

Two-photon transitions in hydrogen: A test of pseudostate summation

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We make another application of a remarkably efficient technique, in which the intermediate states of conventional second-order perturbation theory are replaced by a finite set of pseudostates, and the energy denominators are replaced by the expectation values of the unperturbed Hamiltonian with respect to those pseudostates. In previous calculations (of van der Waals coefficients and frequency-dependent polarizabilities) in which the pseudostates were determined by diagonalization of the unperturbed Hamiltonian, excellent converged results were obtained with as few as five to ten terms representing both bound and continuum states. Following the work of Bassani, Forney, and Quattropani [Phys. Rev. Lett. **39**, 1070 (1977)], we use the method to investigate the $2S$ - $1S$ transition rate using both the $\mathbf{E}\cdot\mathbf{x}$ and $\mathbf{A}\cdot\mathbf{p}$ forms of the interaction. The results mirror closely those obtained in the conventional way, even when only a small number of terms are used, and they are not strongly dependent on the exact form of the basis functions used.

I. INTRODUCTION

The problem of the choice of gauge in multiphoton transitions was investigated some years ago¹ by the illustrative method of actually computing the two-photon transition rates between the $2S$ and $1S$ states of atomic hydrogen in two different gauges. In agreement with their earlier conclusions² in the dipole approximation the same results were obtained using the two interactions $\mathbf{E}\cdot\mathbf{x}$ and $\mathbf{A}\cdot\mathbf{p}$ provided that a complete set of intermediate states was used. They also showed that the sum over intermediate states converged very differently in the two gauges and that the importance of continuum states was also radically different in the two cases.

In the present work we repeat the calculation of Ref. 1 using the pseudostate summation technique which has proven to be of remarkable efficiency in carrying out just such intermediate-state sums in other problems.³ The advantage of this method, if it is successful, lies in the replacement of the doubly infinite sum over both discrete and continuum states by a finite sum over tractable, square-integrable pseudostates some of which lie in the energy range normally occupied by the continuum. In previous applications this finite sum has required only a very small number of terms to achieve quite high accuracy. In the present calculation we wished to test the usefulness of the method in a new sort of problem.

In Sec. II the pseudostates are defined, and we give the mathematical expressions for the two-photon matrix element in both forms and also rewrite the momentum form using a well-known commutator expression which might not seem to be valid for the pseudostates. In Sec. III numerical results are presented for all three forms and at a range of photon energies, and excellent agreement with the previous work is demonstrated.

II. MATHEMATICAL FORMULATION

The pseudostate method begins with the choice of N_0 basis functions of angular momentum 1 (because we are going to use the dipole approximation for the radiation field and are coupling two S states.) We take them to have the simple form

$$\phi_j = e^{-ar} r^{1+j} P_1 \quad (j=0, 1, \dots, N_0-1), \quad (1)$$

and linear combinations of these are used to diagonalize approximately the hydrogen Hamiltonian ($H_0 = -\nabla^2 - 2/r$ in rydberg units)

$$\psi_N = \sum_j C(N, j) \phi_j, \quad (2)$$

$$\langle N | H_0 | N' \rangle = \hat{E}_N \delta_{NN'}, \quad \langle N | N' \rangle = \delta_{NN'}.$$

In what follows we will use finite sums (N_0 terms) of these pseudostates ψ_N or $|N\rangle$ wherever infinite summations over complete sets of hydrogenic functions usually occur and will use the corresponding energies \hat{E}_N in the denominators of perturbation sums. The parameter a is adjustable and might be set equal to 1 corresponding to the range of the hydrogenic $1S$ state or to $\frac{1}{2}$ to simulate the $2S$ state. In fact, the results we will present later are remarkably insensitive to the value of a .

The finite-dimensional eigenvalue problem corresponding to the hydrogenic Hamiltonian H_0 is the following:

$$(\underline{H} - \hat{E}_N \underline{\Delta}) |N\rangle = 0, \quad (3)$$

where the elements of the Hamiltonian matrix \underline{H} and the overlap matrix $\underline{\Delta}$ are given by the following simple expressions:

$$H_{jk} = \frac{(j+k)!}{(2a)^{j+k+1}} \left[\frac{5}{2} - \frac{1}{a} + \left(\frac{1}{4} - \frac{1}{a} \right) (j+k) - \frac{(j-k)^2}{4} \right], \quad (4)$$

$$\Delta_{jk} = \frac{(2+j+k)!}{(2a)^{j+k+3}}.$$

The matrix eigenvalue problem in Eq. (3) is fairly standard, although for the larger expansion lengths N_0 it is necessary to work in multiple precision to avoid loss of significance. Knowing the eigenvalues \hat{E}_N and eigenfunctions $|N\rangle$ we can proceed to the evaluation of the two-photon matrix elements in their various forms.

