

## Rydberg states of helium: Nuclear-recoil corrections

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The nuclear-recoil terms contributing to the fine-structure splitting of  $1sNL$  Rydberg states of helium are calculated. A great simplification resulting from the use of nonsymmetric Jacobi coordinates makes it possible to write the corrections to all terms in the asymptotic optical potential without additional computation. The isotope shifts between  $^4\text{He}$  and  $^3\text{He}$  are displayed and evaluated.

In two previous papers<sup>1,2</sup> the effective optical potential acting on the outer Rydberg electron in helium has been derived and applied to the fine-structure splitting for states of high angular momentum, as an extension of earlier work.<sup>3</sup> The somewhat surprising observation was made that the lowest-order correction for the nuclear recoil (mass polarization, specific mass effect) is proportional to the dipole polarization potential. As derived in Ref. 1 this appears to be a coincidence, and higher-order recoil corrections would need to be calculated one by one. In fact, the similarity between the dipole part of the perturbing potential and the recoil term in the perturbation has been noted recently,<sup>4</sup> while Martinis and Pilkuhn<sup>5</sup> made the same observation as the by-product of a calculation of relativistic effects. In this report the use of a slightly different coordinate system simplifies the recoil calculation greatly and explains the apparent coincidence.

Ordinarily, in calculations involving two-electron atoms, it is convenient to use as coordinates the vectors  $\mathbf{r}_i - \mathbf{r}_M$  describing the positions of the two electrons ( $i = 1, 2$ ) relative to the nucleus of mass  $M$ . This choice makes it easy to satisfy the Pauli principle, since the symmetry of the Hamiltonian is explicit, but it introduces the mass-polarization operator  $V_{MP} = -2/M \nabla_1 \cdot \nabla_2$ , which must be treated as a separate perturbation along with the usual perturbing potential.<sup>1,6</sup> For highly excited Rydberg states, where the outer electron does not overlap appreciably with the  $1s$  core electron, it is not necessary to use a symmetric coordinate system. Instead we may use the Jacobi coordinates defined as follows (we use Rydberg atomic units with the electron mass set equal to unity):

$$\mathbf{R} = \frac{M\mathbf{r}_M + \mathbf{r}_1 + \mathbf{r}_2}{M + 2}, \quad \mathbf{r}' = \mathbf{r}_1 - \mathbf{r}_M, \quad \mathbf{x}' = \mathbf{r}_2 - \frac{M\mathbf{r}_M + \mathbf{r}_1}{M + 1}. \quad (1)$$

Here  $\mathbf{R}$  is the position of the center of mass of the atom,  $\mathbf{r}'$  is the position of the inner electron relative to the nucleus, and  $\mathbf{x}'$  is the position of the outer electron relative to the center of mass of the core; the new feature is the definition of this last coordinate. The Hamiltonian in this form does not contain any cross terms in the momenta; the effect of the finite nuclear mass will appear in the potential. We next drop the kinetic energy of the center of

mass (which may be taken at rest) and further scale the coordinates as follows:  $\mathbf{r}' = \mathbf{r}/m$  and  $\mathbf{x}' = \mathbf{x}/m$ , where  $m$  is the reduced mass. The Hamiltonian is

$$H = - \left[ \nabla_r^2 + \frac{4}{r} \right] - \left[ \frac{1}{\mu} \nabla_x^2 + \frac{2}{x} \right] + \frac{2}{|\mathbf{x} - m\mathbf{r}|} + \frac{2}{x} - \frac{4}{|\mathbf{x} + \frac{1}{2}K\mathbf{r}|}, \quad (2)$$

in reduced Rydberg units  $R = mR_\infty$ , where  $m = 1 - K/2$ ,  $1/\mu = 1 - K^2/4$ , and  $K \equiv 2/(1 + M)$ . The first set of parentheses in Eq. (2) is the atomic Hamiltonian of the core while the second set is the Hamiltonian of the shielded outer electron, with a slightly modified mass  $\mu$ . Since both  $m$  and  $K$  are less than unity we can make the usual multipole expansion of the potential for  $r < x$ ,

$$V = \sum_{l=1}^{\infty} \left[ m^l - 2 \left[ -\frac{K}{2} \right]^l \right] \frac{2r^l}{x^{l+1}} P_l(\hat{\mathbf{x}} \cdot \hat{\mathbf{r}}). \quad (3)$$

Notice that this expansion differs from the conventional one<sup>1</sup> only because of the square bracket appearing as a factor in each term; for that reason it is trivial to rewrite every term in the optical potential to include the finite-mass effects. In addition, however, one must rewrite every expectation value appearing in the energy to account for the modified mass  $\mu$ ,

$$\langle x^{-s} \rangle \rightarrow \mu^s \langle x^{-s} \rangle. \quad (4)$$

Thus the apparently accidental proportionality of the leading mass-polarization correction to the dipole polarization term is easily understood, and it is obvious that every term in the long-range optical potential will have its nuclear-recoil counterpart. For example, the lowest term in the expansion of Eq. (3) corresponds to  $l = 1$ ,

$$\begin{aligned} \bar{U}(l=1) &= -\frac{\alpha_1}{x^4} \mu^4 (m + K)^2 \\ &= -\frac{\alpha_1}{x^4} \frac{1}{\left[ 1 - \frac{K}{2} \right]^2 \left[ 1 - \frac{K^2}{4} \right]^2} \\ &\simeq -\frac{\alpha_1}{x^4} (1 + K). \end{aligned} \quad (5)$$

