Leading-order relativistic corrections to the hydrogen molecular ion

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We present the improved calculations over the results of Korobov [Phys. Rev. A **74**, 052506 (2006)] for the energy levels and hyperfine structure of the hydrogen molecular ion H_2^+, including the relativistic corrections of orders α^2 Ry and $\alpha^2 m_e/m_p$ Ry, using variationally constructed basis sets in Hylleraas coordinates. In particular, the computational uncertainty of 3 kHz in Korobov's calculation for the α^2 Ry correction to the two-photon transition frequency $(L=0,v=0)\rightarrow(L=0,v=1)$ is eliminated.

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I. INTRODUCTION

In recent years, H_2^+ and its isotopes were cooled to a few mK sympathetically with laser-cooled beryllium ions by Schiller and co-workers [1,2] aiming at a spectroscopic accuracy of 10^{-10} level. Meanwhile, experiments on high-resolution spectroscopy for the two-photon transition $(L,v=0)\rightarrow(L,v=1)$ in H_2^+ were proposed by Karr *et al.* [3,4] to achieve a few kHz accuracy. The two-photon transition between the lowest rovibrational states of H_2^+ can be used to derive an improved value of the electron-proton mass ratio m_e/m_p [5,6]. Schiller and Korobov [7] studied in detail the theoretical dependence of the rovibrational transition frequency of H_2^+ on m_e/m_p.

The CODATA recommended values [8] of the fundamental constants for m_e/m_p is accurate to about five parts in 10^{10}. In order to obtain an improved value for m_e/m_p by 1 order of magnitude in precision, the theory and experiment must be accurate at least to a few parts in 10^{12} or better [2,9,10]. Thus, relativistic and QED corrections of orders α^5 Ry and α^6 Ry should be included in the calculations, which in turn requires very high-precision nonrelativistic wave functions. Korobov [11] evaluated the nonrelativistic rovibrational energy levels H_2^+ to a precision of 10^{-15} for $v=0-4$ and $L=0-4$. After that, the relativistic and radiative corrections up to α^6 Ry for H_2^+ and HD^+ have been calculated [11–14]. Relativistic corrections of order $\alpha^6(m/M)$ Ry to the hyperfine structure have also been estimated as well [15,16]. However, these results have not been confirmed independently. Furthermore, the computational uncertainty in the lowest-order relativistic correction of α^2 Ry is about 3 kHz, which is at the level of 40 parts in 10^{12} to the total transition frequency of $(0,0)\rightarrow(0,1)$ [11,14]. Up to now, the most precise nonrelativistic energies for the low-lying states of H_2^+ were determined variationally by Li *et al.* [10] and Hajikata *et al.* [17] using a completely different approach from [11]. As an example, the ground-state energy of H_2^+ has been calculated to 30 significant digits. The purpose of this Brief Report is to report our calculations for the leading relativistic corrections to the low-lying rovibrational states in H_2^+, which provides an independent verification of Korobov's calculations. At the same time, much-improved results for the energy levels and hyperfine structure will be presented. Atomic units $(\hbar=e=m_e=1)$ are used throughout this Brief Report, unless otherwise stated. The proton-electron mass ratio is chosen to be 1836.152 672 61(85) [18].

II. NONRELATIVISTIC WAVE FUNCTIONS

Consider the hydrogen molecular ion H_2^+. After eliminating the center of mass degrees of freedom, the eigenvalue problem for the nonrelativistic Hamiltonian H_0 becomes [10]

$$H_0\Psi = E_0\Psi, \tag{1}$$

with

$$H_0 = \lambda_1\nabla_{r_1}^2 + \lambda_2\nabla_{r_2}^2 + \lambda_{12}\boldsymbol{\nabla}_{r_1}\cdot\boldsymbol{\nabla}_{r_2} + V. \tag{2}$$