In the length formulation, the second-order perturbation sum involved in the two-photon transition in which we are interested takes the following form:

$$D_1 = \frac{1}{2} \sum_N \left[\frac{1}{1 + \hat{E}_N - \omega} + \frac{1}{\frac{1}{4} + \hat{E}_N + \omega} \right] R_{1S}^N R_{2S}^N. \quad (5)$$

In the momentum formulation the corresponding expression is

$$D_2 \rightarrow D_3 = -\frac{1}{2} \sum_N \left[\frac{1}{1 + \hat{E}_N - \omega} + \frac{1}{\frac{1}{4} + \hat{E}_N + \omega} \right] R_{1S}^N R_{2S}^N \frac{(1 + \hat{E}_N)(1/4 + \hat{E}_N)}{\omega(3/4 - \omega)}. \quad (8)$$

It will be interesting to see how close this third form of the matrix element approaches the second in spite of its even more approximate nature.

III. RESULTS AND CONCLUSIONS

We will now present our numerical results for the three forms D_1 , D_2 , and D_3 of the two-photon transition-

TABLE I. Pseudostate energies \hat{E}_N obtained by solving the matrix eigenvalue problem Eq. (3). Results are given for the scale parameter $a = 1$ and 0.55.

N	Energy \hat{E}_N (Ry)	
	$a = 1$	$a = 0.5$
1	-0.250 000 000	-0.250 000 000 0
2	-0.111 110 988	-0.111 111 111 1
3	-0.062 144 252	-0.062 499 999 6
4	-0.030 756 376	-0.039 993 737 3
5	0.012 423 677	-0.027 020 193 7
6	0.073 441 629	-0.013 633 007 6
7	0.155 140 247	0.005 564 222
8	0.262 302 992	0.031 983 483
9	0.402 262 529	0.067 384 920
10	0.586 056 239	0.114 609 309
11	0.830 537 004	0.178 109 107
12	1.162 203 753	0.264 970 794
13	1.624 445 117	0.386 886 306
14	2.292 104 254	0.564 149 133
15	3.303 178 358	0.834 402 826
16	4.935 224 971	1.273 850 916
17	7.816 342 057	2.056 203 128
18	13.630 802 343	3.651 314 772
19	28.296 327 562	7.727 908 591
20	87.071 218 883	24.376 920 544

$$D_2 = -2 \sum_N \left[\frac{1}{1 + \hat{E}_N - \omega} + \frac{1}{\frac{1}{4} + \hat{E}_N + \omega} \right] \frac{Q_{1S}^N Q_{2S}^N}{\omega(3/4 - \omega)}. \quad (6)$$

In these expressions the length and momentum matrix elements are defined as follows:

$$R_{kS}^N = \int_0^\infty dr r^3 \chi_N \chi_{kS}, \quad Q_{kS}^N = \int_0^\infty dr r^2 \chi_N \chi'_{kS}. \quad (7)$$

Here the functions χ_N are the radial parts of the pseudostate functions ψ_N defined in Eq. (1), the functions χ_{kS} are the corresponding parts of the 1S and 2S eigenfunctions of hydrogen, and they are all unit normalized in the usual way. The quantity ω is the energy of one of the photons emitted in the transition; energy conservation requires the energy of the second photon to be equal to $3/4 - \omega$ and this condition has been used to eliminate its mention.

If the functions ψ_N were true eigenfunctions of the Hamiltonian H we could make use of the well-known commutator relation $\nabla = [\mathbf{r}, H_0]/2$ to rewrite the second form of matrix element as

matrix element to be compared with the results of Ref. 1. First, however, we would like to present, in tabular form, the energy values \hat{E}_N generated in the diagonalization process discussed above; two different values of the scale

TABLE II. Convergence of the matrix elements for $\omega = 0.3750$, $a = 1$, and $N_0 = 20$. The cumulative sum over N is shown; D_3 is not distinguishable from D_2 under these conditions. Items in parentheses are the results of Ref. 1 for the sum over discrete states and total, respectively.

N	D_1	D_2
1	-17.878 45	$\approx 10^{-13}$
2	-14.796 88	-2.705 36
3	-14.066 45	-3.620 47
4	-13.518 32	-4.448 77
	(-13.382 4)	(-4.692 4)
5	-12.989 60	-5.447 67
6	-12.562 92	-6.501 14
7	-12.255 91	-7.522 84
8	-12.052 57	-8.457 96
9	-11.926 89	-9.275 35
10	-11.854 14	-9.961 33
11	-11.814 80	-10.514 76
12	-11.795 07	-10.943 16
13	-11.786 02	-11.259 70
14	-11.782 30	-11.480 95
15	-11.780 98	-11.625 13
16	-11.780 59	-11.710 72
17	-11.780 50	-11.755 22
18	-11.780 48	-11.774 09
19	-11.780 48	-11.779 71
20	-11.780 48	-11.780 48
	(-11.780 5)	(-11.780 5)

TABLE III. Convergence of the matrix elements as in Table II, but for $a = 0.55$.

