

## High-accuracy calculation of muonic molecules using random-tempered basis sets

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We use random-tempering formulas and explicitly correlated Slater-type geminals to calculate the  $S$  and  $P$  bound-state energies of the muonic molecules  $xy\mu$ , where  $x, y = p, d, t$ . The final binding energies are accurate to about  $1 \mu\text{eV}$  or better except for the weakly bound  $td\mu(11)$  state. For this state we get a binding energy of  $0.660\,1721 \text{ eV}$ , which is better than any previous calculation.

### I. INTRODUCTION

The binding energy of muonic molecules has been the subject of several recent papers because of the importance of this quantity to muon-catalyzed fusion (see, for example, Refs. 1–10). For simplicity, we shall refer to each bound state simply as  $xy\mu(mn)$ , where the label  $m$  denotes the angular momentum ( $0=S, 1=P$ ) and  $n$  specifies whether the molecule is in the ground state ( $n=0$ ) or the first excited state ( $n=1$ ). Of all the systems, the  $td\mu$  molecule has perhaps been the most extensively examined. Using large generalized Hylleraas basis sets, Szalewicz *et al.*<sup>1</sup> calculated the binding energies of this system to an accuracy of about  $1 \mu\text{eV}$  except for the weakly bound  $td\mu(11)$  state. For the other muonic molecules, Puzynin and Vinitsky have recently reviewed the various computational studies.<sup>9</sup> For many systems, however, the accuracy obtained by these studies was relatively poor. For this reason we have computed the binding energies of all these molecules using large basis sets of explicitly correlated Slater-type geminals. The parameters of this basis are chosen using random tempering formulas. This method allows us to easily increase the size of our basis until the desired convergence is reached. As a result, we have been able to calculate the binding energies of all the muonic molecules to an accuracy of about  $1 \mu\text{eV}$  except for the weakly bound  $td\mu(11)$  state. This state converges much more slowly than the others and is only accurate to about  $10^{-4} \text{ eV}$ . Like most of the other calculations, our work assumes that only the Coulomb interaction is present although the importance of relativistic and other effects has been recognized.<sup>11–14</sup> For the  $td\mu(11)$  state these terms can shift the binding energy by  $10^{-2}$  to  $10^{-3} \text{ eV}$ . Since the accurate calculation of these changes requires a very accurate wave function, this study also shows how an accurate basis set for this function can be constructed.

### II. METHOD

If the muonic molecules  $xy\mu$  are assumed to interact in a purely Coulombic manner, the complete three-body Hamiltonian for these systems can be defined as

$$H = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\left(\frac{\mu_1}{\mu_2}\right)\nabla_2^2 - \left(\frac{\mu_1}{m}\right)\nabla_1 \cdot \nabla_2 - \frac{1}{r_1} - \frac{1}{r_2} + \frac{1}{r_{12}}, \quad (1)$$

where  $\mathbf{r}_1$  and  $\mathbf{r}_2$  are the vectors from the muon to particles  $x$  and  $y$ , respectively,  $r_{12}$  is the distance between  $x$  and  $y$ , and  $\mu_i = mM_i/(m+M_i)$  are the reduced mass of the muon- $x$  ( $i=1$ ) and muon- $y$  ( $i=2$ ) systems, respectively. Our units are chosen such that  $\hbar = e = \mu_1 = 1$  and it is assumed that nucleus  $x$  is the heavier one.

Earlier studies by Smith<sup>10,15</sup> and by Frolov<sup>2–4</sup> have shown that explicitly correlated Slater-type geminals form an excellent basis for this problem. Since the present work is concerned with only  $S$  and  $P$  bound states, the appropriate wave functions can be written as

$$\Psi = (1 + P_{12}) \sum_{i=1}^K c_i e^{-\alpha_i r_1 - \beta_i r_2 - \gamma_i r_{12}} \quad (2)$$

for homonuclear  $S$  states,

$$\Psi = \sum_{i=1}^K c_i e^{-\alpha_i r_1 - \beta_i r_2 - \gamma_i r_{12}} \quad (3)$$

