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Two-Photon Decay Rate of the $2\,^1S_0$ Metastable State of Helium†

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Two alternative expressions for the two-photon decay rate of the $2\,^1S_0$ metastable state of helium are evaluated by performing the summations over intermediate $^1P_1$ states. The values obtained from the "length" and "velocity" expressions for the decay rate are 50.85 sec^{-1} and 50.89 sec^{-1}, respectively, in good agreement with a recent experimental value and with previous calculations done by different methods. Incidental results are given for the photoionization cross sections of the $1\,^1S_0$ and $2\,^1S_0$ states of helium in the energy region 0–0.2 Ry.

I. INTRODUCTION

The radiative lifetime of the $2\,^1S_0$ metastable state of helium is of fundamental interest because single-photon decay to the $1\,^1S_0$ ground state is strictly forbidden by the selection rule $J=0 \nrightarrow J=0$. This selection rule is violated only by the presence of nuclear spin. Breit and Teller[1] pointed out that the $2\,^1S_0$ state of helium should decay predominantly by two-photon electric-dipole emission. This decay process gives rise to a continuous emission spectrum, since the only restriction on the photon energies is that their sum equal the energy difference between the $2\,^1S_0$ and $1\,^1S_0$ states.

The two-photon decay of the $2\,^1S_0$ state proceeds through virtual transitions to intermediate $^1P_1$ states. In order to evaluate directly the expression for the decay rate, an infinite summation over the intermediate $^1P_1$ states (which includes integrations over the continua) has to be performed for each pair of photon energies. Dalgarno[2] first attempted a direct evaluation of the expression for the two-photon decay rate of the $2\,^1S_0$ state by explicitly carrying out the summations over the intermediate states. In subsequent calculations, Dalgarno and Victor[3,4] avoided doing the infinite summations over intermediate states by carrying out variational calculations for the complete perturbed wave function for each pair of photon energies. In a recent calculation of the two-photon decay rates of the $2\,^1S_0$ and $2\,^3S_1$ states of heliumlike ions, Drake, Victor, and Dalgarno[5] conveniently performed the summations over intermediate states by replacing the sets of true excited-state wave functions by discrete sets of variationally determined functions.

In the calculation of the two-photon decay rate described in this paper, the summations over the intermediate $^1P_1$ states are explicitly performed. Use is made of the oscillator strengths recently obtained by Schiff, Pekeris, and Accad[6] for the electric-dipole transitions from the $1\,^1S_0$ and $2\,^1S_0$ states to the lowest four discrete $^1P_1$ states. An expansion into a complete discrete basis set (a modification of the close-coupling method used to describe electron scattering by hydrogenlike ions) has been carried out for the $^1P_1$ continuum wave functions. These wave functions have been applied with success in an evaluation of the photoionization cross sections of the $1\,^1S_0$ and $2\,^1S_0$ states of helium.[7]

Pearl[8] has reported an experimental value of $(38\pm8)\times10^{-3}$ sec for the radiative lifetime τ of the $2\,^1S_0$ metastable state of helium. However, a more recent series of measurements made by Van Dyck and co-workers[9,10] has yielded the result $\tau=(20\pm1)\times10^{-3}$ sec, which is in good agreement with the theoretical value of $\tau=19.5\times10^{-3}$ sec calculated by Drake, Victor, and Dalgarno.[5]

II. THEORY OF TWO-PHOTON EMISSION

The probability for the simultaneous electric-dipole emission of two photons with one photon in the frequency range from ν_1 to $\nu_1+d\nu_1$ is given (in sec^{-1}) by[1]

$$A(\nu_1)\,d\nu_1=\frac{2^{10}\pi^6e^4}{h^2c^6}\,\nu_1^3\nu_2^3\,d\nu_1 \times\left\langle\left|\sum_{n'}\left[\frac{\langle 1\,^1S|\vec{R}\cdot\vec{\epsilon}_1|n'\rangle\langle n'|\vec{R}\cdot\vec{\epsilon}_2|2\,^1S\rangle}{\nu_{n'2}+\nu_2}\right.\right.$$

$$+\frac{\langle 1\,^1S|\vec{R}\cdot\vec{\epsilon}_2|n'\rangle\langle n'|\vec{R}\cdot\vec{\epsilon}_1|2\,^1S\rangle}{\nu_{n'2}+\nu_1}\Bigg]\Bigg|^2\Bigg\rangle_{\rm av}, \quad (1)$$