N	D_1	D_2
1	-17.878 45	$\approx 10^{-19}$
2	-14.796 93	-2.705 31
3	-14.102 61	-3.573 22
4	-13.825 77	-3.970 11
5	-13.650 75	-4.240 14
6	-13.441 05	-4.587 79
	(-13.382 4)	(-4.692 4)
7	-13.194 98	-5.037 47
8	-12.933 69	-5.578 19
9	-12.675 66	-6.199 79
10	-12.436 69	-6.890 40
11	-12.229 38	-7.633 92
12	-12.062 26	-8.408 08
13	-11.938 91	-9.182 85
14	-11.857 50	-9.920 03
15	-11.811 18	-10.575 37
16	-11.789 69	-11.104 84
17	-11.782 27	-11.476 51
18	-11.780 65	-11.685 94
19	-11.780 49	-11.766 67
20	-11.780 48	-11.780 48
	(-11.780 5)	(-11.780 5)

factor a have been used. In Table I these energies are shown for the expansion length $N_0 = 20$ and for $a = 1$ and 0.55 . Notice that in the first case only four of these energies are negative and thus represent the true discrete spectrum of the hydrogen atom, while in the second case there are seven of them. In addition, the highest-lying eigenvalue in the first case is much higher than in the second case. Nevertheless, we shall see later that in both cases the matrix elements are very well represented. It is also interesting to note how well the lowest negative eigenvalues approximate the true hydrogenic values of $-1/(N+1)^2$. (Similar results are obtained for smaller values of N_0 .)

In Tables II and III the convergence of the sum over N is shown, for one particular value of ω , for both values of a , and again for the expansion length $N_0 = 20$. Notice in both tables that the part of the sum representing the discrete spectrum is quite close to the discrete sum done the usual way in Ref. 1, while the complete sum is in exact agreement to five decimal places for both values of a . It is not surprising that the separation into "discrete" and "continuum" parts of the sum is not exact; perhaps more startling is how good the separation actually is and how excellent is the total.

Finally, in Table IV we display the ω dependence of the matrix elements, as compared with the results of Ref. 1.

TABLE IV. ω dependence of the three forms of matrix element and their convergence with expansion length. For each value of ω the first line is the result for $N_0 = 5$, the second for $N_0 = 10$, and the third for $N_0 = 15$ ($a = 1$). The results of Ref. 1 are in the last column.

ω	D_1	D_2	D_3	Ref. 1
0.3750	-11.779 1	-11.780 7	-11.092 4	-11.7805
	-11.780 48	-11.780 48	-11.786 00	
	-11.780 483	-11.780 483	-11.780 449	
0.5250	-14.720 5	-14.735 0	-13.903 0	-14.7319
	-14.731 87	-14.731 87	-14.738 44	
	-14.731 87	-14.731 87	-14.731 83	
0.6750	-40.738 8	-41.470 2	-38.831 4	-41.1484
	-41.148 36	-41.148 44	-41.163 68	
	-41.148 41	-41.148 41	-41.148 32	
0.6875	-49.052 1	-50.268 3	-46.804 8	-49.6878
	-49.687 68	-49.687 84	-49.705 74	
	-49.687 78	-49.687 78	-49.687 67	
0.7000	-61.606 8	-63.802 2	-58.847 9	-62.6595
	-62.659 28	-62.659 63	-62.681 45	
	-62.659 47	-62.659 47	-62.659 34	
0.7125	-82.590 6	-87.098 3	-78.976 4	-84.5252
	-84.524 75	-84.525 64	-84.553 78	
	-84.525 17	-84.525 17	-84.524 99	
0.7250	-124.360 6	-136.046 5	-119.032 8	-128.683
	-128.682 43	-128.685 28	-128.725 23	
	-128.683 52	-128.683 52	-128.683 26	
0.7375	-246.273	-299.359	-235.798	-262.165
	-262.161 05	-262.177 39	-262.245 22	
	-262.165 42	-262.165 42	-262.164 90	
0.7475	-1042.68	-2296.20	-991.00	-1334.33
	-1334.247	-1334.794	-1334.662	
	-1334.3261	-1334.3262	-1334.3236	

In addition, we show their convergence with N_0 , and it is clear that no more than about 15 terms are needed to match the accuracy (six significant figures) of Ref. 1. As expected, the form derived from the commutator identity (D_3) converge slowest.

We conclude once again that the pseudostate summation is an accurate, powerful, and effective technique for carrying out the second-order sums common in atomic theoretical calculations. Since it does not require sums over large numbers of discrete states and difficult integrations over continuum functions, this method should continue to prove useful.

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