In the above, $\mathbf{r}_1$ and $\mathbf{r}_2$ are, respectively, the position vectors of the electron and one proton, relative to the other proton situated at the origin, $\lambda_1=-(1+m_p)/(2m_p)$, $\lambda_2=\lambda_{12}=-1/m_p$, $V=-1/r_1+1/r_2-1/r_{12}$ is the coulomb interaction among the three particles, and $\mathbf{r}_{12}=\mathbf{r}_1-\mathbf{r}_2$. The energy eigenvalue problem for H_0 is solved variationally in the following Hylleraas basis set:

$$\phi_{ijk}(\mathbf{r}_1,\mathbf{r}_2) = r_1^i r_2^j r_{12}^k e^{-\alpha r_1-\beta r_2}\mathcal{Y}_{l_1 l_2}^{LM}(\mathbf{r}_1,\mathbf{r}_2), \tag{3}$$

where $\mathcal{Y}_{l_1 l_2}^{LM}(\mathbf{r}_1,\mathbf{r}_2)$ is the vector coupled product of spherical harmonics for the electron and the proton. More details on the construction of basis set for H_2^+ can be found in [10]. The basic type of integrals required in the calculation of matrix elements can be evaluated analytically [19] using Perkins' expansion for r_{12}^c. Singularities of singular integrals that appear in the evaluation of Breit interaction can be canceled out according to the procedure in [20].

III. LEADING-ORDER RELATIVISTIC CORRECTIONS

The leading-order relativistic corrections of order α^2 from the Breit-Pauli Hamiltonian are well established and may be found in [11,21]. The complete spin-independent contributions are

$$H_{\text{spin-indep}} = \alpha^2 H_{\text{Breit}} + H_{\text{nuc}}, \tag{4}$$

where

$$H_{\mathrm{Breit}} = H_{\mathrm{rc}} + E_{\mathrm{kin}} + H_{\mathrm{tr\text{-}ph}} + H_{\mathrm{Darwin}}, \tag{5}$$

$$H_{\mathrm{rc}} = -\frac{\nabla_{r_1}^4}{8} + \frac{4\pi}{8}[\delta(\mathbf{r}_1) + \delta(\mathbf{r}_{12})], \tag{6}$$

$$H_{\mathrm{kin}} = 2\left[-\frac{1}{8m_p^3}\nabla_{r_2}^4\right], \tag{7}$$

$$\begin{aligned} H_{\mathrm{tr\text{-}ph}} = {} & \frac{1}{2m_p r_1^3}[r_1^2 \boldsymbol{\nabla}_{r_1}\cdot(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2}) + \mathbf{r}_1\mathbf{r}_1{:}(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2})\boldsymbol{\nabla}_{r_1}] \\ & - \frac{1}{2m_p^2 r_2^3}[r_2^2 \boldsymbol{\nabla}_{r_2}\cdot(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2}) + \mathbf{r}_2\mathbf{r}_2{:}(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2})\boldsymbol{\nabla}_{r_2}] \\ & - \frac{1}{2m_p r_{12}^3}[r_{12}^2 \boldsymbol{\nabla}_{r_1}\cdot\boldsymbol{\nabla}_{r_2} + \mathbf{r}_{12}\mathbf{r}_{12}{:}\boldsymbol{\nabla}_{r_1}\boldsymbol{\nabla}_{r_2}], \end{aligned} \tag{8}$$

$$H_{\mathrm{Darwin}} = \frac{1}{8m_p^2}[\delta(\mathbf{r}_1) + \delta(\mathbf{r}_{12})], \tag{9}$$

$$H_{\mathrm{nuc}} = \frac{2\pi(R_p/a_0)^2}{3}[\delta(\mathbf{r}_1) + \delta(\mathbf{r}_{12})]. \tag{10}$$

In the above, H_{rc} is the major relativistic contribution of order α^2 from the bound electron, H_{kin} is the correction due to the relativistic kinetic energy of the two protons, $H_{\mathrm{tr\text{-}ph}}$ is from the transverse photon exchange, H_{Darwin} is the Darwin term, and H_{nuc} is the leading-order correction from the finite nuclear charge distribution, where $R_p = 0.8768(69)$ fm [8] is the proton root-mean-square charge radius and a_0 is the Bohr radius. The global operator method, proposed by Drachman [22], is used to calculate some singular operators such as $\delta(\mathbf{r}_i)$ and $\nabla_{r_i}^4$.

