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Bethe logarithm for the $H_2{}^+$ and HD^+ molecular ions

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The Bethe logarithm is calculated for the lowest rotational-vibrational states of $H_2{}^+$ and HD^+ hydrogen molecular ions in a wide range of vibrational ($v = 0$–4) and total orbital momentum ($L = 0$–4) quantum numbers. Numerical results with eight to nine significant digits are obtained for all the states within this range. This allows us to reduce an error in the leading-order radiative contribution, which results eventually in the relative uncertainty of rovibrational frequency intervals at a level lower than 10^{-11}. This high precision is important for the rovibrational spectroscopy experiments of hydrogen molecular ions aiming to determine the electron-to-proton mass ratio.

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I. INTRODUCTION

Several laser spectroscopy experiments [1–3] have been carried out recently to get high-precision measurements of vibrational spectra of the hydrogen molecular ions $H_2{}^+$ and HD^+. Aiming at sub-ppb precision, these measurements are supposed to be used [4,5] to improve a value of the electron-to-proton mass ratio by comparing with theoretical data. The importance of the m_p/m_e problem is supported by recent experiments [6] with rubidium atoms, which allow to deduce a new value of the fine-structure constant, $\alpha = e^2/(\hbar c)$, with a relative uncertainty 6.6×10^{-10}. Further improvement may be hindered by the present limits on the proton-to-electron mass ratio, which is known with a relative uncertainty 5.2×10^{-10} [7,8].

Nonrelativistic energies are obtained with numerical precision of 10^{-15} [5,9] for a wide range of vibrational states and up to 10^{-30} [10–14] for some particular low vibrational states of $H_2{}^+$ and HD^+. To calculate the observable transition frequency interval, one needs as well to include quantum electrodynamics (QED) corrections. For light systems, the most natural way is to use the nonrelativistic QED (NRQED) [15,16], where a bound state energy is expanded in powers of the fine-structure constant α. The leading-order relativistic corrections ($R_\infty\alpha^2$) are now available with very high precision [17,18]. The next term is the one-loop radiative corrections that contribute to the order $R_\infty\alpha^3$ [17,19,20]. The main difficulty at this order is to calculate the Bethe logarithm, and this has remained the major source of numerical uncertainty for the fundamental vibrational transitions $[(L = 0, v = 0) \to (L' = 0, v' = 1)]$ in $H_2{}^+$ and HD^+ ions of about 20 kHz. For higher-order corrections, recoil effects become negligible, and the contribution of the $R_\infty\alpha^4$ order can already be calculated in a nonrecoil limit [21] with sufficient accuracy.

The major aim of this work is to recalculate improved values of the Bethe logarithm for a wide range ($v = 0$–4, $L = 0$–4) of rovibrational states in $H_2{}^+$ and HD^+ hydrogen molecular ions using a recently developed method [22] based on direct integration over the virtual photon energy. The latter method evolved from the Schwartz approach [23], which was the best calculation of the Bethe logarithm for the ground state of a helium atom for over 30 years. Atomic units ($\hbar = e = m_e = 1$) are used throughout the paper.

We use the notation conventional for molecular-type three-body systems. Thus, the space configuration of a molecular ion is described by the following coordinates: $\mathbf{r}_i = \mathbf{r}_e - \mathbf{R}_i$ ($i = 1,2$) and $\mathbf{R} = \mathbf{R}_2 - \mathbf{R}_1$, where $\mathbf{R}_i$ denotes the position vectors of nuclei. Correspondingly, the charges are $Z_e = -1$, $Z_1 = Z_2 = 1$ and the masses are m_e, M_i.

II. THEORY

The complete spin-independent effective Hamiltonian of order $R_\infty\alpha^3$ and $R_\infty\alpha^3(m_e/M)$ for a one-electron molecular system may be expressed as follows [24,25]:

$$\begin{aligned}\delta^{(3)}E = \alpha^3 \sum_i \Bigg[&\frac{4Z_i}{3}\left(-\ln\alpha^2 - \beta(v,L) + \frac{19}{30}\right)\langle\delta(\mathbf{r}_i)\rangle \\ &+ \frac{2Z_i^2}{3M_i}\left(-\ln\alpha - 4\beta(v,L) + \frac{31}{3}\right)\langle\delta(\mathbf{r}_i)\rangle \\ &- \frac{14Z_i^2}{3M_i}Q(r_i)\Bigg], \end{aligned} \tag{1}$$

