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$2p^2\ ^3P$ and $2p3p\ ^1P$ States of the Helium Isoelectronic Sequence*

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Accurate eigenvalues and radiative lifetimes are obtained for the bound $2p^2\ ^3P$ and $2p3p\ ^1P$ states of the heliumlike ions He I to Ne IX. A high-order Z -expansion perturbation procedure is used that does not require the explicit solution of perturbation equations. The results are compared with variational calculations. The predicted wavelengths of the $1s2p\ ^3P$ - $2p^2\ ^3P$ and $1s3p\ ^1P$ - $2p3p\ ^1P$ transitions in helium are, respectively, 320.31 and 308.97 Å. Lines have been observed in helium at 320.39 and 309.04 Å.

I. INTRODUCTION

Because of the conservation of angular momentum and parity, the $2p^2\ ^3P$ and $2p3p\ ^1P$ states of helium and heliumlike ions, although embedded in the continuum, are stable against autoionization within the LS coupling approximation. Their lifetimes depend on the probability of radiative transitions to lower states and are much longer than the lifetimes characteristic of autoionizing states. Becker and Dahler¹ have pointed out the importance of such highly energetic states as initiators of reactions involved in radiation chemistry and in the chemistry of high-temperature gases.

The $2p^2\ ^3P$ state of helium has been produced through electron impact by Burrow and Schulz² and identified by the trapped-electron method. This state should also be observable in the electron energy-loss spectrum, although it has not yet been identified. The cross sections for the excitation of the $2p^2\ ^3P$ and $2p3p\ ^1P$ states by electron impact from the ground state have been calculated by Becker and Dahler,¹ along with several other doubly excited states. The $2p3p\ ^1P$ state will be more difficult to observe in electron-impact experiments because its excitation cross section is much smaller than the $2p^2\ ^3P$ cross section. A

radiative transition at 320.39 Å, observed by Kruger³ and included in Martin's⁴ tables, has been tentatively identified as the $2p^2\ ^3P$ - $1s2p\ ^3P$ transition.

The helium $2p^2\ ^3P$ eigenvalue has been calculated by Holóien⁵ using a configuration-interaction wave function. Several other less accurate calculations are cited by Becker and Dahler.¹ In this paper we report precise variational upper bounds for the $2p^2\ ^3P$ and $2p3p\ ^1P$ helium eigenvalues and oscillator strengths for radiative transitions to the $1s2p$, $1s3p$, and $1s4p\ ^3P$ and 1P states, respectively. The results are extended to the heliumlike ions by a high-order Z -expansion technique. We also calculate the sum of all oscillator strengths from the $1s2p\ ^1,^3P$ state to the $npn'p\ ^1,^3P$ states.

II. THEORY

The eigenvalue problem to be solved is (in Z^2 a. u.)

$$H\Psi_s = (H_0 + Z^{-1}V)\Psi_s = E_s\Psi_s, \quad (1)$$

where H_0 is a sum of two hydrogen-atom Hamiltonians, V is equal to $1/r_{12}$ and r_{12} and is the inter-electronic coordinate. A variational approximation to E_s and Ψ_s is obtained from the stationary values

of the functional

$$\tilde{E}_s = \langle \tilde{\Psi}_s | H | \tilde{\Psi}_s \rangle / \langle \tilde{\Psi}_s | \tilde{\Psi}_s \rangle \quad (2)$$

for arbitrary variations of $\tilde{\Psi}_s$. Because the $2p^2\ ^3P$ and $2p3p\ ^1P$ states are the lowest-lying triplet and singlet P states of even parity, the application of the variational method is straightforward.

If we expand $\tilde{\Psi}_s$ in a correlated orthonormal M -dimensional basis set

$$\tilde{\Psi}_s = \sum_{i=1}^M a_i \phi_i, \quad (3)$$

and vary the coefficients a_i , M roots are obtained, of which one may be further optimized through the choice of the ϕ_i to represent a particular state.