IV. LEADING-ORDER RELATIVISTIC CORRECTIONS TO HYPERFINE STRUCTURE

The leading-order relativistic corrections to hyperfine structure of H_2^+ are described by the spin-dependent terms of Breit-Pauli Hamiltonian that sums up pairwise interactions of the three particles. The electron-proton and proton-proton spin-orbit interactions are expressed as

$$\begin{aligned} V_{\mathrm{so\text{-}pe}} = \alpha^2\Bigg[& -\frac{1+2\kappa_e}{2}\left(\frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_1}) + \frac{1}{r_{12}^3}(i\mathbf{r}_{12}\times\nabla_{r_1})\right)\cdot\mathbf{s}_e \\ & + \frac{1+\kappa_e}{m_p}\left(\frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_1}) + \frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_2}) \right. \\ & \left. + \frac{1}{r_{12}^3}(i\mathbf{r}_{12}\times\nabla_{r_2})\right)\cdot\mathbf{s}_e + \frac{1+2\kappa_p}{m_p^2}\left(\frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_1})\cdot\mathbf{I}_0 \right. \\ & \left. + \frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_2})\cdot\mathbf{I}_0 + \frac{1}{r_{12}^3}(i\mathbf{r}_{12}\times\nabla_{r_2})\cdot\mathbf{I}_2\right) \\ & - \frac{1+\kappa_p}{2m_p}\left(\frac{1}{r_1^3}(i\mathbf{r}_1\times\nabla_{r_1})\cdot\mathbf{I}_0 + \frac{1}{r_{12}^3}(i\mathbf{r}_{12}\times\nabla_{r_1})\cdot\mathbf{I}_2\right)\Bigg], \end{aligned} \tag{11}$$

$$\begin{aligned} V_{\mathrm{so\text{-}pp}} = \alpha^2\Bigg[& \frac{1+2\kappa_p}{2m_p^2}\left(\frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_1})\cdot\mathbf{I}_0 + \frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_2})\cdot\mathbf{I}_0 \right. \\ & \left. + \frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_2})\cdot\mathbf{I}_2\right) + \frac{\mu_p}{m_p^2}\left(\frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_1})\cdot\mathbf{I}_2 \right. \\ & \left. + \frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_2})\cdot\mathbf{I}_2 + \frac{1}{r_2^3}(i\mathbf{r}_2\times\boldsymbol{\nabla}_{r_2})\cdot\mathbf{I}_0\right)\Bigg], \end{aligned} \tag{12}$$

TABLE I. Convergence of $\langle\nabla_{r_1}^4\rangle$ and $\langle\nabla_{r_2}^4\rangle$ in the state (0,0). N denotes the size of basis set.

N	$\langle\nabla_{r_1}^4\rangle$	$\langle\nabla_{r_2}^4\rangle$
6084	6.2856600593122250	79.7976493122364
6795	6.2856600593122253	79.7976493122401
7560	6.2856600593122214	79.7976493122417
8381	6.2856600593122213	79.7976493122422
9260	6.2856600593122207	79.7976493122427
Extrap.	6.2856600593122200(7)	79.7976493122432(5)

TABLE II. Expectation values of various operators for some rovibrational states of H_2^+ and comparison to Korobov's work [11].