where

$$\beta(v,L) = \frac{\mathcal{N}(v,L)}{\mathcal{D}(v,L)} = \frac{\langle \mathbf{J}(H_0 - E_0)\ln[(H_0 - E_0)/R_\infty]\mathbf{J}\rangle}{\langle[\mathbf{J},[H_0,\mathbf{J}]]/2\rangle} \tag{2}$$

is the Bethe logarithm. Here, H_0 is the three-body nonrelativistic Hamiltonian and E_0 is an energy of a state with quantum numbers v and L, vibrational and total orbital momentum, respectively. The state is a solution of the stationary Schrödinger equation,

$$(H_0 - E_0)\psi_0 = 0.$$

The operators, which appear in Eqs. (1) and (2), are $\mathbf{J} = -Z_e\mathbf{p}_e + \sum_{i=1}^{2} Z_i\mathbf{P}_i/M_i$, which is the electric current density

TABLE I. Asymptotic coefficient C_3, Eq. (11), for $H_2{}^+$ of rovibrational states ($v = 0$–4, $L = 0$–4). $N = 4000$.

	$v = 0$	$v = 1$	$v = 2$	$v = 3$	$v = 4$
$L = 0$	2.24754287	2.24333059	2.23916276	2.23501032	2.23086435
$L = 1$	2.24752822	2.24331484	2.23914249	2.23499095	2.23084327
$L = 2$	2.24749705	2.24328179	2.23910696	2.23494986	2.23080007
$L = 3$	2.24744640	2.24322774	2.23904902	2.23488251	2.23072687
$L = 4$	2.24737144	2.24314762	2.23896023	2.23478453	2.23061456

operator of the system, and $Q(r)$ is the Q term introduced by Araki and Sucher [26],

$$Q(r) = \lim_{\rho\to 0}\left\langle \frac{\Theta(r-\rho)}{4\pi r^3} + (\ln\rho + \gamma_E)\delta(\mathbf{r})\right\rangle . \tag{3}$$

The denominator in Eq. (2) can be expanded as follows:

$$\mathcal{D}(v,L) = 2\pi \sum_{i=1}^{2} Z_i \left(\frac{1}{m_e} + \frac{Z_i}{M_i}\right)^2 \langle\delta(\mathbf{r}_i)\rangle. \tag{4}$$

Here we neglect the small term proportional to $\langle\delta(\mathbf{R})\rangle$, because the latter is of order 10^{-15} or even less. It is convenient to express the numerator in the form of integration over photon energy [19,22]:

$$\begin{aligned}\mathcal{N}(v,L) = {} & \int_0^{E_h} k\,dk \left\langle \mathbf{J}\left(\frac{1}{E_0 - H_0 - k} + \frac{1}{k}\right)\mathbf{J}\right\rangle \\ & + \int_{E_h}^{\infty} \frac{dk}{k}\left\langle \mathbf{J}\frac{(E_0-H_0)^2}{E_0-H_0-k}\mathbf{J}\right\rangle,\end{aligned} \tag{5}$$

where E_h is the Hartree energy. Thus, $\beta(v,L)$ may be easily obtained if precise approximation of the following functions is available:

$$\begin{aligned}J(k) &= \langle \mathbf{J}(E_0 - H_0 - k)^{-1}\mathbf{J}\rangle \\ &= -\frac{1}{k}\langle\mathbf{J}^2\rangle + \frac{1}{k^2}\frac{\langle[\mathbf{J},[H_0,\mathbf{J}]]\rangle}{2} + \frac{1}{k^2}\,w(k), \\ w(k) &= \left\langle \mathbf{J}\frac{(E_0-H_0)^2}{E_0-H_0-k}\mathbf{J}\right\rangle.\end{aligned} \tag{6}$$