where the summation is taken over the triply degenerate $^1P_1$ states and includes integrations over the continua; $\nu_{n'2}$ is the frequency of the $n'\,^1P_1 \rightarrow 2\,^1S_0$ transition; ν_2 is the frequency of the second photon; $\vec{R}=\vec{r}_1+\vec{r}_2$, where $\vec{r}_1$ and $\vec{r}_2$ are the electronic coordinates measured with respect to the nucleus; and $\vec{\epsilon}_1$ and $\vec{\epsilon}_2$ are unit vectors parallel to the directions of polarization of the two photons. The averaging indicated by $\langle\,\rangle_{\rm av}$ is to be taken over the directions of propagation and over the directions of polarization independently for ν_1 and ν_2. Since the $2\,^1P_1$ level lies above the $2\,^1S_0$ level, no cascade emission process of the type $2\,^1S_0 \rightarrow 2\,^1P_1 \rightarrow 1\,^1S_0$ can affect the probability of two-photon emission.

An alternative expression for the probability of two-photon emission can be derived by proceeding directly from the usual expression for the second-order transition amplitude[11] written down with matrix elements of the momentum operator $\vec{P}=\vec{p}_1+\vec{p}_2$ in place of the position operator $\vec{R}$. The expression thus obtained for $A(\nu_1)\,d\nu_1$ is given by

$$A(\nu_1)\,d\nu_1=\frac{2^6\pi^2e^4}{h^2c^4m^4}\,\nu_1\nu_2\,d\nu_1 \times\Bigg\langle\Bigg|\sum_{n'}\Bigg[\frac{\langle 1\,^1S|\vec{P}\cdot\vec{\epsilon}_1|n'\rangle\langle n'|\vec{P}\cdot\vec{\epsilon}_2|2\,^1S\rangle}{\nu_{n'2}+\nu_2}$$

$$+\frac{\langle 1\,^1S|\vec{P}\cdot\vec{\epsilon}_2|n'\rangle\langle n'|\vec{P}\cdot\vec{\epsilon}_1|2\,^1S\rangle}{\nu_{n'2}+\nu_1}\Bigg]\Bigg|^2\Bigg\rangle_{\rm av}. \quad (2)$$

The equivalence of Eqs. (1) and (2) can be established by employing the following identities[11]:

$$\langle i|\vec{P}|j\rangle=2\pi i m\nu_{ij}\langle i|\vec{R}|j\rangle, \quad (3)$$

$$\sum_{n'}[\langle i|\vec{R}\cdot\vec{\epsilon}_1|n'\rangle\langle n'|\vec{R}\cdot\vec{\epsilon}_2|j\rangle - \langle i|\vec{R}\cdot\vec{\epsilon}_2|n'\rangle\langle n'|\vec{R}\cdot\vec{\epsilon}_1|j\rangle]=0, \quad (4)$$

$$\sum_{n'}[\langle i|\vec{P}\cdot\vec{\epsilon}_1|n'\rangle\langle n'|\vec{R}\cdot\vec{\epsilon}_2|j\rangle - \langle i|\vec{R}\cdot\vec{\epsilon}_2|n'\rangle\langle n'|\vec{P}\cdot\vec{\epsilon}_1|j\rangle] = -i\hbar\vec{\epsilon}_1\cdot\vec{\epsilon}_2\,\delta_{ij}. \quad (5)$$

These two expressions for the probability of two-photon emission would give identical results if they could be evaluated using exact solutions of the nonrelativistic wave equation. When only approximate wave functions are available, the difference between the two results should provide some measure of the uncertainties in the wave functions.

