-\frac{1}{x^4}\right] , \tag{15}$$

the superscripts (0) and (1) denote unperturbed and perturbed wave function of the Rydberg electron, and F_{NL} is the unperturbed expectation value[16] of $1/x^4$. In the usual way, the first-order perturbed function satisfies the inhomogeneous differential equation

$$\left[\nabla^2+\frac{2}{x}-\frac{1}{N^2}\right]\Psi_{NL}^{(1)}(\mathbf{x})=\alpha_1 W_{NL}(x)\Psi_{NL}^{(0)}(\mathbf{x}) . \tag{16}$$

Letting

$$\Psi_{NL}^{(1)}(\mathbf{x})=f_{NL}(x)\Psi_{NL}^{(0)}(\mathbf{x})$$

and

$$\Psi_{NL}^{(0)}(\mathbf{x})=u_{NL}(x)Y_{NL}(\hat{\mathbf{x}})/x ,$$

one obtains the following differential equation for $f(x)$:

$$\frac{d^2f}{dx^2}+\frac{d}{dx}(\ln u^2)\frac{df}{dx}=\alpha_1 W(x) . \tag{17}$$

This can be easily integrated once

$$\frac{df}{dx}=\frac{\alpha_1}{u^2(x)}\int dx\,u^2(x)W(x) , \tag{18}$$

and with careful handling of the zeroes in u^2, f itself can be found. Finally, the expression for the second-order shift is

$$\Delta_2^{N,L}=\alpha_1\int_0^\infty dx\,f_{NL}(x)u_{NL}^2(x)W(x) . \tag{19}$$

I previously[4] evaluated f and Δ for the special case $(N=L+1)$ for which u has no nodes except at $x=0$ and obtained the result

$$\Delta_2^{N,N-1}=-\frac{4(\alpha_1)^2(128N^4-560N^3+848N^2-518N+105)}{N^8[(2N-1)(N-1)(2N-3)]^3(2N-5)} . \tag{20}$$

With the aid of the symbolic manipulation program MACSYMA,[17] I have now also solved for the one-node case $(N=L+2)$ and obtained the result

TABLE I. Second-order energy shifts $(-\Delta_2)$ for the ^{4}He in MHz. All entries were obtained analytically as discussed in the text.

$N\backslash L$	3	4	5	6	7	8	9
4	39.002 24						
5	26.265 20	0.956 18					
6	17.143 33	0.721 23	0.052 88				
7	11.510 55	0.517 15	0.042 84	4.868×10^{-3}			
8	8.010 65	0.373 14	0.032 82	4.137×10^{-3}	6.382×10^{-4}		
9	5.764 78	0.274 52	0.024 99	3.326×10^{-3}	5.609×10^{-4}	1.086×10^{-4}	
10	4.271 95	0.206 40	0.019 21	2.640×10^{-3}	4.676×10^{-4}	9.781×10^{-5}	2.259×10^{-5}
11	3.246 60	0.158 44	0.014 97	2.102×10^{-3}	3.836×10^{-4}	8.386×10^{-5}	2.072×10^{-5}

TABLE II. Theoretical level splittings for ^{4}He, including all corrections except retardation, in MHz. The second column is the total uncorrected splitting from Ref. 4, and the fourth and fifth columns include $l=1$ only. The results of this paper are in the fourth and sixth columns. When possible, the totals are compared with experiment (Ref. 7).

NL-NL'	$V_4+V_6+\cdots$	∇_x^4	Relativistic polarizability	Mass polarization	Δ_2	Theory sum	Experiment	Experiment − Theory
6G-6H	2043.067	32.752	−0.455	0.565	0.668	2076.60±1.19		
7G-7H	1337.876	20.625	−0.298	0.370	0.474	1359.05±1.04	1359.16±0.11	0.11±1.04
8G-8H	918.566	13.817	−0.205	0.254	0.340	932.77±0.84	931.34±0.44	−1.43±0.95
9G-9H	655.871	9.704	−0.146	0.182	0.250	665.86±0.66		
10G-10H	483.727	7.074	−0.108	0.134	0.187	491.01±0.52	490.990±0.010	−0.02±0.52
11G-11H	366.542	5.315	−0.082	0.102	0.143	372.02±0.41		
7H-7I	409.167	14.279	−0.091	0.113	0.038	423.506±0.008		
8H-8I	284.695	9.566	−0.063	0.078	0.029	294.304±0.011		
9H-9I	205.046	6.718	−0.045	0.056	0.022	211.797±0.011		
10H-10I	152.135	4.898	−0.034	0.042	0.017	157.057±0.010	157.068±0.013	0.011±0.016
11H-11I	115.778	3.680	−0.026	0.032	0.013	119.477±0.008		
8I-8K	105.1222	7.0149	−0.0233	0.0289	0.0035	112.1462±0.0011		
9I-9K	76.5489	4.9268	−0.0169	0.0210	0.0028	81.4826±0.0006		
10I-10K	57.2214	3.5916	−0.0127	0.0157	0.0022	60.8183±0.0004	60.818±0.010	−0.000±0.010
11I-11K	43.7790	2.6985	−0.0097	0.0120	0.0017	46.4815±0.0002		
9K-9L	32.3790	3.7676	−0.0072	0.0089	4.5×10^{-4}	36.1487±0.0002		
10K-10L	24.4277	2.7465	−0.0054	0.0067	3.7×10^{-4}	27.1759±0.0002		
11K-11L	18.8105	2.0635	−0.0042	0.0052	3.0×10^{-4}	20.8754±0.0001		
10L-10M	11.450 79	2.168 33	-2.53×10^{-3}	3.14×10^{-3}	7.5×10^{-5}	13.6198±0.000 05		
11L-11M	8.886 97	1.629 10	-1.96×10^{-3}	2.44×10^{-3}	6.3×10^{-5}	10.5166±0.000 04		

