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Spin-forbidden radiative decay rates from the $3\,^3P_{1,2}$ and $3\,^1P_1$ states of helium

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We have calculated atomic helium spontaneous decay rates and absorption oscillator strengths for the spin-forbidden transitions from $3\,^3P_{1,2}$ and $3\,^1P_1$ to all lower $^1S_0$ and $^3S_1$ states. In particular we found $A_{10} = 44.33(4)\ \mathrm{s}^{-1}$ for the $E1$ transition $3\,^3P_1$–$1\,^1S_0$ and $0.1147(1)\ \mathrm{s}^{-1}$ for the $M2$ transition $3\,^3P_2$–$1\,^1S_0$.

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I. INTRODUCTION

The study of spin-forbidden transitions in helium and the heliumlike ions has a long history as a fundamental testing ground for the interactions between radiation and matter. In the limit of LS coupling, electric dipole ($E1$) transitions between singlet and triplet states are strictly forbidden in the lowest order due to spin orthogonality because the leading terms in the electric dipole transition operator are just $-e(\mathbf{r}_1 + \mathbf{r}_2)$, which is not spin dependent. However, relativistic spin-dependent corrections to the wave functions and retardation corrections to the transition operator both contribute nonvanishing terms of relative order $(\alpha Z)^2$, where Ze is the nuclear charge and α is the fine-structure constant.

The best-studied example is the astrophysically important $1s2p\,^3P_1$–$1s^2\,^1S_0$ transition of helium and the heliumlike ions, where the subscript is the eigenvalue of the total angular momentum operator $\mathbf{J} = \mathbf{L} + \mathbf{S}$. For this case, the intermixing of the $1s2p\,^3P_1$ and $1s2p\,^1P_1$ states is the dominant effect, but the contribution from all the higher-lying $1snp\,^3P_1$ states and final $1s^2\,^1S_0$ state perturbations due to mixing with doubly-excited P states of the form $npn'p\,^3P^e_1$ are also significant. Following the earlier work by Elton [1] and Drake and Dalgarno [2], Drake [3] performed a complete calculation, which included spin-dependent perturbations to both the initial and final states. This work also demonstrated the equivalence of the length and velocity forms of the transition operator, provided that an extra spin-dependent term is added to the velocity form of the transition operator, as further discussed in the present work. Also, the spin-dependent part of the magnetic quadrupole ($M2$) operator directly enables $1s2p\,^3P_2$–$1s^2\,^1S_0$ [4].

A general derivation of relativistic corrections to allowed and spin-forbidden electric dipole transitions was carried out from basic QED by Drake [5], including the effects of the electron-electron interaction and negative energy states. It was proven there that the nonrelativistic Pauli form of the Breit interaction remains valid for off-diagonal matrix elements in the presence of radiation emission, and that a semiclassical representation of the radiation field remains valid up to terms of relative order α^2Z^2. The former point is important because the Breit interaction is normally taken to be correct only as a first-order perturbation correction to the nonrelativistic energy [6]. These theoretical results were recently confirmed by Łach and Pachucki [7], and much more accurate numerical values for matrix elements were obtained by them, as well as by Morton, Moffatt, and Drake [8].

The purpose of the present work is to extend the results of Ref. [8] on the calculation of He I spin-forbidden A and f values for the electric dipole ($E1$) decays $3\,^3P_1$–$n\,^1S_0$ and $3\,^1P_1$–$n\,^3S_1$, as well as the magnetic quadrupole ($M2$) decays $3\,^3P_2$–$n\,^1S_0$ and $3\,^1P_1$–$n\,^3S_1$. Since Baldwin and his colleagues [9,10] are considering measuring $3\,^3P_1$–$1\,^1S_0$ and $3\,^3P_2$–$1\,^1S_0$, this theoretical study is timely. In emission, the spin-forbidden transitions are dominated by ordinary allowed $E1$ transitions to lower states, but in absorption, the spin-forbidden transitions can readily be observed.

II. CALCULATIONS

As outlined in the Introduction, if spin-orbit coupling is weak, spin-forbidden transitions can be calculated to order $(\alpha Z)^2$ relative to allowed $E1$ transitions from the perturbation of the nonrelativistic wave function $|nPSLJ\rangle$ by the Breit spin-orbit (SO) and spin-other-orbit (SOO) operators. The spin-spin operator does not contribute to $S = 1$ to $S = 0$ transitions because it is a scalar formed by coupling irreducible tensors of rank 2. The perturbed wave function can be expanded in terms of a complete set of virtual intermediate states. The spin-changing transitions come from the virtual states with opposite spin but the same parity P and angular momentum J. As described in [8], we approximate these intermediate states, including the continuum, by the complete set of N nonrelativistic pseudostates derived from the variational solution of the energy eigenvalue equations for a basis set with N terms.