Carrying out the averaging over propagation and polarization directions as described by Breit and Teller[1] and converting to atomic units, Eqs. (1) and (2) become

$$A_L(\nu_1)\,d\nu_1=\tfrac{1}{3}(E_2-E_1)^7\,\alpha^6y^3(1-y)^3\,dy \times\Bigg|\sum_{n=2}^{\infty}\langle 1\,^1S|z_1+z_2|n^1P\rangle\langle n\,^1P|z_1+z_2|2\,^1S\rangle\Bigg(\frac{1}{E_n-E_2+(E_2-E_1)y}+\frac{1}{E_n-E_2+(E_2-E_1)(1-y)}\Bigg)\Bigg|^2, \quad (6)$$

$$A_V(\nu_1)\,d\nu_1=\tfrac{16}{3}(E_2-E_1)^3\,\alpha^6y(1-y)\,dy \times\Bigg|\sum_{n=2}^{\infty}\Bigg\langle 1\,^1S\Bigg|\frac{\partial}{\partial z_1}+\frac{\partial}{\partial z_2}\Bigg|n\,^1P\Bigg\rangle\Bigg\langle n\,^1P\Bigg|\frac{\partial}{\partial z_1}+\frac{\partial}{\partial z_2}\Bigg|2\,^1S\Bigg\rangle\Bigg(\frac{1}{E_n-E_2+(E_2-E_1)y}+\frac{1}{E_n-E_2+(E_2-E_1)(1-y)}\Bigg)\Bigg|^2. \quad (7)$$

The summation is now taken over the principal quantum number n, which must be extended into the continuous spectrum. α is the fine-structure constant, and y is the fraction of the $2\,^1S_0-1\,^1S_0$ energy difference carried away by one of the two photons ($\nu_1=y\nu_{21}$). E_1, E_2, and E_n denote the energies of the $1\,^1S_0$, $2\,^1S_0$, and $n\,^1P_1$ states, respectively, measured in rydbergs.

The radiative lifetime τ and the total transition probability A (decay rate) are obtained (in rydberg frequency units) from Eqs. (6) and (7) by

$$1/\tau=A=\tfrac{1}{2}\int_0^{\nu_{21}}A(\nu_1)\,d\nu_1=\tfrac{1}{2}\int_0^1A(y)\,dy. \quad (8)$$

The factor of $\frac{1}{2}$ in Eq. (8) is needed because only pairs of photons are counted, whereas a given pair will occur twice in Eqs. (1) and (2) when ν_1 is varied through the range $0-\nu_{21}$.

III. EVALUATION OF MATRIX ELEMENTS

The $m\,^1S_0$ bound-state wave functions obtained by Pekeris[12,13] and the $n\,^1P_1$ bound-state wave functions obtained by Schiff, Lifson, Pekeris, and Rabinowitz[14] can be reexpressed in the forms

$$\psi_m(\vec{r}_1,\vec{r}_2)=\frac{(1+P_{12})}{\sqrt{2}}\,e^{-ar_1-br_2}\sum_{ijk}c_{ijk}r_1^ir_2^jr_{12}^k \quad (9)$$

and

$$\psi_n(\vec{r}_1,\vec{r}_2)=\frac{(1+P_{12})}{\sqrt{2}}\,e^{-ar_1-br_2}\,r_2\cos\theta_2$$

$$\times \sum_{ijk} d_{ijk}\, r_1^i r_2^j r_{12}^k, \quad (10)$$

where P_{12} indicates the interchange of the labels 1 and 2. In the $1\,^1S_0$ ground-state wave function,[12] $a=b=(-0.5E_1)^{1/2}$, where $-0.5E_1$ is the binding energy measured in atomic units. In the $2\,^1S_0$ and $n\,^1P_1$ wave functions,[14] $a=Z$, while b was optimized to obtain the lowest-energy eigenvalue for a given number of terms in the expansion. Employing these wave functions, Schiff, Pekeris, and Accad[6] have evaluated the matrix elements $\langle m\,^1S|z_1+z_2|n\,^1P\rangle$ and $\langle m\,^1S|\partial/\partial z_1+\partial/\partial z_2|n\,^1P\rangle$ (for $m=1, 5$ and $n=2, 5$). They obtained length and velocity values for the corresponding electric-dipole-transition oscillator strengths which are believed to be accurate to within 1% or better.