(L,v)	(0,0)	(1,0)	(0,1)
$\langle\nabla_{r_1}^4\rangle$	6.28566006	6.27803905	6.12451981
This work	6.2856600593122200(7)	6.278039037386287(5)	6.124519807747879(3)
$\langle\nabla_{r_2}^4\rangle$	79.7976	85.0505	334.898
This work	79.7976493122432(5)	85.050455613540(5)	334.89830192549(4)
$\langle\delta(\mathbf{r}_1)\rangle$	0.206736476	0.206491321	0.201310665
This work	0.20673647628881478 50(1)	0.20649132015881730 0(3)	0.20131066470208266 75(1)
R_{pe}	1.17012	1.16881	1.14081
This work	1.17011762503375469 4(1)	1.16881866381904511(8)	1.14080522713004732(2)
R_{pp}	4.60193	4.83433	12.8961
This work	4.60193431250485649 8(2)	4.83433647367036965 70(6)	12.89614649485648(2)

TABLE III. Numerical values for reduced matrix elements of orbital parts of spin-dependent operators in the state (1,0).

Operator	Value
$S_1^{(1)}$	$0.2704494968631(4)\times10^{-4}$
$S_2^{(2)}$	−0.2996739237238803(7)
$S_2^{(1)}$	−0.2968096504450887(5)
$S_1^{(2)}$	$0.139939978335(3)\times10^{-4}$
$S_1^{(12)}$	$0.270449496868(5)\times10^{-4}$
$S_2^{(12)}$	0.296782605495398(5)
T_{pe}	0.215395211855521(1)
T_{pp}	0.3283262297065680565838(1)

where $\mu_e=-(1+\kappa_e)$ is the magnetic moment of the electron in Bohr magneton, $\mu_p=1+\kappa_p$ is the magnetic moment of the proton in nuclear magneton, and $\mathbf{I}_0$, $\mathbf{I}_2$, and $\mathbf{s}_e$ are the spin operators for the two nuclei and the electron, respectively. The electron-proton and proton-proton spin-spin tensor interactions are expressed as

$$V_{\text{ss-ep}}=\alpha^2\frac{\mu_e\mu_p}{m_p}\left\{\frac{r_1^2(\mathbf{s}_e\cdot\mathbf{I}_0)-3(\mathbf{r}_1\cdot\mathbf{s}_e)(\mathbf{r}_1\cdot\mathbf{I}_0)}{r_1^5}+\frac{r_{12}^2(\mathbf{s}_e\cdot\mathbf{I}_2)-3(\mathbf{r}_{12}\cdot\mathbf{s}_e)(\mathbf{r}_{12}\cdot\mathbf{I}_2)}{r_{12}^5}-\frac{8\pi}{3}[\delta(\mathbf{r}_1)(\mathbf{s}_e\cdot\mathbf{I}_0)+\delta(\mathbf{r}_{12})(\mathbf{s}_e\cdot\mathbf{I}_2)]\right\}, \quad (13)$$

$$V_{\text{ss-pp}}=\alpha^2\frac{\mu_p^2}{m_p^2}\left[\frac{r_2^2(\mathbf{I}_0\cdot\mathbf{I}_2)-3(\mathbf{r}_2\cdot\mathbf{I}_0)(\mathbf{r}_2\cdot\mathbf{I}_2)}{r_2^5}-\frac{8\pi}{3}\delta(\mathbf{r}_2)(\mathbf{I}_0\cdot\mathbf{I}_2)\right]. \quad (14)$$

The nuclear spin-spin tensor interaction cannot be neglected and was first considered in [16].