The remaining $M-1$ solutions satisfy oscillator-strength sum rules⁶ if the ϕ_i are well chosen and may be used to evaluate the summations occurring in formal perturbation theory.^{7,8} The calculations must be repeated for each value of the nuclear charge.

Alternatively, we may expand $\tilde{\Psi}_s$ and \tilde{E}_s in the forms

$$\tilde{\Psi}_s = \sum_{n=0}^{\infty} Z^{-n} \psi_n \quad (4)$$

$$\text{and } \tilde{E}_s = \sum_{n=0}^{\infty} Z^{-n} E_n, \quad (5)$$

where, from Eq. (1),

$$(H_0 - E_0)\psi_0 = 0, \quad (6)$$

$$(H_0 - E_0)\psi_n + V\psi_{n-1} = \sum_{i=1}^n E_i \psi_{n-i}. \quad (7)$$

Substituting Eqs. (4) and (5) into Eq. (2), we obtain the n th-order functional

$$\begin{aligned} \tilde{E}_n = & \langle \psi_n | H_0 - E_0 | \psi_n \rangle + 2 \langle \psi_n | V | \psi_{n-1} \rangle \\ & - \sum_{i=1}^{n-1} E_i \sum_{j=0}^i \langle \psi_j | \psi_{n-i-j} \rangle. \end{aligned} \quad (8)$$

Scherr and Knight⁹ and Sanders and Scherr¹⁰ have obtained perturbed eigenvalues and eigenfunctions directly from the extrema of (8) by substituting trial forms $\tilde{\psi}_n$ for the ψ_n .

A simpler procedure, discussed by Dalgarno and Drake,¹¹ is obtained by introducing a set of M functions ϕ_i , one of which, say ϕ_k , is the exact hydrogenic solution ψ_0 with eigenvalue E_0 . A unitary transformation is then applied such that

$$\langle \phi_i | H_0 | \phi_j \rangle = \epsilon_i \delta_{ij}, \quad i, j = 1, 2, \dots, M, \quad (9)$$

$$\langle \phi_i | \phi_j \rangle = \delta_{ij}, \quad (10)$$

and $\epsilon_k = E_0$. If we expand the perturbed-trial wave function in the basis set ϕ_i

$$\psi_n = \sum_{i=1}^M a_i^{(n)} \phi_i \quad (11)$$

and evaluate E_n , demanding that its value be stationary with respect to arbitrary variations of the

$a_i^{(n)}$, then

$$\begin{aligned} \psi_n = & \sum_{i \neq k}^M \frac{\langle \psi_{n-1} | V | \phi_i \rangle \langle \phi_i |}{\epsilon_k - \epsilon_i} \\ & - \sum_{i \neq k}^M \sum_{p=1}^n \frac{E_p \langle \psi_{n-p} | \phi_i \rangle \langle \phi_i |}{\epsilon_k - \epsilon_i} \end{aligned} \quad (12)$$

$$\text{and } E_n = \langle \psi_{n-1} | V | \psi_0 \rangle - \sum_{p=1}^{n-1} E_p \langle \Psi_{n-p} | \psi_0 \rangle. \quad (13)$$

Exactly the same results are obtained from (7) by writing the resolvent operator in the form

$$(E_0 - H_0)^{-1} = \sum_{i \neq k}^M \frac{|\phi_i\rangle \langle \phi_i|}{\epsilon_k - \epsilon_i}. \quad (14)$$

Thus, (14) is the best variational approximation to the resolvent operator within a finite basis set. It is not necessary to solve explicitly each of the perturbation equations. Once the two diagonalizations (9) and (10) have been performed to determine the basis set ϕ_i , the perturbed eigenfunctions and energies are easily generated from the recursion relations (12) and (13). The results are comparable in accuracy with the explicit solutions of Sanders and Scherr⁹ using the same number of basis functions.