for heteronuclear  $S$  states,

$$\Psi = (1 - P_{12}) \sum_{i=1}^K c_i r_1(\cos\theta_1) e^{-\alpha_i r_1 - \beta_i r_2 - \gamma_i r_{12}} \quad (4)$$

for homonuclear  $P$  states,

$$\Psi = \sum_{i=1}^{K'} c_i r_1(\cos\theta_1) e^{-\alpha_i r_1 - \beta_i r_2 - \gamma_i r_{12}} + \sum_{j=1}^{\hat{K}'} \hat{c}_j r_2(\cos\theta_2) e^{-\hat{\alpha}_j r_1 - \hat{\beta}_j r_2 - \hat{\gamma}_j r_{12}}, \quad (5)$$

for heteronuclear  $P$  states, where  $P_{12}$  is the operator that interchanges  $\mathbf{r}_1$  and  $\mathbf{r}_2$ . For simplicity, we set  $\hat{K}' = K'$ ,  $\hat{\alpha} = \alpha$ ,  $\hat{\beta} = \beta$ , and  $\hat{\gamma} = \gamma$  in our calculations and defined the label  $K = K' + \hat{K}'$ . In the sections below we use the value of  $K$  to characterize our calculations since this quantity is both the total number of basis functions and the size of the overlap and Hamiltonian matrices. Because the size of these matrices is intimately related to the amount of computer time needed in any approach, this label allows us to easily compare the quality of different kinds of basis sets, e.g., generalized Hylleraas or Gaussian.

Before we can begin calculating the desired roots of the generalized eigenvalue equation some method of selecting the nonlinear parameters in the basis set needs to be chosen. The most accurate method would be to non-

linearly optimize all  $3K$  parameters. This approach gives the most compact wave function but it also requires a great deal of computer time since such optimizations need at least  $N^2$  function evaluations where  $N$  is the number of nonlinear parameters. Several studies have shown that random tempering is a much more efficient way of selecting these parameters.<sup>15-17</sup> Using this method Frolov<sup>2-4</sup> has calculated the binding energies of several muonic molecules  $xy\mu$  although his results for the weakly bound  $td\mu(11)$  and  $dd\mu(11)$  states have never converged to more than three or four significant figures. In a more recent paper Petelenz and Smith<sup>10</sup> have also used random tempering of exponents to study a number of these systems. Below, we propose a new random-tempering formula with which to perform these calculations. By systematically increasing the number of basis functions, we show that the binding energies can be calculated to a high degree of accuracy and we investigate in detail how such calculations are best performed.

### III. RANDOM TEMPERING

Our algorithm for producing a basis set of random tempered geminals is a simple one.

Step 1. Select a tempering formula for each type of nonlinear parameter.

Step 2. Optimize the tempering parameters using a relatively small number of geminals ( $K=50$  or  $100$ ).

Step 3. Take these optimized parameters and calculate the energy using an increasingly larger number of geminals until convergence is reached, the number of geminals becomes intractable or linear dependence occurs.

For our tempering formulas we choose

$$\begin{aligned}\alpha_i &= \left| \sum_{j=1}^I A_j \langle i, 3j-2 \rangle \right|, \\ \beta_i &= \left| \sum_{j=1}^I B_j \langle i, 3j-1 \rangle \right|, \\ \gamma_i &= \left| \sum_{j=1}^I C_j \langle i, 3j \rangle \right| - \min(\alpha_i, \beta_i),\end{aligned}\quad (6)$$

where  $A_j$ ,  $B_j$ , and  $C_j$  are the tempering parameters,  $\langle k, j \rangle$  is the fractional part of  $\{[k(k+1)/2][P(j)]^{1/2}\}$ , and where  $P(j)$  is the  $j$ th prime number in the sequence  $2, 3, 5, 7, \dots$ . One feature of this equation is that it allows us to increase the number of tempering parameters in a systematic fashion. We felt this flexibility would be important in achieving fast convergence and in avoiding linear dependence. Equation (6) is not the only possible method of tempering the nonlinear parameters  $\alpha_i$ ,  $\beta_i$ , and  $\gamma_i$ , however. Both Smith and Frolov have used slightly different formulas. The advantages and disadvantages of these methods will be discussed in more detail in Sec. IV.