Higher discrete $n\,^1P_1$ states are expected to be less important in the evaluation of expressions (6) and (7). The oscillator strengths estimated by Dalgarno and Lynn[15] and by Dalgarno and Kingston[16] should be sufficiently accurate for the $n\,^1P_1$ states for $n=6, 10$. The contributions from the remaining discrete $n\,^1P_1$ states can be estimated by employing the asymptotic expressions[17,18]

$$\langle m\,^1S|z_1+z_2|n\,^1P\rangle \underset{n\to\infty}{\longrightarrow} (2/n^3)^{1/2}\langle m\,^1S|z_1+z_2|\psi_{k^2=0}\rangle \quad (11)$$

and

$$\left\langle m\,^1S\left|\frac{\partial}{\partial z_1}+\frac{\partial}{\partial z_2}\right|n\,^1P\right\rangle \underset{n\to\infty}{\longrightarrow}\left(\frac{2}{n^3}\right)^{1/2}\left\langle m\,^1S\left|\frac{\partial}{\partial z_1}+\frac{\partial}{\partial z_2}\right|\psi_{k^2=0}\right\rangle, \quad (12)$$

where $\psi_{k^2=0}$ denotes the $^1P_1$ continuum wave function evaluated at the first ionization threshold of the $m\,^1S_0$ states. The continuum wave function is assumed to be normalized per unit rydberg energy.

The techniques employed to obtain the continuum wave functions are described in the theory of electron scattering by hydrogenlike ions with nuclear charge Z. The ^1L continuum wave function (corresponding to nonresonant elastic scattering) can be expanded in the form

$$\psi_k{}^2(\vec{r}_1\vec{r}_2)=\frac{(1+P_{12})}{\sqrt{2}}\sum_{\mu=nl_1l_2}\frac{S_{nl_1}(r_1)}{r_1}\,\frac{F_\mu(r_2)}{r_2}\times\sum_{m_1m_2}\langle l_1l_2LM_L|l_1m_1l_2m_2\rangle Y_{l_1}^{m_1}(\hat{r}_1)\,Y_{l_2}^{m_2}(\hat{r}_2), \quad (13)$$

where the functions $S_{nl_1}(r_1)$ form a complete orthonormal basis and the functions $F_\mu(r_2)$ are to be determined.

The radial basis functions $S_{nl_1}(r_1)$ employed in this expansion are similar to the discrete basis functions introduced by Rotenberg[19] for elastic positron-hydrogen scattering. These functions can be constructed from the complete radial set $r^{n+l_1}e^{-Zr}$ by means of the Schmidt orthogonalization procedure. Their explicit form is given in terms of the associated Laguerre polynomials $L_{n+l+1}^{2l+2}(2Zr)$ by[20]

$$S_{nl}(r)=\left(\frac{(2Z)^3(n-l-1)!}{[(n+l+1)!]^3}\right)^{1/2}\times(2Zr)^l re^{-Zr}L_{n+l+1}^{2l+2}(2Zr). \quad (14)$$

This discrete basis includes the 1s eigenstate, thus ensuring that the expansion for ψ_{k^2} will satisfy the correct scattering boundary conditions. A discrete radial basis appropriate for inelastic scattering can be constructed by orthogonalizing the functions $r^{n+l_1}e^{-Zr}$ to the atomic eigenstates occurring in all open channels.[7] The use of a discrete basis for the functions $S_{nl_1}(r_1)$ circumvents the necessity of explicitly including continuum hydrogenlike eigenstates in order to achieve completeness.

When the continuum wave function is normalized to a Dirac δ function in the total rydberg energy E ($E=k^2-Z^2$), the asymptotic form of the function $F_{1s,l_2=L}(r_2)$ becomes[21]

$$F_{1s,l_2=L}(r_2)\underset{r_2\to\infty}{\longrightarrow}\frac{1}{(\pi k)^{1/2}}\sin\left[kr_2-\tfrac{1}{2}l_2\pi+\frac{(Z-1)}{k}\ln 2kr_2+\arg\Gamma\left(l_2+1-i\,\frac{(Z-1)}{k}\right)+\eta_L\right], \quad (15)$$

where η_L is the additional phase shift due to the non-Coulombic part of the potential. When $k^2=0$, this asymptotic form must be replaced by

$$F_{1s,l_2=L}(r_2)\underset{r_2\to\infty}{\longrightarrow}\left(\frac{r_2}{2\pi^2(Z-1)}\right)^{1/4}\times\sin\{[8(Z-1)r_2]^{1/2}-(l_2+\tfrac{1}{4})\pi+\eta_L\}. \quad (16)$$

The other functions $F_\mu(r_2)$, corresponding to closed channels in the elastic-scattering energy region, have asymptotic forms which represent exponentially decaying waves. In our calculations, the closed channels included in the expansion were $(\bar{2}s, l_2=1)$, $(\bar{2}p, l_2=2)$, and $(\bar{2}p, l_2=0)$. (The bar over the principal quantum number indicates the replacement of He^+ eigenstates by discrete basis functions.)