III. CALCULATIONS

Variational basis sets were constructed from sets of correlated functions of the form

$$\begin{aligned} \chi_{i,j,k}^{LS} = & (1 \pm P_{12}) r_1^i r_2^j r_{12}^k \\ & \times \exp(-\alpha_{LS} r_1 - \beta_{LS} r_2) \mathcal{Y}_{L_1 L_2}^{M L}(\hat{r}_1, \hat{r}_2), \end{aligned} \quad (15)$$

where $\mathcal{Y}_{L_1 L_2}^{M L}(\hat{r}_1, \hat{r}_2)$

$$= \sum_{m_1, m_2} \langle l_1 m_1 l_2 m_2 | L M_L \rangle Y_{l_1}^{m_1}(\hat{r}_1) Y_{l_2}^{m_2}(\hat{r}_2)$$

in the conventional vector-coupling notation of Edmonds,¹² and P_{12} indicates the interchange of electrons 1 and 2, with the plus sign referring to singlet states and the minus sign to triplet states. For the variational calculations involving the complete Hamiltonian H , α and β were chosen to minimize the lowest secular root, thus giving a variational bound to the $2p^2\ ^3P$ and $2p3p\ ^1P$ energies. For the Z -expansion basis sets defined by Eqs. (9) and (10), α and β are specified by their hydrogenic

values. In both calculations, the $\chi_{i,j,k}^{LS}$ basis set was first orthonormalized and then H (or H_0) diagonalized. Up to 50 terms were retained in the basis sets with the powers i, j , and k chosen such that $i, j \geq 1, k \geq 0$ and $i+j+k \leq N_{\max}$.

IV. VARIATIONAL RESULTS

A. Eigenvalues

The optimum values of the scale factors (α_{LS}, β_{LS}), defined by Eq. (15), were found to be (0.475, 0.40) and (0.45, 0.25), respectively, for the $2p^2\ ^3P$ and $2p3p\ ^1P$ states of neutral helium, with the unit of length being $Z^{-1}a_0$. The nonrelativistic eigenvalues obtained from various basis sets are given in Table I. The $2p^2\ ^3P$ eigenvalue $-1.420\ 999$ Ry may be compared with $-1.420\ 21$ Ry calculated by Holóien⁵ using a configuration-interaction wave function. The present eigenvalue lies 59.671 eV above the ground state and predicts that the $1s2p\ ^3P - 2p^2\ ^3P$ wavelength is 320.31 Å. Kruger³ and Compton and Boyce³ observed a line at 320.39 Å. The $2p3p\ ^1P$ eigenvalue is $-1.160\ 493$ Ry and lies 63.215 eV above the ground state. The predicted $1s3p\ ^1P - 2p3p\ ^1P$ wavelength is 308.97 Å. Compton and Boyce³ observed a line at 309.04 Å.

B. Oscillator Strengths

The oscillator strength connecting two M_L magnetic sublevels through the z component of the radiation field is defined in the length formulation by¹³

$$f_z(i-j) = 2\omega_{i,j} |\langle i | z_1 + z_2 | j \rangle|^2, \quad (16)$$

and in the velocity formulation by

$$f_z(i-j) = (2/\omega_{i,j}) |\langle i | p_{1,z} + p_{2,z} | j \rangle|^2, \quad (17)$$

where $p_{1,z}$ is the z component of the momentum operator. It follows from the properties of the Clebsch-Gordan coefficients that $f_z(^3P_1 - ^3P_1) = f_z(^3P_{-1} - ^3P_{-1})$ and $f_z(^3P_0 - ^3P_0) = 0$, where the subscripts denote the values of M_L . Thus, the averaged absorption oscillator strength is

$$f_{\text{av}}(^3P - ^3P) = \frac{2}{3} f_z(^3P_1 - ^3P_1). \quad (18)$$

The $^3P - ^3D$ and $^3P - ^3S$ averaged oscillator

TABLE I. $2p^2\ ^3P$ and $2p3p\ ^1P$ eigenvalues.

No. of terms	Eigenvalue (Ry)	
	$2p^2\ ^3P$	$2p3p\ ^1P$
20	-1.420 995	-1.160 492
30	-1.420 999	-1.160 493
40	-1.420 999	-1.160 493
50	...	-1.160 493

TABLE II. Oscillator strengths obtained from variational wave functions.