In order to find the optimum number of tempering parameters, we calculated the binding energies of the  $td\mu$  states using a total of three, six, and nine parameters. These parameters were optimized using 50 basis functions ( $K=50$ ) except for the ones associated with the  $td\mu(11)$  state. When this relatively small optimization basis was used, none of our expansions (step 3 in our al-

gorithm) had a positive binding energy and so we increased the optimization size to  $K=100$ . For the masses of the various particles we choose set 1 from Table I. The results of these numerical experiments are given in Table II. The first number in each column is the binding energy obtained by the optimization and the others are the binding energies given by the various expansions. To gauge the effectiveness of these calculations, we compared our results with those computed using a large generalized Hylleraas basis set.<sup>1</sup> Table II shows that the quality of our wave function noticeably increases when we go from three to six tempering parameters. In contrast, we find little difference between calculations with six and nine parameters. For this reason we performed the rest of our calculations with six parameters.

Now that the number of tempering parameters has been selected, we must choose the optimum size with which to optimize each calculation. We begin by reoptimizing the systems in Table II with double the number of basis functions used before. When compared with the energies in Table II, the expansions in Table III show no real improvement except for the  $td\mu(01)$  state. This state now converges much faster to the saturated result of Ref. 1. Unless poor convergence occurs or linear dependence appears before we have reached our goal of calculating the binding energies to about  $10^{-1}$  eV, we shall perform all future optimizations with 50 basis functions. In cases like the  $td\mu(01)$  and  $td\mu(11)$  states, a  $K=100$  optimization will be used.

Tables II and III also illustrate the random nature of our algorithm. As expected, the optimized total energy decreases both as the number of tempering parameters increase and as the number of basis functions in the optimization increase. When the tempering parameters are expanded, however, there is no reason for either trend to continue. As a result, comparisons between different expansions can sometimes seem almost nonphysical, e.g., many of the  $td\mu(11)$  calculations with six parameters are much better than the same nine-parameter calculations. These differences occur when the tempering parameters fortuitously place a number of basis functions in an important region of the wave function. For this reason such differences always become smaller as the number of basis functions increases.

TABLE I. Masses (in  $m_e$ ) and conversion constants (in eV). The conversion constant is defined such that the binding energy of  $xy\mu$  is  $(-0.5 - E)C(xy\mu)$ , where  $E$  is energy in  $C(xy\mu)$  units.

Mass set 1	
$m_t = 5496.899$	$C(t\mu) = 5422.5347$
$m_d = 3670.481$	$C(d\mu) = 5326.4520$
$m_p = 1836.1515$	$C(p\mu) = 5057.0346$
$m_\mu = 206.7686$	
Mass set 2	
$m_t = 5496.918$	$E(t\mu) = 5422.5354$
Rest same as set 1	

TABLE II. Selected binding energies (in eV) as a function of the number of tempering parameters. An optimization size of  $K = 100$  is used for the  $td\mu(11)$  state and  $K = 50$  for the others. The masses in set 1 from Table I are used.

$K$	$td\mu(00)$	$td\mu(01)$	$td\mu(10)$	$td\mu(11)$
Three parameters				
50	317.626 164	22.396 905	229.077 508	
100	319.031 755	32.188 508	231.507 356	-8.276 530
200	319.135 774	34.680 471	232.402 839	-0.467 962
300	319.139 534	34.790 281	232.453 974	0.409 753
400	319.139 268	34.827 467	232.467 807	0.564 978
500	319.139 747	34.831 500	232.470 284	0.591 478
Six parameters				
50	318.384 116	26.520 021	229.377 947	
100	319.066 398	32.455 404	231.233 280	-2.618 614
200	319.137 714	34.646 858	232.380 234	-0.003 939
300	319.139 455	34.792 507	232.458 847	0.562 870
400	319.139 720	34.823 764	232.469 212	0.635 954
500	319.139 746	34.832 000	232.470 712	0.647 551
Nine parameters				
50	318.807 441	28.736 492	230.293 695	
100	319.073 763	33.049 394	231.329 488	-1.740 285
200	319.138 661	34.667 302	232.369 410	-0.605 089
300	319.139 590	34.795 877	232.456 752	0.385 429
400	319.139 734	34.826 407	232.467 758	0.624 622
500	319.139 743	34.831 661	232.469 795	0.645 585
Ref. 1	319.139 752 161	34.834 465	232.471 537	0.660 01