A. Low-energy contribution

For the low-energy part, $k \in [0,K_{\max}]$, we solve the equation

$$(E_0 - H_0 - k)\psi_1 = i\mathbf{J}\psi_0 \tag{7}$$

using variational expansion for ψ_1 [22]. In earlier calculations, we solved this equation on a sequence of energy intervals $[k_i,k_{i+1}]$ to comply with the requirement that ψ_1 should contain terms which behave as $e^{-\sqrt{2k}\,r_i}$. If one collects basis sets made up for these intervals into one set of intermediate states, one may expect that the final result would not be less accurate than in a previous approach [19]. On the other hand, the Hamiltonian can be diagonalized to get energies E_n and dipole matrix elements $\langle\psi_0|i\mathbf{J}|\psi_1^{(n)}\rangle$ for states and pseudostates of the Hamiltonian spanned over the subspace of the variational basis set. Then, using obtained data, the function $J(k)$ is expressed as

$$J(k) = -\sum_n \frac{\left|\langle\psi_0|i\mathbf{J}|\psi_1^{(n)}\rangle\right|^2}{E_0 - E_m - k}\,, \tag{8}$$

and integration of (8) can be performed analytically:

$$\begin{aligned}&\int_0^{K_{\max}} kd\,k\,J(k) \\ &\quad = \sum_n \left|\langle\psi_0|i\mathbf{J}|\psi_1^{(n)}\rangle\right|^2 \\ &\qquad \times \left[K_{\max} - (E_0 - E_n)\ln\left|\frac{E_0 - E_n}{E_0 - E_n - K_{\max}}\right|\right].\end{aligned} \tag{9}$$

Here $K_{\max}$ is some intermediate energy ($K_{\max} \sim 10^3$–10^5), which is taken to optimize the precision of the calculation. The larger $K_{\max}$ is, the larger the basis set has to be for the intermediate states to provide the necessary precision for $J(k)$ within the range of $k \in [0,K_{\max}]$. That in turn improves extrapolation of an asymptotic expansion (see the next subsection). On the other hand, convergence of numerical $J(k)$ to its exact value becomes worse with an increase of k, which forces the choice of $K_{\max}$ to be as low as possible for a given precision.

TABLE II. Asymptotic coefficient C_3, Eq. (11), for HD^+ of rovibrational states ($v = 0$–4, $L = 0$–4). $N = 4000$.

	$v = 0$	$v = 1$	$v = 2$	$v = 3$	$v = 4$
$L = 0$	2.24841853	2.24476334	2.24114194	2.23754139	2.23395070
$L = 1$	2.24840722	2.24475167	2.24112987	2.23752807	2.23393654
$L = 2$	2.24838417	2.24472700	2.24110278	2.23749760	2.23390349
$L = 3$	2.24834681	2.24468755	2.24106022	2.23745069	2.23385023
$L = 4$	2.24829292	2.24463111	2.24099934	2.23738344	2.23377521

TABLE III. Test of convergence of the Bethe logarithm quantity for the $H_2{}^+$ ($v = 4, L = 0$) state. N_a is the basis length for the initial state; N_b is the basis length for the intermediate state.

	N_a			
N_b	3000	4000	5000	∞
7000	3.0123774610	3.0123774692	3.0123773656	
8000	3.0123777946	3.0123777400	3.0123777225	
9000	3.0123778020	3.0123777707	3.0123777551	
∞				3.01237775(6)

B. High-energy contribution

For $k \in [K_{\max},\infty]$, an asymptotic expansion for $w(k)$ is used:

$$\begin{aligned} w(k) = &-\sum_{i=1}^{2}\left(\frac{1}{m_e}+\frac{Z_i}{M_i}\right)^2 \frac{1}{k}\left[Z_i^2\sqrt{2\mu_i k} - Z_i^3\mu_i \ln k\right]4\pi\langle\delta(\mathbf{r}_i)\rangle \\ &+2\pi\left[\sum_{i=1}^{2} Z_i\langle\delta(\mathbf{r}_i)\rangle\right]\left\{-\frac{C_3}{k}+\sum_{m=1}^{\infty}\frac{1}{k^{m+1}}\left[C_{1m}\sqrt{k}+C_{2m}\ln(k)+C_{3m}\right]\right\}, \end{aligned} \tag{10}$$

where $\mu_i = m_e M_i/(m_e+M_i)$ are the reduced masses. In Eq. (10), the coefficient C_3 may be calculated explicitly from the initial state solution:

$$\begin{aligned} C_3 \times 2\pi\left[\sum_{i=1}^{2} Z_i\langle\delta(\mathbf{r}_i)\rangle\right] = &\,2Z_1Z_2\left(\frac{1}{m_e}+\frac{Z_1}{M_1}\right)\left(\frac{1}{m_e}+\frac{Z_2}{M_2}\right)\left\langle\frac{\mathbf{r}_1\mathbf{r}_2}{r_1^2 r_2^2}\right\rangle \\ &+\sum_{i=1}^{2} Z_i^2\left(\frac{1}{m_e}+\frac{Z_i}{M_i}\right)^2\{4\pi\mathcal{R}_i + Z_i\mu_i(-\ln\mu_i+\ln 2+1)4\pi\langle\delta(\mathbf{r}_i)\rangle\}, \end{aligned} \tag{11}$$