Specifically, for the spin-changing transition $3\,^3P_1$–$1\,^1S_0$, we computed the sums

$$\begin{aligned}\langle 1\,^1S_0 \mid H_1 \mid 3\,^3P_1\rangle &= \sum_m^N \langle 1\,^1S_0 \mid H_1 \mid m\,^1P_1\rangle \frac{\langle m\,^1P_1 \mid B \mid 3\,^3P_1\rangle}{\epsilon(3\,^3P_1) - \epsilon(m\,^1P_1)} \\ &\quad + \sum_n^N \frac{\langle 1\,^1S_0|B|n\,^3P^e_0\rangle}{\epsilon(1\,^1S_0) - \epsilon(n\,^3P^e_0)} \langle n\,^3P^e_0|H_1|3\,^3P_1\rangle, \qquad (1)\end{aligned}$$

where $H_1 = z_1 + z_2$ is the interaction operator, B is the sum of the SO and SOO operators, and all the wave functions on the right are nonrelativistic. In the atomic units used here, lengths r and energies ϵ are related to laboratory values by $R = a_0 r$ and $E = \alpha^2 m_e c^2 \epsilon$, where a_0, α, m_e, and c are the usual atomic constants. The only states affecting $1\,^1S_0$ are the doubly-excited ones beginning with $2p^2\,^3P^e_0$ above the first ionization of helium. Similarly for $2\,^3S_1$, the intermediate

TABLE I. Matrix Elements M and f- and A-values for $3\,^3P_{1,2}$–$1\,^1S_0$, $2\,^1S_0$, and $3\,^1S_0$. The error on each entry indicates the numerical uncertainty in the convergence as the size of the basis sets is increased and does not include any estimate for the theoretical approximations.

$E1$ Transition	$3\,^3P_1$–$1\,^1S_0$	$3\,^3P_1$–$2\,^1S_0$	$3\,^3P_1$–$3\,^1S_0$
$\Delta\epsilon_\infty$ (a.u.) theory	0.845 643 292 8	0.087 892 961 78	$3.190\,905\,466 \times 10^{-3}$
$\Delta\epsilon$ (a.u.) actual	0.845 494 46	0.087 878 693 0	$3.188\,316 \times 10^{-3}$
λ (nm) actual	53.889 594	518.489 09	14 290.72
Length calculation[a]			
${}^1\langle n\,{}^1S_0 \vert z_1+z_2 \vert 3\,{}^3P_1\rangle^0$	$-0.008\,24(1) \times 10^{-4}$	$-0.000\,523(3) \times 10^{-4}$	$-0.000\,043\,9(2) \times 10^{-3}$
${}^0\langle n\,{}^1S_0 \vert z_1+z_2 \vert 3\,{}^3P_1\rangle^1$	$1.021\,445(3) \times 10^{-4}$	$5.487\,346(3) \times 10^{-4}$	$3.100\,340(1) \times 10^{-3}$
Total M_L	$1.013\,20(1) \times 10^{-4}$	$5.486\,823(4) \times 10^{-4}$	$3.100\,296(1) \times 10^{-3}$
Velocity calculation[a]			
${}^1\langle n\,{}^1S_0 \vert \partial/\partial z_1+\partial/\partial z_2 \vert 3\,{}^3P_1\rangle^0/\Delta\epsilon_\infty$	$0.021\,72(1) \times 10^{-4}$	$0.018\,80(4) \times 10^{-4}$	$0.021\,93(9) \times 10^{-3}$
${}^0\langle n\,{}^1S_0 \vert \partial/\partial z_1+\partial/\partial z_2 \vert 3\,{}^3P_1\rangle^1/\Delta\epsilon_\infty$	$1.140\,782\,9(1) \times 10^{-4}$	$5.501\,376\,4(3) \times 10^{-4}$	$3.272445(1) \times 10^{-3}$
Correction $C/\Delta\epsilon_\infty$	$-0.149\,283\,75(1) \times 10^{-4}$	$-0.033\,312\,00(5) \times 10^{-4}$	$-0.194\,178\,64(3) \times 10^{-3}$
Total $M_V/\Delta\epsilon_\infty$	$1.013\,22(1) \times 10^{-4}$	$5.486\,86(4) \times 10^{-4}$	$3.100\,20(9) \times 10^{-3}$
$E1$ f_{01}	$5.787\,6(2) \times 10^{-9}$	$1.764025(3) \times 10^{-8}$	$2.044\,697(1) \times 10^{-8}$
$E1$ A_{10} (s^{-1})	44.326(1)	1.459 495(2)	$2.229\,700(1) \times 10^{-3}$
$M2$ Transition	$3\,^3P_2$–$1\,^1S_0$	$3\,^3P_2$–$2\,^1S_0$	$3\,^3P_2$–$3\,^1S_0$
$\Delta\epsilon_\infty$ (a.u.) theory	0.845 643 292 8	0.087 892 961 78	$3.190\,905\,466 \times 10^{-3}$
$\Delta\epsilon$ (a.u.) actual	0.845 494 36	0.087 878 592 9	$3.188\,216 \times 10^{-3}$
λ (nm) actual	53.889 600	518.480 07	14 291.17
$\langle n\,{}^1S_0 \mid z_1-z_2 \mid 3\,{}^3P_2\rangle$	0.417 847 2(2)	2.142 383(1)	12.013 381 85(1)
$M2$ f_{02}	$2.495035(3) \times 10^{-11}$	$7.364\,387(7) \times 10^{-13}$	$1.108\,032\,35(1) \times 10^{-15}$
$M2$ A_{20} (s^{-1})[b]	$0.114\,654\,7(1)$	$3.655\,827(4) \times 10^{-5}$	$7.249\,719\,30(6) \times 10^{-11}$

