Binding energies of muonic molecules

Piotr Petelenz* and Vedene H. Smith, Jr.

Department of Chemistry, Queen's University, Kingston, Ontario, Canada K7L 3N6

(Received 15 June 1987)

The integral-transform (generator-coordinate) method has been applied to calculate the binding energies of the lowest S states of a set of muonic molecules. The wave functions obtained are both compact and accurate, as indicated by energy eigenvalues. The calculated values of the energies of $dd\mu$, $tt\mu$, and $tt\mu^*$ are the lowest reported so far, and they lead to a new estimation of the corresponding asymptotic values for a saturated basis set.

I. INTRODUCTION

There has been a revival of interest in the literature in the calculation of the binding energies and other properties of three-body systems. Possibly the most intriguing of such systems to be studied most recently are muonic molecules such as $dd\mu$, $tt\mu$, etc., due to their involvement in catalyzing nuclear fusion.[1-4]

The required accuracy (of the order of 1 meV in energy) is a demanding test of the quality of calculated wave functions. The trial functions used in such calculations are either of Hylleraas type,[2,3] or of explicitly correlated pseudo-Slater type[4] with the exponents generated in a pseudorandom manner from a set of nonlinear variational parameters.

The latter approach is essentially a version of the integral-transform (generator-coordinate) method developed in this laboratory and extensively tested for small atoms.[5-7] In a simplified form, it has been recently applied with success by Frolov and Efros.[4] They used only three nonlinear parameters, whose optimization was rather perfunctory, if done at all. The values of those parameters have been set equal for most muonic molecules and states they considered, instead of being individually optimized for each molecule.

In spite of the simplicity of their approach, Frolov and Efros achieved impressive numerical accuracy. However, in view of the high standard of precision required for nuclear fusion applications, the accuracy of some of the results of Ref. 4 may not suffice. It is therefore logical to apply the earlier, more sophisticated approach[5-7] to the same problem. As we are going to demonstrate, in a number of cases this allows one to reduce the size of the basis set without loss of accuracy, and sometimes even with its gain. In some cases, this produces a considerable change in the estimate of the binding energy obtained by extrapolation to the limit of a saturated basis set.

II. METHOD

The standard approximation in treating muonic molecules consists in the neglect of non-Coulomb contributions to the interaction potential between the particles. With this approximation, the Hamiltonian of a muonic molecule is the sum of the potential energy of the pair-wise Coulomb interactions, and of the kinetic energy of the three particles involved. We used the following values of the constants: $m_t = 5496.899m_e$, $m_d = 3670.481m_e$, $m_p = 1836.151\,527m_e$, $m_\mu = 206.7686m_e$, and $Ry = 13.605\,804\,1$ eV.[2-4]

To solve the corresponding Schrödinger equation, we have applied a modified version of the integral-transform (generator-coordinate) method, originally developed in this laboratory.[5-7] It is equivalent to the variational treatment with the trial function of the form

$$\psi(r_1, r_2, r) = (4\pi)^{-1} \sum_{k=1}^{N} C_k (1 + P_{12})$$

$$\times \exp(-\alpha_k r_1 - \beta_k r_2 - \gamma_k r),$$

$$(1)$$

where r_1 and r_2 denote the distances between each of the two nucleons and the muon, and r is the distance between the nucleons. P_{12} stands for the permutation operator and enters only for homonuclear molecules. The C_k are linear combination coefficients, determined by diagonalizing the Hamiltonian matrix.

The exponents α_k, β_k, and γ_k are generated in a pseudorandom manner from the set of six nonlinear variational parameters A_1, A_2, B_1, B_2, G_1, and G_2, according to the formulas:

$$\alpha_k = \exp[A_1 \langle \tfrac{1}{2}k(k+1)\sqrt{2}\rangle + A_2 \langle \tfrac{1}{2}k(k+1)\sqrt{7}\rangle],$$

$$(2a)$$

$$\beta_k = \exp[B_1 \langle \tfrac{1}{2}k(k+1)\sqrt{3}\rangle + B_2 \langle \tfrac{1}{2}k(k+1)\sqrt{11}\rangle],$$

$$(2b)$$

$$\gamma_k = \exp[G_1 \langle \tfrac{1}{2}k(k+1)\sqrt{5}\rangle + G_2 \langle \tfrac{1}{2}k(k+1)\sqrt{13}\rangle],$$

$$(2c)$$

where $\langle x \rangle$ is the fractional part of x.

This algorithm for generating the exponents (tempering scheme) has been recently proposed by Alexander *et al.*[8] Its advantage, with respect to that previously developed in this laboratory,[5-7] is due to a wider spread of exponents obtained, which alleviates problems with quasilinear dependences between basis functions, en-

TABLE I. Binding energies (in eV) of the lowest S states of homonuclear muonic molecules. N is the number of terms in the trial function according to Eq. (1). Asterisks denote the lowest excited state of S symmetry ($2S$). P, present results; FE, results from Ref. 4; BD, results from Ref. 2 obtained with a Hylleraas-type variational ansatz (number of terms in parentheses).