The computer code of Burke[22] was used to solve the coupled integrodifferential equations for the functions $F_\mu(r_2)$ in the energy region $0\le k^2\le 2.4$ Ry. An asymptotic expansion,[23] whose leading term has the form given by Eq. (15), is employed in Burke's code to obtain starting values of the functions $F_\mu(r_2)$ for inward integration of the coupled equations. When k^2 is very small, this asymp-

TABLE I. The P-wave e^--He^+ elastic-scattering phase shift η_1 and differential oscillator strengths for helium evaluated in the energy region $0 \le k^2 \le 0.2$ Ry, using the $1s$, $\bar{2}s$, $\bar{2}p$ expansion for the ^1P continuum wave function.

		$1\,^1S$ state		$2\,^1S$ state	
k^2 (Ry)	η_1	$\left(\frac{df}{dE}\right)_L$	$\left(\frac{df}{dE}\right)_V$	$\left(\frac{df}{dE}\right)_L$	$\left(\frac{df}{dE}\right)_V$
0	−0.0424	0.9218	0.9117	1.144	1.1384
0.02	−0.0429	0.9072	0.8971	1.026	1.0306
0.04	−0.0435	0.8926	0.8827	0.9263	0.9364
0.06	−0.0439	0.8783	0.8685	0.8408	0.8538
0.08	−0.0444	0.8642	0.8546	0.7671	0.7809
0.10	−0.0448	0.8502	0.8408	0.7030	0.7165
0.12	−0.0452	0.8365	0.8274	0.6466	0.6591
0.14	−0.0455	0.8230	0.8141	0.5967	0.6079
0.16	−0.0459	0.8097	0.8011	0.5523	0.5621
0.18	−0.0462	0.7966	0.7884	0.5125	0.5209
0.20	−0.0465	0.7838	0.7756	0.4768	0.4837

totic expansion for the functions $F_\mu(r_2)$ converges only for extremely large values of r_2. Norcross and Seaton[24] have developed an alternative asymptotic expansion for the functions $F_\mu(r_2)$ which is based on the WKB method and is appropriate for small values of k^2 (including $k^2 = 0$). The computer code of Norcross[25] was employed in our calculations to obtain starting values for the functions $F_\mu(r_2)$ in the energy region $0 \le k^2 \le 0.2$ Ry.

The matrix elements $\langle m\,^1S | z_1 + z_2 | \psi_k{}^2 \rangle$ and $\langle m\,^1S | \partial/\partial z_1 + \partial/\partial z_2 | \psi_k{}^2 \rangle$ (for $m = 1, 2$) were evaluated in the energy region $0 \le k^2 \le 2.4$ Ry, using the 56-term $m\,^1S_0$ bound-state wave functions of Pekeris and the $1s$, $\bar{2}s$, $\bar{2}p$ expansion for the $^1P_1$ continuum wave function. The "length" and "velocity" expressions for the differential oscillator strengths, which would yield identical values if they could be evaluated using exact eigenstates of the nonrelativistic Hamiltonian, are given in terms of the above matrix elements by

$$\left(\frac{df}{dE}\right)_L = (I + k^2) \left| \langle m\,^1S | z_1 + z_2 | \psi_k{}^2 \rangle \right|^2 \tag{17}$$

and

$$\left(\frac{df}{dE}\right)_V = \frac{4}{I + k^2} \left| \left\langle m\,^1S \left| \frac{\partial}{\partial z_1} + \frac{\partial}{\partial z_2} \right| \psi_k{}^2 \right\rangle \right|^2, \tag{18}$$

where I is the first ionization potential of the $m\,^1S_0$ state. Results are given in Table I for the P-wave phase shift and the differential oscillator

TABLE II. Various contributions to the sums in Eqs. (6) and (7) for representative values of y:

$$M_n(y) = \langle 1\,^1S | D_z | n\,^1P \rangle \langle n\,^1P | D_z | 2\,^1S \rangle \left(\frac{1}{E_n - E_2 + (E_2 - E_1)y} + \frac{1}{E_n - E_2 + (E_2 - E_1)(1-y)} \right).$$