#### IV. COMPUTATIONAL CONSIDERATIONS

We performed a number of calculations in order to benchmark our program with respect to the literature. One series of tests attempted to reproduce the calculations of Thakkar and Smith.<sup>15</sup> Since our wave function is identical to the one they used to calculate the energies of the helium atom, our results should agree to a large number of digits. Using the same random-tempering method they described, we were able to obtain the same energies to all figures given.

A second set of calculations was done to compare our converged binding energies with those obtained by Szalewicz *et al.*<sup>1</sup> This work uses a large generalized Hylleraas basis set and, with the exception of the  $td\mu(11)$  state, has converged to 1  $\mu\text{eV}$  or better. We obtain quite good agreement with these values and see in Table IV that our expansions are slightly longer for the  $td\mu(00)$

and  $td\mu(10)$  states, slightly shorter for the  $td\mu(01)$  state and much shorter for the  $td\mu(11)$  state. Although computational limitations prevented us from expanding this last state even further, our  $K = 2000$  value is better than any previous calculation. We see also that this result agrees quite well with the predicted limit of  $0.6601 \pm 0.0001$  eV in Ref. 1.

The last series of tests was designed to reproduce the work of Frolov.<sup>2-4</sup> In a number of papers, he reports the energies and binding energies of muonic ions using random tempered Slater-type geminals. We attempted to recalculate several of these values with less than satisfactory results. One example is his  $K = 250$  energy for the  $td\mu(10)$  state.<sup>2</sup> Frolov obtained a value of  $-0.542 838 011$  for this system; our value is  $-0.542 834 992$ . Although an error in his program, a series of misprints in his papers, numerical precision problems in his calculations or some unknown combina-

TABLE III. Selected binding energies (in eV) using an optimization size of  $K = 200$  for the  $td\mu(11)$  state and  $K = 100$  for the others. Six tempering parameters and the masses in set 1 from Table I are used.

$K$	$td\mu(00)$	$td\mu(01)$	$td\mu(10)$	$td\mu(11)$
100	319.073 315	34.240 373	231.915 650	
200	319.138 788	34.773 046	232.379 594	0.304 859
300	319.139 608	34.826 499	232.455 201	0.550 044
400	319.139 730	34.833 321	232.467 884	0.636 934
500	319.139 743	34.834 111	232.470 300	0.643 036

tion of the above cannot be ruled out, we believe this difference is due to his method of diagonalization. During the testing phase of this project we noticed that a random tempered Slater-type geminal basis is almost linearly dependent and a similar observation has been made by Petelenz and Smith.<sup>10</sup> To avoid the numerical problems associated with such basis sets, we performed all calculations in 128-bit precision (double precision on the ETA-10 and CYBER 205 and extended precision on the IBM 3090). The generalized eigenvalue problem was solved using the inverse moment method<sup>18</sup> or the QL method.<sup>19</sup> Other than stating that he also used the inverse iteration method, Frolov does not mention any problems with linear dependence. One reason may be that he has used a regularization scheme similar to the one described by Korobov, Puzynin, and Vinitsky.<sup>8</sup> This procedure shifts the diagonal of the overlap by some small amount and thus eliminates any possibility of linear dependence. The disadvantage of this method is that the resulting eigenvalues are not as accurate. If we shift the diagonal of the overlap matrix by  $10^{-11}$  in the example above, our energy becomes  $-0.542\,827\,017$ . The difference in the energy between this result and our previous value is of the same order of magnitude as the difference between our result and Frolov's. Until more details are known about these calculations, we believe that Frolov's results should not be regarded as more reliable than this amount even if they appear to have converged.