where

$$\begin{aligned} \mathcal{R} &= \lim_{\rho\to 0}\left\{\left\langle\frac{1}{4\pi r^4}\right\rangle_{\rho} - \left[\frac{1}{\rho}\langle\delta(\mathbf{r})\rangle + (\ln\rho+\gamma_E)\langle\delta'(\mathbf{r})\rangle\right]\right\}, \\ \langle\phi_1|\delta'(\mathbf{r})|\phi_2\rangle &= \langle\phi_1|\frac{\mathbf{r}}{r}\boldsymbol{\nabla}\delta(\mathbf{r})|\phi_2\rangle = -\langle\partial_r\phi_1|\delta(\mathbf{r})|\phi_2\rangle - \langle\phi_1|\delta(\mathbf{r})|\partial_r\phi_2\rangle\,. \end{aligned} \tag{12}$$

Subtracting the known terms of the asymptotic expansion from the numerically obtained $w(k)$ [Eqs. (6) and (8)], one may approximate the remaining part by

$$f_{\text{fit}}(k) \approx \sum_{m=1}^{M}\frac{C_{1m}\sqrt{k}+C_{2m}\ln k + C_{3m}}{k^{m+3}}. \tag{13}$$

The coefficients C_{1m}, C_{2m}, and C_{3m} are evaluated by using a least-squares approximation of $f_{\text{fit}}(k)$ at a set of points $k_i \in [k_{\min},k_{\max}]$ for $k_{\min} \sim 10$ and $k_{\max} \sim 10^3$–10^4. Then $w(k)$ is integrated analytically on $[K_{\max},\infty]$. In actual calculations, we use the best fit of $f_{\text{fit}}(k)$ with a number of terms $n = 10$–16.

III. CALCULATION AND RESULTS

For vibrational calculations in $H_2{}^+$ and HD^+, the wave functions both for the initial bound states and for the

TABLE IV. The Bethe logarithm for the lowest rotational, L, and vibrational, v, states of the hydrogen molecular ion $H_2{}^+$.

	$v = 0$	$v = 1$	$v = 2$	$v = 3$	$v = 4$
$L = 0$	3.012230335(1)	3.012547548(3)	3.01267873(2)	3.01262269(4)	3.01237775(6)
$L = 1$	3.01220132(1)	3.01251393(2)	3.01264054(3)	3.01258051(4)	3.0123316(1)
$L = 2$	3.01214395(1)	3.01244742(2)	3.01256542(3)	3.01249674(4)	3.0122395(1)
$L = 3$	3.01205949(2)	3.01234936(3)	3.01245429(4)	3.01237302(5)	3.0121036(1)
$L = 4$	3.01194983(3)	3.01222182(3)	3.01230955(5)	3.01221169(6)	3.0119263(2)

TABLE V. The Bethe logarithm for the lowest rotational, L, and vibrational, v, states of the hydrogen molecular ion HD^+.

	$v=0$	$v=1$	$v=2$	$v=3$	$v=4$
$L=0$	3.01233626(2)	3.01263268(3)	3.01278948(4)	3.01280622(6)	3.0126822(1)
$L=1$	3.01231470(2)	3.01260814(3)	3.01276198(5)	3.0127760(1)	3.0126490(1)
$L=2$	3.01227206(2)	3.01255942(3)	3.01270766(5)	3.0127160(1)	3.0125836(2)
$L=3$	3.01220877(3)	3.01248727(4)	3.01262691(6)	3.0126269(1)	3.0124865(2)
$L=4$	3.01212616(4)	3.01239292(4)	3.0125211(1)	3.0125102(1)	3.0123593(2)

intermediate state are taken in the form

$$\Psi_L(l_1,l_2) = \sum_{i=1}^{\infty}\{U_i\,\mathrm{Re}[e^{-\alpha_i R-\beta_i r_1-\gamma_i r_2}] + W_i\,\mathrm{Im}[e^{-\alpha_i R-\beta_i r_1-\gamma_i r_2}]\}\mathcal{Y}^{l_1,l_2}_{LM}(\hat{\mathbf{R}},\hat{\mathbf{r}}_1), \qquad (14)$$