No. of terms	M	20	30	40
$f_{\text{av}}(1s2p\ ^3P - 2p^2\ ^3P)$		0.1778	0.1779	0.1810
$f_{\text{av}}(1s2p\ ^1P - 2p3p\ ^1P)$		0.0838	0.0811	0.0830

strengths are similarly

$$f_{\text{av}}(^3P - ^3D) = \frac{5}{6} f_z(^3P_0 - ^3D_0) \quad (19)$$

$$\text{and } f_{\text{av}}(^3P - ^3S) = \frac{1}{3} f_z(^3P_0 - ^3S_0). \quad (20)$$

The values of $f_{\text{av}}(1s2p\ ^3P - 2p^2\ ^3P)$ and $f_{\text{av}}(1s2p\ ^1P - 2p3p\ ^1P)$, calculated in the length formulation, are given in Table II.

If we define oscillator-strength sums S_z out of the 2^3P state

$$S_z(^3P_1) = \sum_{n=2}^{\infty} f_z(2^3P_1 - n^3P_1), \quad (21)$$

where \sum denotes summation over discrete states and integration over continuum states, and similarly for $S_z(^3S_0)$ and $S_z(^3D_0)$, then it follows from the Thomas-Reiche-Kuhn oscillator-strength sum rule¹³ that

$$S_z(^3S_0) + S_z(^3D_0) = 2 \quad (22)$$

from the z component of the radiation field and

$$\frac{1}{3} S_z(^3S_0) + \frac{5}{6} S_z(^3D_0) + \frac{2}{3} S_z(^3P_1) = 2 \quad (23)$$

from the average of all three components. Thus, only one of the three S_z summations is independent, the other two being determined by Eqs. (22) and (23). An analogous set of equations may be written for the singlet states.

The oscillator strength sums S_z were evaluated by replacing the infinite sets of excited state S, P , and D wave functions (including the continuum) by the variationally determined discrete sets described at the beginning of Sec. II. The results are given in Table III. There is some uncertainty in the final figures quoted due to incomplete convergence of the variational sets. The oscillator strength sum rules given by (21) and (22) are well satisfied.

V. Z-EXPANSION RESULTS

A. Eigenvalues

The leading several terms in the Z expansions of the wave functions were calculated by the recursion procedure described at the end of Sec. II. The coefficients in the eigenvalue expansions of the $2p^2\ ^3P$ and $2p3p\ ^1P$ states are given in Table IV. The eigenvalues summed through ninth order for

TABLE III. Oscillator-strength sums from the $1s2p$ 1P and 3P states.

	Singlet	Triplet
$S_z(S_0)$	-0.043	0.086
$S_z(D_0)$	2.040	1.909
$S_z(P_1)$	0.472	0.560
Sum, Eq. (22)	1.997	1.995
Sum, Eq. (23)	1.997	1.993

$Z = 2$ are -1.420999 and -1.160490 Ry for the $2p^2\ ^3P$ and $2p3p\ ^1P$ states, respectively, while the variational eigenvalues from Table I are -1.420999 and -1.160493 Ry. The good agreement indicates that the individual perturbation coefficients are accurate and that satisfactory convergence is achieved by summing through ninth order, even for $Z = 2$.

The $2p^2\ ^3P$ and $2p3p\ ^1P$ eigenvalues summed through ninth order for H^- are -0.2496 and -0.2471 Ry, respectively, while a graphical extrapolation to infinite order of the partial eigenvalue sums yielded -0.2510 Ry for both states. The extrapolated eigenvalue for the $2p^2\ ^3P$ state is in agreement with -0.2507 Ry calculated variationally by Drake¹⁴ and indicates that the state is bound below the $n = 2$ threshold of H , as suggested by Holþien.⁵ The $2p3p\ ^1P$ state is not known to be bound and one would expect the extrapolated eigenvalue to be -0.2500 Ry. The above sums provide a sensitive check on the accuracy of the calculations.