## V. CALCULATIONS

Using the algorithm outlined in Secs. II and III we computed the binding energies of the muonic molecules  $xy\mu$ . For this set of calculations we chose the masses in set 2 of Table I because these values are more recent estimates<sup>20</sup> than those used in the tests. The results of our calculations are given in Table V and the final optimization parameters for these systems are listed in Table VI.

In general, all the binding energies have converged to an accuracy of about  $10^{-6}$ . The one exception is the  $td\mu(11)$  state. Like their counterparts in Table IV, these calculations converge slowly.

The  $td\mu$  calculations in Tables IV and V show that even a relatively small change in the masses can influence the binding energies to about  $10^{-4}$  eV. Our calculations also show, however, that the differences in the energies produced by these two sets of masses converge to a constant value much faster than the binding energies. This is because the Hamiltonian for one tritium mass can be written as a similar Hamiltonian for another tritium mass plus an additional term. Since a large random-tempered basis set is nearly complete regardless of which masses are chosen, diagonalizing these two Hamiltonians with the same basis and then subtracting the result is the same as evaluating a small perturbation to first order. Because of the amount of computer time needed by the larger calculations, we elected not to repeat all the  $td\mu(11)$  expansions and have used this computed constant to estimate three of the entries in Table V.

A comparison of Table V with the literature also shows the advantages and disadvantages of our tempering method. In Ref. 2 Frolov reports calculations using a method similar to the one used by Thakkar and Smith for helium. For the bulk of the systems examined this form converges slightly faster than ours. In addition, the tempering formula used by Petelenz and Smith<sup>10</sup> also seems to be somewhat better. This is probably because our method selects exponents from the range  $[0, X]$  where  $X$  is the maximum of  $A_1$  and  $A_2$  in Eq. (5), for example. In particular, the interval from zero to the minimum of  $A_1$  and  $A_2$  is extensively sampled. These very small exponents do not appear to contribute substantially to the energy and thus slow the convergence. Frolov eliminates these choices in this calculations by sampling  $[Y, Z]$  ( $0 \leq Y < Z$ ) and Petelenz and Smith use an exponential factor. For the weakly bound  $td\mu(11)$  and  $dd\mu(11)$

TABLE IV. Converged binding energies (in eV) for  $td\mu$ . The optimization size is  $K = 100$  for the  $td\mu(01)$  and  $td\mu(11)$  states and  $K = 50$  for the other. Six tempering parameters and the masses in set 1 from Table I are used.

$K$	$td\mu(00)$	$td\mu(01)$	$td\mu(10)$	$td\mu(11)$
50	318.384 116 914 8		229.377 947 0	
100	319.066 398 092 0	34.240 373 4	231.233 280 1	-2.618 614 2
200	319.137 714 817 5	34.773 046 3	232.380 234 7	-0.003 937 4
300	319.139 455 700 6	34.826 499 6	232.458 847 0	0.562 870 3
400	319.139 720 174 4	34.833 321 7	232.469 212 8	0.635 954 0
500	319.139 746 258 1	34.834 111 0	232.470 712 7	0.647 551 2
600	319.139 749 930 2	34.834 339 7	232.471 230 3	0.655 709 0
800	319.139 752 005 2	34.834 448 2	232.471 459 7	0.658 929 9
1000	319.139 752 131 9	34.834 462 0	232.471 520 5	0.659 673 6
1200	319.139 752 155 4	34.834 464 4	232.471 530 5	0.659 951 6
1400	319.139 752 161 8	34.834 464 7	232.471 534 9	0.660 065 3
1600			232.471 536 4	0.660 105 7
1800			232.471 536 9	0.660 136 5
2000				0.660 146 9
Ref. 1	319.139 752 161 ( $K = 1158$ )	34.834 465 ( $K = 1995$ )	232.471 537 ( $K = 1072$ )	0.660 01 ( $K = 3063$ )

TABLE V. Converged binding energies (in eV) for the muonic ion  $xy\mu$  where  $x, y = p, d, t$ . The optimization size is  $K = 100$  for the  $td\mu(01)$ ,  $td\mu(11)$ ,  $dd\mu(11)$ ,  $tt\mu(01)$ , and  $tt\mu(11)$  states. All others were optimized with  $K = 50$ . Six tempering parameters and the masses in set 2 from Table I are used.