V. CALCULATIONS AND RESULTS

Table I presents a convergence study for $\langle\nabla_{r_1}^4\rangle$ and $\langle\nabla_{r_2}^4\rangle$ in (0,0) evaluated using the global operator method. Numerical values for various spin-independent operators of the Breit interaction are listed in Table II, together with a comparison to Korobov's work [11]. Table III contains the numerical values for various spin-dependent operators in the state (1,0). The notations for orbit-orbit operators are as follows:

$$R_{pe}=-\left\langle\frac{1}{r_1^3}[r_1^2\boldsymbol{\nabla}_{r_1}\cdot(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2})+\mathbf{r}_1\mathbf{r}_1:(\boldsymbol{\nabla}_{r_1}+\boldsymbol{\nabla}_{r_2})\boldsymbol{\nabla}_{r_1}]\right\rangle, \quad (15)$$

and the definition for R_{pp} is obtained from R_{pe} by $1\leftrightarrow2$. The notations for the reduced matrix elements of orbital parts of spin-dependent operators are

$$S_j^{(i)}=\left\langle\left\|\frac{1}{r_i^3}(i\mathbf{r}_i\times\nabla_{r_j})\right\|\right\rangle,\quad S_j^{(12)}=\left\langle\left\|\frac{1}{r_{12}^3}(i\mathbf{r}_{12}\times\nabla_{r_j})\right\|\right\rangle,$$

$$T_{pe}=\langle\|[\mathbf{r}_1\otimes\mathbf{r}_1]^{(2)}\|\rangle,\quad T_{pp}=\langle\|[\mathbf{r}_2\otimes\mathbf{r}_2]^{(2)}\|\rangle, \quad (16)$$

where $i,j=1,2$ and the superscript 2 stands for a second-order tensor.

TABLE IV. Contributions to the transition frequency $(0,0)\rightarrow(0,1)$ in MHz.

	This work	Korobov
ΔE_{nr}	65687511.0686(4)	65687511.0686[a]
ΔE_{Breit}	1091.081355(2)	1091.081(03)[b]
ΔE_{nuc}	−0.0410(6)	−0.0410(3) [b]

[a]Table VI of Ref. [11].
[b]Obtained using data of Table IV in Ref. [11].

Table IV lists the contributions to the two-photon transition frequency $(0,0)\rightarrow(0,1)$ from the nonrelativistic energy, the Breit interaction, and the nuclear charge distribution. In the table, the uncertainty of 0.4 kHz in ΔE_{nr} is due entirely to the uncertainty in the Rydberg constant [8]. ΔE_{Breit} is the leading relativistic correction of order α^2. In Korobov's calculations [11,14], the uncertainty in ΔE_{Breit} is estimated to be 3 kHz that mainly comes from the computational uncertainty in $\langle\nabla_{r_1}^4\rangle$. By contrast, the uncertainty of 2 Hz in our value is due to the uncertainty in the fine-structure constant α. Additional, the uncertainty due to proton-electron mass ratio [16] in Breit interactions is about 0.003 Hz. The largest uncertainty of 0.6 kHz (about ten parts in 10^{12} to the total transition frequency) is from the proton root-mean-square charge radius, which is at the level of 0.8% accuracy. A muonic hydrogen Lamb-shift experiment is currently underway at

TABLE V. Hyperfine structure of H_2^+ for the state (1,0), in MHz.

	ΔE_{hfs}		$[C_1^{\pm},C_3^{\pm}]$	
$(\tilde{F},J)$	This work	Ref. [15]	This work	Ref. [[4]].
(1/2,3/2)	−930.433201(8)	−930.4332	[−0.999878130, 0.015611707]	[−0.999878, 0.015612]
(1/2,1/2)	−910.757885(8)	−910.7579	[−0.999243469, 0.038890742]	[−0.999243, 0.038891]
(3/2,5/2)	474.106321(4)	474.1063	[0, 1]	[0, 1]
(3/2,3/2)	481.953402(4)	481.9534	[0.015611707, 0.999878130]	[0.015612, 0.999878]
(3/2,1/2)	385.398521(4)	385.3985	[0.038890742, 0.999243469]	[0.038891, 0.999243]

TABLE VI. Numerical values of the hyperfine coefficients of the effective Hamiltonian for the state (1,0), in MHz.