Continuum contributions are denoted by $M_E(y)$. Numbers in parentheses are the powers of 10 by which the entries are to be multiplied.

y	$\sum_{n=2}^{5} M_n(y)$	$\sum_{n=2}^{\infty} M_n(y)$	$\int_{k^2=0}^{2.4} M_E(y)\,dE$	$\int_{k^2=2.4}^{\infty} M_E(y)\,dE$
		$D_z = z_1 + z_2$		
0.025	−1.433 (1)	1.20 (−1)	9.551 (−1)	2.67 (−3)
0.100	−6.119 (0)	1.03 (−1)	8.285 (−1)	2.64 (−3)
0.300	−2.892 (0)	7.40 (−2)	6.738 (−1)	2.57 (−3)
0.500	−2.450 (0)	6.77 (−2)	6.356 (−1)	2.54 (−3)
		$D_z = \frac{\partial}{\partial z_1} + \frac{\partial}{\partial z_2}$		
0.025	−1.510 (−1)	1.64 (−2)	3.084 (−1)	1.16 (−2)
0.100	−3.538 (−2)	1.30 (−2)	2.790 (−1)	1.15 (−2)
0.300	−2.892 (−3)	9.34 (−3)	2.406 (−1)	1.13 (−2)
0.500	+8.277 (−5)	8.52 (−3)	2.307 (−1)	1.12 (−2)

TABLE III. Two-photon decay energy distribution $A(y)$ (in $\sec^{-1}$) as a function of y, the fraction of the $2^1S_0 - 1^1S_0$ energy difference carried away by one of the two photons. The numbers in parentheses are the powers of 10 by which the entries are to be multiplied.

y	$A_L(y)$	$A_V(y)$
0.025	7.726 (0)	7.745 (0)
0.050	2.513 (1)	2.515 (1)
0.075	4.324 (1)	4.327 (1)
0.100	5.959 (1)	5.964 (1)
0.125	7.378 (1)	7.384 (1)
0.150	8.593 (1)	8.600 (1)
0.175	9.629 (1)	9.638 (1)
0.200	1.051 (2)	1.052 (2)
0.225	1.127 (2)	1.128 (2)
0.250	1.191 (2)	1.192 (2)
0.275	1.245 (2)	1.246 (2)
0.300	1.291 (2)	1.293 (2)
0.325	1.330 (2)	1.331 (2)
0.350	1.362 (2)	1.364 (2)
0.375	1.389 (2)	1.390 (2)
0.400	1.410 (2)	1.411 (2)
0.425	1.425 (2)	1.427 (2)
0.450	1.436 (2)	1.438 (2)
0.475	1.443 (2)	1.444 (2)
0.500	1.445 (2)	1.447 (2)

strengths in the energy region $0 \leq k^2 \leq 0.2$ Ry. Results were given in the previous paper[7] for the energy region $0.2 \leq k^2 \leq 2.4$ Ry. The photoionization cross sections are obtained (in units of a_0^2) by multiplying the differential oscillator strengths by $4\pi^2\alpha$.

The contributions to the sums in Eqs. (6) and (7) from the continuum states for $k^2 \geq 2.4$ Ry can be estimated with sufficient accuracy by employing the various independent-particle approximations used by Salpeter and Zaidi[26] and by Suh and Zaidi[27] in their calculations of the Lamb-shift mean excitation energies.

IV. RESULTS FOR TWO-PHOTON DECAY RATE

Various contributions to the sums in Eqs. (6) and (7) are presented in Table II for representative values of y. The continuum contributions are of relatively greater importance in the velocity expression. This is explained by the different energy dependence of the matrix elements implied by Eqs. (17) and (18). The two-photon decay energy distributions resulting from the evaluation of the two alternative expressions for $A(y)$ are tabulated in Table III, and they both differ from the distribution obtained by Drake, Victor, and Dalgarno[5] by less than 1%. The integrated decay rates obtained from these two distributions are $A_L = 50.85$ $\sec^{-1}$ and $A_V = 50.89$ $\sec^{-1}$, in good agreement with the value $A = 51.3$ $\sec^{-1}$ obtained by Drake, Victor, and Dalgarno.[5] It is very possible that the uncertainty in our two values for the decay rate is greater than the difference between them.

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