$K$	$td\mu(00)$	$td\mu(01)$	$td\mu(10)$	$td\mu(11)$
50	318.384 0856 83		229.378 001 3	
100	319.066 3679 37	34.240 398 0	231.233 492 1	-2.618 602 8
200	319.137 6849 04	34.773 072 4	232.380 291 1	-0.003 916 4
300	319.139 4258 00	34.826 525 9	232.458 903 8	0.562 897 6
400	319.139 6902 76	34.833 348 1	232.469 269 6	0.635 979 2
500	319.139 7163 60	34.834 137 4	232.470 769 5	0.647 576 5
600	319.139 7200 32	34.834 366 2	232.471 287 1	0.655 734 2
800	319.139 7221 07	34.834 474 6	232.471 516 5	0.658 955 1
1000	319.139 7222 33	34.834 488 4	232.471 577 3	0.659 698 9
1200	319.139 7222 56	34.834 490 9	232.471 587 3	0.659 976 8
1400	319.139 7222 59	34.834 491 2	232.471 591 7	0.660 090 5
1600			232.471 593 2	0.660 130 9 <sup>a</sup>
1800			232.471 593 5	0.660 161 7 <sup>a</sup>
2000				0.660 172 1 <sup>a</sup>
	$dd\mu(00)$	$dd\mu(01)$	$dd\mu(10)$	$dd\mu(11)$
50	325.051 354 9	35.643 998 8	226.497 862 3	
100	325.072 911 3	35.812 249 9	226.654 258 3	1.746 057 0
200	325.073 533 0	35.843 202 7	226.681 354 3	1.954 376 9
300	325.073 539 6	35.844 330 8	226.681 640 8	1.971 314 6
400	325.073 540 1	35.844 358 6	226.681 671 7	1.974 040 4
500	325.073 540 2	35.844 360 5	226.681 676 5	1.974 665 9
600			226.681 678 0	1.974 786 8
800			226.681 678 6	1.974 866 6
1000				1.974 871 2
1200				1.974 871 7
	$tt\mu(00)$	$tt\mu(01)$	$tt\mu(10)$	$tt\mu(11)$
50	362.851 055 5		288.614 623 0	
100	362.907 297 7	83.758 320 6	289.106 265 3	44.922 698 1
200	362.909 758 1	83.770 989 9	289.141 231 9	45.183 706 5
300	362.909 768 8	83.771 207 1	289.141 735 9	45.203 982 2
400	362.909 769 6	83.771 215 7	289.141 777 3	45.205 463 9
500	362.909 769 6	83.771 216 5	289.141 782 3	45.205 748 5
600			289.141 782 9	45.205 820 2
800				45.205 852 4
1000				45.205 856 0
1200				45.205 856 3
	$pp\mu(00)$	$pp\mu(10)$	$dp\mu(00)$	$dp\mu(10)$
50	253.144 881 7	107.085 113 5	221.162 275 1	96.290 978 9
100	253.152 100 0	107.246 833 9	221.514 113 8	97.212 789 3
200	253.152 324 3	107.265 669 2	221.548 569 9	97.480 945 9
300	253.152 330 8	107.265 945 3	221.549 301 0	97.495 459 8
400	253.152 331 9	107.265 966 6	221.549 386 7	97.497 791 7
500	253.152 332 2	107.265 969 6	221.549 404 3	97.498 047 6
600		107.265 970 7	221.549 407 6	97.498 117 2
800		107.265 971 4	221.549 409 6	97.498 152 4
1000				97.498 157 8
1200				97.498 159 7
1400				97.498 160 2
	$tp\mu(00)$	$tp\mu(10)$		
50	213.388 294 2	97.783 746 5		
100	213.794 554 7	98.788 460 8		
200	213.839 124 9	99.106 517 4		
300	213.840 004 9	99.123 458 3		
400	213.840 146 3	99.126 053 6		
500	213.840 171 8	99.126 373 8		

TABLE V. (Continued).