In the same way, we find that the quadrupole potential, falling off like  $x^{-6}$ , is to be multiplied by  $1-2K$ , to first order. To the same order in  $K$ , the third-order potential term, falling like  $x^{-7}$ , is unmodified; since it contains two dipole terms and one quadrupole term it is multiplied by the factor  $(1+K)^2(1-2K)=1+O(K^2)$ . Simply by noting which multipole terms contribute to each term in the optical potential, one can derive the mass-polarization correction of any desired order in  $1/x$ . As a practical matter, no terms of higher order in  $K$  than the first need to be retained at the present level of accuracy of the experiments<sup>7</sup> (about 10 kHz). The main theory itself has conservatively estimated errors,<sup>1,2</sup> based on the observed rate of convergence of the asymptotic expansion in  $1/x$ , which are always of the same order of magnitude as the  $x^{-6}$  term in the mass-polarization correction. Thus, until further terms in the expansion have been evaluated only the dipole correction is of observational significance.

It is possible, however, to calculate the isotope effect on the fine-structure intervals for comparison with future experiments. The  $L$ -dependent shift in the  $NL$  level correct to first order in  $K$  is<sup>8</sup>

$$\begin{aligned} \frac{E(N,L)}{R_\infty} = & -\alpha_1 \left[ 1 + \frac{K}{2} \right] F_4(N,L) \\ & + \left[ 6\beta_1 \left[ 1 + \frac{K}{2} \right] - \alpha_2 \left( 1 - \frac{5}{2}K \right) \right] F_6(N,L) \\ & - \frac{\alpha^2(1-2K)}{N^3(L + \frac{1}{2})}, \end{aligned} \quad (6)$$

where  $\alpha_1, \alpha_2$  are dipole and quadrupole polarizabilities,  $\beta_1$  is the first nonadiabatic correction parameter,  $F_j(N,L)$  is the expectation value<sup>9</sup> of  $x^{-j}$ , and the last term is the usual correction for the relativistic increase of mass involving the fine-structure constant  $\alpha$ . Let  $\Delta_M(N,L) \equiv E(N,L+1) - E(N,L)$  be the level splitting for the isotope of mass  $M$ , and  $\delta(N,L) \equiv \Delta_3(N,L) - \Delta_4(N,L)$  be the shift between the two isotopes of helium in MHz. Then

TABLE I.  ${}^3\text{He}$ — ${}^4\text{He}$  isotope shifts in kHz for selected fine-structure intervals, defined as  $[E_3(N,L+1) - E_3(N,L)] - [E_4(N,L+1) - E_4(N,L)]$ .

$N,L - N,L + 1$	Isotope shift
6G-6H	81.4
7G-7H	53.1
8G-8H	36.4
9G-9H	25.9
10G-10H	19.1
11G-11H	14.5
7H-7I	15.5
8H-8I	10.8
9H-9I	7.8
10H-10I	5.8
11H-11I	4.4
8I-8K	3.4
9I-9K	2.5
10I-10K	1.9
11I-11K	1.5
9K-9L	0.8
10K-10L	0.6
11K-11L	0.5

$$\begin{aligned} \delta(N,L) = & -4.150 \times 10^4 \bar{F}_4(N,L) + 2.47 \times 10^5 \bar{F}_6(N,L) \\ & - \frac{125.7}{N^3(2L+1)(2L+3)}, \end{aligned} \quad (7)$$

where

$$\bar{F}_j(N,L) = F_j(N,L+1) - F_j(N,L).$$

[In this expression we have used the values  $K({}^3\text{He}) = 3.6384 \times 10^{-4}$  and  $K({}^4\text{He}) = 2.7415 \times 10^{-4}$ ]. In Table I the isotope shifts for selected transitions are displayed; some of them are large enough to be measurable.

<sup>1</sup>R. J. Drachman, Phys. Rev. A 26, 1228 (1982).

<sup>2</sup>R. J. Drachman, Phys. Rev. A 31, 1253 (1985).

<sup>3</sup>H. A. Bethe, *Handbuch der Physik* (Edwards, Ann Arbor, 1943), Vol. 24, Part 1, p. 339; A. Temkin and A. Silver, Phys. Rev. A 10, 1439 (1974); C. Deutsch, *ibid.* 13, 2311 (1976).

<sup>4</sup>C. Froese Fischer and L. Smentek-Mielczarek, J. Phys. B 16, 3479 (1983).

<sup>5</sup>M. Martinis and H. Pilkuhn, J. Phys. B 15, 1797 (1982).

<sup>6</sup>M. D. Girardeau, Phys. Rev. A 28, 3635 (1983).

<sup>7</sup>D. R. Cok and S. R. Lundeen, Phys. Rev. A 23, 2488 (1981); S.

L. Palfrey and S. R. Lundeen, Phys. Rev. Lett. 53, 1145 (1984).

<sup>8</sup>The  $K$ -dependent factors appearing in Eq. (6) include the factor  $1 - K/2$  coming from the ordinary reduced-mass effect. It is interesting that including the mass polarization correction to the leading (dipole) term produces an effect of magnitude equal to that of the reduced mass alone but of opposite sign. Note that only terms falling more slowly than  $x^{-7}$  have been retained here.

<sup>9</sup>K. Bockasten, Phys. Rev. A 9, 1087 (1974).