where $\mathcal{Y}^{l_1,l_2}_{LM}(\hat{\mathbf{R}},\hat{\mathbf{r}}_1)$ are the solid bipolar harmonics as defined in Ref. [27], and L is the total orbital angular momentum of a state. Complex parameters α_i, β_i, and γ_i are generated in a quasirandom manner [10]:

$$\alpha_i = \left[\left\lfloor \frac{1}{2}i(i+1)\sqrt{p_\alpha}\right\rfloor (A_2 - A_1) + A_1\right] + i\left[\left\lfloor \frac{1}{2}i(i+1)\sqrt{q_\alpha}\right\rfloor (A'_2 - A'_1) + A'_1\right], \qquad (15)$$

where $\lfloor x \rfloor$ designates the fractional part of x, p_α and q_α are some prime numbers, and $[A_1,A_2]$ and $[A'_1,A'_2]$ are real variational intervals which need to be optimized. Parameters β_i and γ_i are obtained in a similar way.

For an initial state with nonzero L, its intermediate states span over $L' = L, L\pm 1$ with the spatial parity $\pi = -(-1)^L$. A basis set of intermediate states is composed of a regular part and two extra short-distance trial functions (for $\mathbf{r}_i \to 0$, $i = 1,2$) with exponentially growing parameters (see details in Ref. [22]). To maintain the required numerical stability, quadruple and sextuple precision arithmetics have been used.

The numerically obtained values of C_3 for particular rovibrational states are presented in Table I for the ${H_2}^+$ molecular ion and Table II for the HD^+ molecular ion, respectively. The data have been obtained from the variational bound state wave functions, Eq. (14), with the basis size $N = 4000$. A relative accuracy of about 10^{-7}–10^{-8} is reached, which corresponds approximately to the precision of the δ-function operator expectation values.

Convergence of the numerical value for the nonrelativistic Bethe logarithm, $\beta(4,0)$, for the rovibrational state with total angular momentum $L = 0$ and vibrational quantum number $v = 4$ is studied in Table III. As is seen, it is essential to analyze convergence in two ways: as a function of an increasing basis set (N_a) of the initial state and as a function of the basis size of an intermediate state (N_b). It is worth noting that the lower the vibrational state is, the better precision may be achieved. From this table, one may conclude that for the vibrational state, $v = 4$, an accuracy of eight significant digits is reached.

Tables IV and V present numerical results of the Bethe logarithm calculations for the ${H_2}^+$ and HD^+ rovibrational states; the numerical uncertainty is indicated in parentheses. The discrepancies of our results with previously published data [19,20] has already been discussed in Ref. [22] and is due to inclusion of reduced masses, μ_i, in the improved asymptotic expansion [see Eq. (10)].

Using Eq. (1), improved radiative corrections of the $R_\infty\alpha^3$ order for ${H_2}^+$ and HD^+ fundamental transitions $(0,0) \to (1,0)$ may be obtained,

$$\Delta v({H_2}^+) = -276.545\,049(4)\mathrm{MHz},$$
$$\Delta v(HD^+) = -242.126\,26(4)\mathrm{MHz}.$$

For these estimates, we have used numerical data for mean values of operators, $\langle\delta(\mathbf{r}_i)\rangle$ and $Q(\mathbf{r}_i)$, obtained with 11 and 8 significant digits, respectively. To do this, the Schrödinger wave functions for the states of interest were calculated with a basis size of $N = 5000$. The error bar due to numerical evaluation of these operators is below 1 Hz. Uncertainty, which is introduced by the Bethe logarithm calculations, is about 4 Hz for the ${H_2}^+$ molecular ion, while for HD^+ it is slightly higher, ~40 Hz. For other transitions, either pure rotational or vibrational overtones, the final fractional uncertainty in theoretical frequency, which stems from the $R_\infty\alpha^3$ order contribution, does not exceed 10^{-11}.

In conclusion, a systematic study of the Bethe logarithm for a wide range of ro-vibrational states in the hydrogen molecular ions ${H_2}^+$ and HD^+ has been carried out, and numerical accuracy of eight to nine significant digits has been achieved. This allowed us to reduce the numerical errors in the theoretical contribution of order $R_\infty\alpha^3$ and to comply with precision requirements necessary for a determination of the electron-to-proton mass ratio m_e/m_p.

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