[a]The superscripts 1 and 0 outside the angle brackets denote the perturbation order of the adjacent wave functions.
[b]Jacobs *et al.* [13] had calculated 0.115 s^{-1} and Kundu *et al.* [14] 0.120 63(1) s^{-1} for $3\,^3P_2$–$1\,^1S_0$.

states begin with $2p3p\,{}^1P_1^e$, while for $1s2p\,{}^1P_1$, they are the odd states $1s2p\,{}^3P_1$ and higher. For these calculations we have accurate nonrelativistic energies and wave functions for infinite nuclear mass with successively larger basis sets up to 1000 terms to assess convergence. As a test of this perturbation procedure in Ref. [8], we obtained $A_{10} = 177.578\ s^{-1}$ for

TABLE II. Matrix Elements M and f- and A-values for $3\,^1P_1$–$2\,^3S_1$ and $3\,^3S_1$. The error on each entry indicates the numerical uncertainty in the convergence as the size of the basis sets is increased and does not include any estimate for the theoretical approximations.

$E1$ Transition	$3\,^1P_1$–$2\,^3S_1$	$3\,^1P_1$–$3\,^3S_1$
$\Delta\epsilon_\infty$ (a.u.) theory	0.120 083 016 14	$1.354\,270\,538 \times 10^{-2}$
$\Delta\epsilon$ (a.u.) actual	0.120 074 88	$1.354\,404\,0 \times 10^{-2}$
λ (nm) actual	379.457 81	3364.088 7
Length calculation[a]		
${}^1\langle n\,{}^3S_1 \vert z_1+z_2 \vert 3\,{}^1P_1\rangle^0$	–0.001 649 3(5) $\times 10^{-4}$	$0.000\,130\,64(4) \times 10^{-3}$
${}^0\langle n\,{}^3S_1 \vert z_1+z_2 \vert 3\,{}^1P_1\rangle^1$	$1.213\,415\,8(1) \times 10^{-4}$	$2.898\,049\,27(4) \times 10^{-3}$
Total M_L	$1.211\,766\,5(5) \times 10^{-4}$	$2.898\,179\,91(6) \times 10^{-3}$
Velocity calculation[a]		
${}^1\langle n\,{}^3S_1 \vert \partial/\partial z_1+\partial/\partial z_2 \vert 3\,{}^1P_1\rangle^0/\Delta\epsilon_\infty$	$0.037\,517(5) \times 10^{-4}$	–0.024 429(4) $\times 10^{-3}$
${}^0\langle n\,{}^3S_1 \vert \partial/\partial z_1+\partial/\partial z_2 \vert 3\,{}^1P_1\rangle^1/\Delta\epsilon_\infty$	$1.368\,098\,3(1) \times 10^{-4}$	$2.803\,014(4) \times 10^{-3}$
Correction $C/\Delta\epsilon_\infty$	–0.193 852 94(1) $\times 10^{-4}$	$0.119\,599\,95(1) \times 10^{-3}$
Total $M_V/\Delta\epsilon_\infty$	$1.211\,762(5) \times 10^{-4}$	$2.898\,185(6) \times 10^{-3}$
E1 f_{01}	$3.918\,384(4) \times 10^{-10}$	$2.527\,805\,2(1) \times 10^{-8}$
E1 A_{10} (s^{-1})	0.181 543 6(2)	0.148 958 52(1)
$M2$ Transition	$3\,^1P_1$–$2\,^3S_1$	$3\,^1P_1$–$3\,^3S_1$
$\Delta\epsilon_\infty$ (a.u.) theory	0.120 083 016 14	$1.354\,270\,538 \times 10^{-2}$
$\Delta\epsilon$ (a.u.) actual	0.120 074 88	$1.354\,404\,0 \times 10^{-2}$
λ (nm) actual	379.457 81	3364.088 7
$\langle n\,{}^3S_1 \mid z_1-z_2 \mid 3\,{}^1P_1\rangle$	0.443 407 2(1)	11.258 337 6(1)
$M2$ f_{01}	$2.681\,693(1) \times 10^{-14}$	$2.479\,839\,86(4) \times 10^{-14}$
$M2$ A_{10} (s^{-1})	$1.242\,462(1) \times 10^{-5}$	$1.461\,320\,16(2) \times 10^{-7}$

[a]The superscripts 1 and 0 outside the angle brackets denote the perturbation order of the adjacent wave functions.