$K$	$tp\mu(00)$	$tp\mu(10)$
600	213.840 176 3	99.126 450 2
800	213.840 178 9	99.126 492 3
1000	213.840 179 4	99.126 498 8
1200		99.126 500 9
1400		99.126 501 4

<sup>a</sup>Computed using the corresponding value in Table IV and a constant shift.

states, however, small exponents seem to be essential. Our values converge faster than any other published results and for the first time these important states have been calculated to an accuracy of about  $10^{-4}$  eV or better.

## VI. CONCLUSIONS

We have computed the binding energies for all the muonic molecules  $xy\mu$  where  $x, y = p, d, t$ . The accuracy of these values is about  $1 \mu\text{eV}$  except for the very weakly bound  $td\mu(11)$  state. Because of computational limita-

tions we were only able to converge this last state to about  $10^{-4}$  eV. For the  $td\mu$  molecule our calculations confirm the accuracy of Ref. 1 and for the  $td\mu(11)$  state is better than any previous calculation. In contrast, our calculations bring into question the work of Frolov<sup>2-4</sup> but this disagreement must remain unresolved until further information is available about how his calculations were performed. Finally we have shown that random-tempering methods can be used to efficiently produce a converged set of Slater-type geminals and our binding-energy calculations with these basis sets has produced a

TABLE VI. Optimized tempering parameters for each muonic system.

	$td\mu(00)$	$td\mu(01)$	$td\mu(10)$	$td\mu(11)$
$A_1$	1.271 120 65	1.586 390 00	1.131 563 23	1.304 509 43
$B_1$	1.368 897 28	1.289 637 00	0.947 054 64	1.209 778 09
$C_1$	3.093 182 89	2.255 942 73	2.329 805 43	2.588 810 22
$A_2$	-0.135 864 89	-0.351 806 42	0.136 899 07	0.148 780 77
$B_2$	-0.230 165 24	-0.181 098 35	-0.006 499 85	-0.119 062 10
$C_2$	-0.016 219 88	0.085 242 90	0.030 455 09	-1.381 567 32
	$dd\mu(00)$	$dd\mu(01)$	$dd\mu(10)$	$dd\mu(11)$
$A_1$	1.287 947 58	1.101 207 30	1.155 111 91	1.303 806 08
$B_1$	1.096 259 16	1.087 829 77	1.105 351 64	1.420 947 33
$C_1$	2.441 116 35	2.035 808 98	1.663 022 61	3.030 866 04
$A_2$	0.059 450 90	0.002 438 70	-0.064 201 31	-0.002 042 25
$B_2$	0.134 892 17	-0.113 660 07	-0.044 063 25	-1.379 548 39
$C_2$	0.854 443 69	0.820 064 51	1.316 742 68	-1.801 680 69
	$tt\mu(00)$	$tt\mu(01)$	$tt\mu(10)$	$tt\mu(11)$
$A_1$	1.212 719 53	1.173 887 07	0.619 905 22	1.048 428 99
$B_1$	1.107 333 14	1.026 973 33	1.383 155 82	1.110 496 84
$C_1$	2.680 683 08	0.933 089 96	2.807 695 34	1.731 515 07
$A_2$	0.136 556 81	0.150 260 93	0.980 444 47	0.142 798 90
$B_2$	0.127 818 37	-0.045 010 48	-0.263 287 34	0.062 730 65
$C_2$	1.078 803 09	2.529 656 81	0.949 684 08	0.890 535 90
	$pp\mu(00)$	$pp\mu(10)$	$dp\mu(00)$	$dp\mu(10)$
$A_1$	1.221 079 96	1.219 698 01	1.197 698 08	1.226 850 17
$B_1$	1.034 146 80	1.020 543 46	1.029 420 45	0.916 019 81
$C_1$	1.870 594 59	1.678 151 04	1.816 218 29	1.586 084 39
$A_2$	0.037 008 32	-0.057 854 25	-0.054 842 60	-0.082 766 21
$B_2$	0.091 005 81	-0.075 535 87	-0.075 504 52	0.121 145 30
$C_2$	0.753 954 10	0.616 653 26	0.621 630 14	0.648 621 67
	$tp\mu(00)$	$tp\mu(10)$		
$A_1$	1.167 523 64	1.217 263 80		
$B_1$	1.012 512 70	0.903 710 75		
$C_1$	1.833 757 97	1.640 433 92		
$A_2$	-0.038 274 23	-0.074 334 00		
$B_2$	-0.070 144 39	0.116 693 52		
$C_2$	0.592 054 56	0.662 649 64		