B. Oscillator Strengths

The Z expansions of the velocity-matrix elements $\langle i | p_{1,z} + p_{2,z} | j \rangle$ were calculated directly from the wave function expansions and the coefficients in the length-matrix elements $\langle i | z_1 + z_2 | j \rangle$ were calculated using the Z expansion of $\omega_{i,j}$ and the relationship

$$\langle i | z_1 + z_2 | j \rangle = (1/\omega_{i,j}) \langle i | p_{1,z} + p_{2,z} | j \rangle, \quad (24)$$

TABLE V. Transition-integral expansions (a.u.).^a

Order	$1s2p\ ^3P-2p^2\ ^3P$	$1s3p\ ^3P-2p^2\ ^3P$	$1s4p\ ^3P-2p^2\ ^3P$	$1s2p\ ^1P-2p3p\ ^1P$	$1s3p\ ^1P-2p3p\ ^1P$	$1s4p\ ^1P-2p3p\ ^1P$
0	0.7449	0	0	-0.2109	0.5267	0
1	-0.1187	-0.253	0.117	-0.3171	-0.1804	0.337
2	-0.1469	-0.091	0.057	-0.1570	-0.3822	-0.137
3	-0.1283	-0.032	0.054	0.0394	-0.1565	-0.261
4	-0.1315	0.020	0.041	0.0561	0.0253	-0.072
5	-0.1385	0.079	-0.010	-0.0348	0.0263	0.076
6	-0.1348	0.140	-0.054	-0.0512	-0.0927	...
7	-0.1165	0.136	...	0.0660	-0.0712	...
8	-0.0843	0.094	-0.0799	...
9	-0.0423	0.073

^a $\langle i | z_1 + z_2 | j \rangle = R_0 Z^{-1} + R_1 Z^{-2} + \dots + R_9 Z^{-10} a_0$.

TABLE IV. Eigenvalue expansions (Ry).^a

Order	$2p^2\ ^3P$	$2p3p\ ^1P$
0	-0.5000000	-0.3611111
1	0.3281250	0.1624776
2	-0.078789	-0.038436
3	0.003677	-0.002314
4	-0.000676	-0.002484
5	-0.000688	-0.001684
6	-0.000454	-0.001184
7	-0.000336	-0.000886
8	-0.000245	-0.000686
9	-0.000189	-0.000542

^a $E = Z^2 E_0 + Z E_1 + \dots + Z^{-7} E_9$ Ry.

a procedure that is apparently more accurate than calculating the coefficients directly in the length formulation. The coefficients in the Z expansions of $\langle i | z_1 + z_2 | j \rangle$ are given in Table IV for several transitions involving the $2p^2\ ^3P$ and $2p3p\ ^1P$ states. The leading terms vanish for those transitions involving two-electron jumps. The transition energies, absorption oscillator strengths, and emission Einstein A coefficients are given in Tables V, VI, and VII for the ions through Ne ix. In each case, the results obtained from the length and velocity formulations agree to at least the number of figures given. The Z -expansion oscillator strengths for the $1s2p\ ^3P - 2p^2\ ^3P$ and $1s2p\ ^1P - 2p3p\ ^1P$ transitions of neutral helium are in excellent agreement with the direct variational calculations of Table II. Transitions to lower states not included in Tables VI and VII are relatively improbable.

VI. DISCUSSION

In addition to obtaining accurate eigenvalues and radiative lifetimes of the $2p^2\ ^3P$ and $2p3p\ ^1P$ states of the heliumlike ions, we have shown that the results are obtainable for the entire isoelectronic sequence in a single calculation. The application of the perturbation recursion relations (12) and

TABLE VI. Z -Expansion transition energies, oscillator strengths, and Einstein A coefficients. Superscripts indicate the powers of 10 by which the entries are to be multiplied.