$2\,^3P_1$–$1\,^1S_0$ compared with 177.5771 s^{-1} calculated by Łach and Pachucki [7] and $177 \pm 8\ s^{-1}$ measured by Dall *et al*. [11]. We also found $A_{11} = 1.548\,935\ s^{-1}$ for $2\,^1P_1$–$2\,^3S_1$, very close to $1.548\,926\ s^{-1}$ in [7].

This calculation uses the dipole length operator $z_1 + z_2$. Within our $(\alpha Z)^2$ approximation, Drake [3] has shown that a similar one with the dipole velocity operator $\partial/\partial z_1 + \partial/\partial z_2$ in Eq. (1), plus the correction

$$C = \frac{\sqrt{2}\alpha^2}{4}\langle 1\,^1S_0|\frac{Z}{r_1^3}z_1 - \frac{Z}{r_2^3}z_2 + \frac{2}{r_{12}^3}(z_1 - z_2)|3\,^3P_1\rangle, \quad (2)$$

should be equivalent to the length result multiplied by the energy difference. Note that the correction depends on only the initial and final nonrelativistic states and also applies to $3\,^1P_1$–$2\,^3S_1$.

The $M2$ calculation also uses these states. The analysis of Drake [4] with the energy difference $\Delta\varepsilon$ and the reduced matrix element in atomic units gives the statistical weight times the transition rate in s^{-1} as

$$g_2 A_{20} = \frac{\alpha^8 c}{6a_0}\Delta\epsilon^5\ |\ \langle 1\,^1S_0\ ||\ z_1 - z_2\ ||\ 3\,^3P_2\rangle\ |^2, \quad (3)$$

and similarly with the same factors for $g_1 A_{11}$ for the $M2$ part of $^1P_1$–$^3S_1$. For $2\,^3P_2$–$1\,^1S_0$, we found $A_{20} = 0.327\,031\,5\ s^{-1}$ compared with $0.327\ 032\ 6\ s^{-1}$ in Ref. [7] and $0.324 \pm 0.016\ s^{-1}$ measured by Hodgman *et al.* [9].

III. RESULTS AND DISCUSSION

Tables I and II summarize our results. We calculated the oscillator strengths and transition rates using the theoretical energy $\Delta\epsilon_\infty$ for infinite nuclear mass without singlet-triplet mixing. The actual energies and wavelengths are from the combination of laboratory measurements and theoretical calculations presented by Morton, Wu, and Drake [12]. Use of these energies would give f and A values slightly closer to the true ones.

The subscripts on the $E1$ matrix elements M label the length (L) and velocity (V) forms from Eq. (1), and C is the correction of Eq. (2) with division by $\Delta\epsilon_\infty$ to match the length forms. In all cases, the dominant contribution for the spin-forbidden $E1$ transitions comes from the perturbed $3\,^3P_1$ or $3\,^1P_1$ state because of the small $\epsilon(3\,^3P_1) - \epsilon(3\,^1P_1)$ term in the denominator of Eq. (1). In fact, this one term dominates the sum over intermediate states. This dominance of the $m = 3$ term in the summation becomes even more pronounced with increasing Z because the singlet-triplet energy difference increases only in proportion to Z.

The comparison of the L and V forms of the transition operator is particularly interesting. Even though the individual contributions are very different, they sum to the same final matrix element to within 3 parts in 10^5 or better, and they agree to within the estimated convergence accuracy. This provides a very useful check on the accuracy of the results, analogous to the corresponding comparison of L and V for ordinary allowed transitions.

Even though both the spin-forbidden $E1$ and $M2$ decay rates are of the same nominal order $\alpha^8 c/a_0$, in every case the $M2$ decay rate is very much smaller.

The final uncertainties must also include the finite nuclear mass and other higher-order relativistic corrections. These could be as large as 0.1%, so realistic estimates are $A_{10} = 44.33(4)\ s^{-1}$ and $A_{20} = 0.1147(1)\ s^{-1}$ for $E1$ and $M2$, respectively, of $3\,^3P_{1,2}$–$1\,^1S_0$, and likewise for all the other results in the tables. Since these final uncertainties are likely to be much less than the corresponding experimental ones in the planned experiments [9,10], the comparison will provide an accurate benchmark to test experimental procedures to determine atomic lifetimes in this regime of strongly forbidden transitions.

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