Coefficient	This work	Ref. [15]	Ref. [24]	Ref. [25][a]
b_F	922.991798(8)	922.992	922.990	922.940(20)
c_e	42.41631862(4)	42.4163	42.421	42.348(29)
c_I	−0.0416729322(4)	−0.04168	−0.0417[b]	
d_1	128.489751(1)	128.490	128.482	128.259(26)
d_2	−0.297559002(5)	−0.2975		

[a]Experimental results.
[b]Reference [26].

the Paul Scherrer Institute [23] with a purpose of reducing the uncertainty of R_p to 0.1%. Thus a further reduction of uncertainty in ΔE_{nuc} to a few parts in 10^{12} is feasible in the near future.

The leading-order relativistic corrections to the hyperfine structure are calculated in the angular coupling scheme

$$\mathbf{I}=\mathbf{I}_0+\mathbf{I}_2, \quad \mathbf{F}=\mathbf{I}+\mathbf{s}_e, \quad \mathbf{J}=\mathbf{L}+\mathbf{F}.$$

The possible values of F and J are listed in [4]. It should be pointed out that F is an approximate quantum number for the case when L is odd. Thus, for an $L=1$ state, I can only be 1 and the eigenstates of $J=L\pm\frac{1}{2}$ are the linear combination of $F=1/2$ and $F=3/2$ hyperfine states [see Eq. (20) in [4]]

$$\left|v,L,s_e,I,\tilde{F},J=L\pm\frac{1}{2}\right\rangle = C_1^{\pm}\left|v,L,\frac{1}{2},1,\frac{1}{2},L\pm\frac{1}{2}\right\rangle + C_3^{\pm}\left|v,L,\frac{1}{2},1,\frac{3}{2},L\pm\frac{1}{2}\right\rangle. \tag{17}$$

Since the mixing between $F=1/2$ and $F=3/2$ states is quite weak, one can label the states by the dominant F, denoted by $\tilde{F}$. Here only the hyperfine structure of (1,0) state is considered. Our results are listed in Table V. The uncertainties in our calculations are mainly due to the proton magnetic moment. One can see that good agreement with [4,15] is obtained.

The effective spin Hamiltonian is used according to [15]

$$H_{\mathrm{eff}}=b_F(\mathbf{I}\cdot\mathbf{s}_e)+c_e(\mathbf{L}\cdot\mathbf{s}_e)+c_I(\mathbf{L}\cdot\mathbf{I}) + \frac{d_1}{(2L-1)(2L+3)}\left(\frac{2}{3}\mathbf{L}^2(\mathbf{I}\cdot\mathbf{s}_e)-[(\mathbf{L}\cdot\mathbf{I})(\mathbf{L}\cdot\mathbf{s}_e) + (\mathbf{L}\cdot\mathbf{s}_e)(\mathbf{L}\cdot\mathbf{I})]\right)+\frac{d_2}{(2L-1)(2L+3)}\left(\frac{1}{3}\mathbf{L}^2\mathbf{I}^2 - \frac{1}{2}(\mathbf{L}\cdot\mathbf{I})-(\mathbf{L}\cdot\mathbf{I})^2\right). \tag{18}$$

Numerical values of the coefficients contained in Eq. (18) are listed in Table VI. Uncertainties in the coefficients are mainly caused by the uncertainty in μ_p [8], while uncertainties from the operators in Table III are negligible. It is worthwhile to emphasize that the 10 Hz difference in c_I with Korobov's calculation [15] in Table VI is related to the discrepancy in $S_1^{(2)}$, where Korobov's value [27] is about 10^{-16} whereas ours is 10^{-5}. However, it will not introduce noticeable effect on the final results of the hyperfine splittings at the level of kHz [16].

In conclusion, we have presented an independent calculation of the leading-order relativistic corrections to the (0,0), (0,1), and (1,0) states of H_2^+, including the hyperfine structure. The computational uncertainties that exist in previous work have now been eliminated and all uncertainties are due entirely to the fundamental physical constants.

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