TABLE III. Inclusion of the theoretical values of the retardation correction (Ref. 9) and comparison of the final result with experiment when available. Items in parentheses have been estimated by extrapolation, as described in the text.

NL-*NL*′	Retardation	Total theory	Experiment-theory
6*G*-6*H*	(−0.177)	(2076.42±1.19)	
7*G*-7*H*	(−0.116)	(1358.93±1.04)	(0.23±1.04)
8*G*-8*H*	(−0.079)	(932.69±0.84)	(−1.35±0.95)
9*G*-9*H*	−0.0563	665.80±0.66	
10*G*-10*H*	−0.0416	490.97±0.52	0.02±0.52
11*G*-11*H*	−0.0315	371.99±0.41	
7*H*-7*I*	(−0.034)	(423.472±0.008)	
8*H*-8*I*	(−0.023)	(294.281±0.011)	
9*H*-9*I*	−0.0168	211.780±0.011	
10*H*-10*I*	−0.0125	157.045±0.010	0.023±0.016
11*H*-11*I*	−0.0095	119.468±0.008	
8*I*-8*K*	(−0.008)	(112.138±0.001)	
9*I*-9*K*	−0.0059	81.4767±0.0006	
10*I*-10*K*	−0.0045	60.8138±0.0004	0.004±0.010
11*I*-11*K*	−0.0034	46.4781±0.0002	
9*K*-9*L*	−0.0024	36.1463±0.0002	
10*K*-10*L*	−0.0018	27.1741±0.0002	
11*K*-11*L*	−0.0014	20.8740±0.0001	
10*L*-10*M*	−0.00079	13.61902±0.00005	
11*L*-11*M*	−0.0006	10.5160±0.00004	

$$\Delta_2^{N,N-2} = -\frac{4(\alpha_1)^2(128N^7-400N^6-2320N^5+12\,666N^4-19\,133N^3+4846N^2+10\,228N-5880)}{N^8[(N-2)(N-1)(2N-3)(2N-5)]^3(2N-7)} \,. \tag{21}$$

I have not been able to derive an expression for general L and N, but can obtain an exact, analytical result for any particular case. In this way I have constructed Table I, which gives Δ_2 for a number of cases, including those of present experimental interest. [Note that the second-order shift scales with increasing nuclear charge as $(Z-1)^6(2/Z)^8\Delta_2^{\mathrm{He}}$.]

IV. COMPARISON WITH EXPERIMENT AND CONCLUSIONS

All the corrections discussed previously along with the terms discussed in Ref. 4 are shown in Table II for a large set of level splittings ($\Delta L=1, \Delta N=0$) that are of present or potential experimental interest. Whenever possible, these are compared with experimental values, and the agreement is seen to be very good.

In Table III the retardation correction recently derived by Au, Feinberg, and Sucher[9] is tabulated, added to the previous theoretical values, and again compared to the experimental results. (In those cases where no numerical values have been given in Ref. 9 a simple extrapolation has been employed. It is based on the fact that the retardation correction for each L is closely proportional to the relativistic correction to the dipole polarizability. In turn this is due to the fact[9] that the retardation term in the effective potential at distances less than about $137a_0$ is proportional to x^{-4}.) Note that it is not possible, at the present time to verify the reality of the retardation correction, due to a combination of theoretical and experimental errors. For the 10*G*-10*H* interval, the experimental error is small enough in principle for the effect to be observable, but the theory is not good enough. On the other hand, for the 10*I*-10*K* interval the theoretical error is ten times smaller than the retardation correction, but here the experimental error is too large. There is a hint, in the 10*H*-10*I* case, of a disagreement; it is probably premature to worry too much about such a small discrepancy.

Future calculations should concentrate on improving the accuracy of the main terms in the energy shift, where the error comes from the poor convergence of the expansion in inverse powers of the distance, especially for the low L cases where the retardation correction is large. It will probably be necessary to abandon the approximation of retaining only the long-range parts of the electrostatic perturbation, although some of the simplicity of the method will unavoidably be lost. The higher terms in the relativistic polarizability and the recoil corrections should also be considered, but they are at present always too small to be significant.

Note added in proof. The authors of Ref. 9 have now calculated the retardation correction for the cases (in parentheses in Table III) previously omitted (private communication from G. Feinberg). The changes are 0.003 MHz for $6G$-$6H$, 0.002 MHz for $7G$-$7H$, 0.001 MHz for $8G$-$8H$ and $7H$-$7I$, and -0.001 MHz for $8H$-$8I$; there is no change for $8I$-$8K$.

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