library of coefficients with which a number of additional calculations, e.g., sticking fractions and expectation values, can be performed. A tape of these values will be made available upon request.

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- <sup>1</sup>K. Szalewicz, W. Kolos, H. J. Monkhorst, and A. Scrinzi, *Phys. Rev. A* **35**, 965 (1987).  
<sup>2</sup>A. M. Frolov and V. D. Efros, *Pis'ma Zh. Eksp. Teor. Fiz.* **39**, 449 (1984) [*JETP Lett* **39**, 544 (1984)].  
<sup>3</sup>A. M. Frolov and V. D. Efros, *J. Phys. B* **18**, L265 (1985).  
<sup>4</sup>A. M. Frolov, *Z. Phys. D* **2**, 61 (1986).  
<sup>5</sup>S. Hara, T. Ishihara, and N. Toshima, *J. Phys. Soc. Jpn.* **53**, 3293 (1986).  
<sup>6</sup>C. Y. Hu, *Phys. Rev. A* **32**, 1245 (1985); **34**, 2536 (1986).  
<sup>7</sup>K. Kamimura, *Muon Catalyzed Fusion* **3**, 335 (1988).  
<sup>8</sup>V. I. Korobov, I. V. Puzynin, and S. I. Vinitsky, *Phys. Lett.* **196B**, 272 (1987).  
<sup>9</sup>I. V. Puzynin and S. I. Vinitsky, *Muon Catalyzed Fusion* **3**, 307 (1988).  
<sup>10</sup>P. Petelenz and V. H. Smith, Jr., *Phys. Rev. A* **36**, 4078 (1987).  
<sup>11</sup>L. Bracci and G. Fiorentini, *Phys. Rep.* **86**, 169 (1982).  
<sup>12</sup>P. Petelenz and V. H. Smith, Jr., *Phys. Rev. A* **35**, 4055 (1987); **36**, 4529 (E) (1987).  
<sup>13</sup>D. D. Bakalov, S. I. Vinitsky, and V. S. Melezhik, *Zh. Eksp. Teor. Fiz.* **79**, 1629 (1980) [*Sov. Phys. JETP* **52**, 820 (1980)].  
<sup>14</sup>D. D. Bakalov, V. S. Melezhik, L. I. Mehshikov, and S. I. Vinitsky, *Phys. Lett.* **161B**, 5 (1985).  
<sup>15</sup>A. J. Thakkar and V. H. Smith, Jr., *Phys. Rev. A* **15**, 1 (1977); **15**, 16 (1977); **15**, 2143 (1977).  
<sup>16</sup>R. D. Poshusta, *Int. J. Quantum Chem. Symp.* **13**, 59 (1979).  
<sup>17</sup>S. A. Alexander, H. J. Monkhorst, and K. Szalewicz, *J. Chem. Phys.* **85**, 5821 (1987); **87**, 3976 (1987).  
<sup>18</sup>G. Dahlquist and A. Bjorck, in *Numerical Methods* (Prentice-Hall, Englewood Cliffs, NJ, 1974), p. 208.  
<sup>19</sup>See, for example, the EIGRS routine in the EISPACK library.  
<sup>20</sup>A. H. Wapstra and G. Audi, *Nucl. Phys.* **A432**, 1 (1985).