Ion	$1s2p\ ^3P - 2p^2\ ^3P$			$1s3p\ ^3P - 2p^2\ ^3P$			$1s4p\ ^3P - 2p^2\ ^3P$		
	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$
He	2.845 36	0.1804	1.173 ¹⁰	2.695 17	9.54 ⁻³	5.57 ⁸	2.6435	2.88 ⁻³	1.62 ⁸
Li ⁺	6.462 13	0.2225	7.465 ¹⁰	5.867 63	3.90 ⁻³	1.08 ⁹	5.6635	9.55 ⁻⁴	2.46 ⁸
Be ⁺⁺	11.584 52	0.2388	2.574 ¹¹	10.263 77	2.05 ⁻³	1.73 ⁹	9.8092	4.66 ⁻⁴	3.60 ⁸
B ³⁺	18.208 81	0.2476	6.593 ¹¹	15.882 65	1.25 ⁻³	2.54 ⁹	15.0801	2.75 ⁻⁴	5.03 ⁸
C ⁴⁺	26.333 96	0.2530	1.410 ¹²	22.724 00	8.44 ⁻⁴	3.50 ⁹	21.4761	1.81 ⁻⁴	6.72 ⁸
N ⁵⁺	35.959 60	0.2568	2.668 ¹²	30.787 68	6.07 ⁻⁴	4.62 ⁹	28.9972	1.28 ⁻⁴	8.67 ⁸
O ⁶⁺	47.085 50	0.2596	4.623 ¹²	40.073 68	4.57 ⁻⁴	5.89 ⁹	37.6434	9.57 ⁻⁵	1.09 ⁹
F ⁷⁺	59.711 58	0.2617	7.495 ¹²	50.581 94	3.56 ⁻⁴	7.33 ⁹	47.4146	7.40 ⁻⁵	1.34 ⁹
Ne ⁸⁺	73.837 80	0.2634	1.153 ¹³	62.312 46	2.86 ⁻⁴	8.91 ⁹	58.3109	5.90 ⁻⁵	1.61 ⁹

TABLE VII. Z -Expansion transition energies, oscillator strengths, and Einstein A coefficients. Superscripts indicate the powers of 10 by which the entries are to be multiplied.

Ion	$1s2p\ ^1P - 2p3p\ ^1P$			$1s3p\ ^1P - 2p3p\ ^1P$			$1s4p\ ^1P - 2p3p\ ^1P$		
	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$	ΔE (Ry)	$f(i-j)$	$A(j-i)$ sec $^{-1}$
He	3.087 18	0.0830	6.35 ⁹	2.949 82	0.0508	3.55 ⁹	2.9018	5.1 ⁻³	3.5 ⁸
Li ⁺	7.183 88	0.0587	2.43 ¹⁰	6.637 58	0.0862	3.05 ¹⁰	6.4455	3.62 ⁻³	1.21 ⁹
Be ⁺⁺	13.053 55	0.0487	6.67 ¹⁰	11.823 91	0.1022	1.15 ¹¹	11.3911	2.42 ⁻³	2.52 ⁹
B ³⁺	20.699 00	0.0434	1.49 ¹¹	18.509 81	0.1110	3.05 ¹¹	17.7393	1.69 ⁻³	4.26 ⁹
C ⁴⁺	30.121 24	0.0400	2.92 ¹¹	26.695 54	0.1165	6.67 ¹¹	25.4902	1.23 ⁻³	6.43 ⁹
N ⁵⁺	41.320 68	0.0378	5.18 ¹¹	36.381 20	0.1202	1.28 ¹²	34.6438	9.34 ⁻⁴	9.01 ⁹
O ⁶⁺	54.297 56	0.0362	8.57 ¹¹	47.566 80	0.1228	2.23 ¹²	44.0014	7.32 ⁻⁴	1.20 ¹⁰
F ⁷⁺	69.051 96	0.0350	1.34 ¹²	60.252 40	0.1249	3.62 ¹²	57.1593	5.88 ⁻⁴	1.54 ¹⁰
Ne ⁸⁺	85.584 00	0.0340	2.00 ¹²	74.437 96	0.1264	5.63 ¹²	70.5212	4.83 ⁻⁴	1.93 ¹⁰

(13) requires a negligible addition of computer time to that already required for the matrix diagonalizations occurring in a standard variational calculation. The perturbation results are com-

parable in accuracy with the variational results for neutral helium and improve with increasing Z . For the ions with $Z \geq 3$, the Z -expansion perturbation procedure is recommended.

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