N	$pp\mu$	$dd\mu$	$dd\mu^*$	$tt\mu$	$tt\mu^*$
			P		
80	253.150 41	325.070 35	35.837 07	362.908 10	83.713 08
120	253.152 05	325.073 49	35.842 93	362.909 40	83.769 86
140	253.152 30	325.073 52	35.843 27	362.909 45	83.769 96
∞	253.1524	325.0735	35.8435	362.9095	83.771
			FE		
100	253.150 40	325.066 95	35.694 46	362.871 01	83.540 17
125	253.152 05	325.071 24	35.810 93	362.895 58	83.684 14
140	253.152 40	325.072 13	35.836 58	362.900 40	83.722 02
∞	253.153	325.074	35.9	362.904	83.78
			BD		
	253.152	325.070	35.815	362.900	83.630
	(125)	(203)	(161)	(203)	(161)

countered for very extended basis sets.

Those problems are usually caused by the loss of accuracy due to cancellations.[5] The typical manifestations of such problems are of two kinds. Firstly, the roundoff errors may accumulate to make the metric matrix in the eigenvalue problem nonpositive definite. Due to a built-in penalty function, the program automatically rejects those sets of nonlinear parameters for which the metric matrix is non-positive-definite and continues the search for optimum parameters in other regions of the parameter space. The only effect of the loss of accuracy consists in the fact that the optimum set of nonlinear parameters cannot be approached too closely (which would be possible with higher numerical precision, i.e., smaller roundoff errors), and the resultant wave function is slightly worse.

In other cases, the loss of accuracy may result in obviously unphysical eigenvalues. As the eigenvalues are not known *a priori*, there is no systematic way for the program to detect this sort of problems. However, errors of this kind may be easily detected by direct inspection of the final results. In doubtful cases the calculations should be repeated with higher precision. Our experience suggests that with the tempering scheme used in this paper [Eqs. (2a)–(2c)], the variational ansatz of up to 100 terms is very safe with the numerical precision obtained with a 16-bit mantissa.

III. RESULTS

In most of our present calculations for heteronuclear muonic molecules, the accuracy of a Perkin Elmer 3251 computer in double precision (53 bit = 16-significant-figure mantissa) turned out to be sufficient to avoid the difficulties mentioned above. In order to safeguard the conclusions against the second kind of errors, we verified the results for a few sample cases by repeating the calculations on an IBM 3081G computer in quadruple precision (109-bit mantissa). In most instances the differences between the two results turned out to be

negligible. For $tp\mu$, although the lower precision results did not exhibit any unphysical features, the higher numerical precision of the IBM yield a substantial improvement of the eigenvalue, probably by allowing the program to approach the energy minimum more closely.

For homonuclear molecules where shorter expansions produce already very good results, the problems with the loss of accuracy for the most extended basis set ($N=140$) are a rule rather than an exception. In those cases, all calculations have been done on an IBM 3081G in quadruple precision.

The calculated binding energies[9] of the lowest S states of several muonic molecules are shown in Tables I and II, and compared with those reported by Frolov and Efros.[4] The lower values of the total energies obtained from our present treatment clearly indicate the very

TABLE II. Binding energies (in eV) of the lowest S states of heteronuclear muonic molecules. N is the number of terms in the trial function according to Eq. (1). Asterisks denote the lowest excited state of S symmetry ($2S$). P, present results; FE, results from Ref. 4; BD, results from Ref. 2 obtained with a Hylleraas-type variational ansatz (number of terms in parentheses).

N	$pd\mu$	$pt\mu$	$dt\mu$	$dt\mu^*$
		P		
80	221.528 89	213.777 70	319.105 69	34.049 83
120	221.546 34	213.837 00	319.134 79	34.669 78
140	221.547 97	213.838 82	319.137 47	34.753 56
∞	221.5503	213.841	319.1411	34.850
		FE		
125	221.533 93	213.828 47	319.049 91	34.342 04
200	221.547 81	213.838 96	319.134 51	34.791 12
250	221.549 23	213.839 75	319.138 05	34.823 81
∞	221.551	213.841	319.140	34.84
		BD		
	221.541	213.829	319.062	34.573
	(440)	(440)	(440)	(440)

good quality of the corresponding wavefunctions. Almost as a rule, the accuracy attained with the present approach is better than that of Frolov and Efros for a basis set with a comparable number of terms. The difference is particularly spectacular for $tt\mu$ where our value of the binding energy even for the least extended basis set ($N=80$) is greater than the limiting (extrapolated to $N=\infty$) value of Ref. 4. This advantage is due to the flexibility added to the trial function by additional nonlinear parameters, and also to their thorough optimization in the present study. It should also be noted that our binding energies are generally lower than those obtained by using Hylleraas-type trial functions, even with a much greater number of terms.[2,3] This observation agrees very well with previous results for atomic systems,[5] demonstrating the ability of this version of the integral transform method to produce wavefunctions that are both accurate and compact.

From the point of view of nuclear fusion applications, the required accuracy of the binding energies is of the order of 1 meV. Although our calculated binding energies are often better (at least in most cases of homonuclear molecules) than those of Frolov and Efros, they usually seem to converge to the same limit, thereby confirming most of the results of Ref. 4.

The discrepancies in the extrapolated values go beyond the 1-meV margin for $dt\mu^*$, $dd\mu$, $tt\mu$, and $tt\mu^*$. (Asterisks denote the first excited S state.) We believe that in the first case the result of Ref. 4 may be more reliable, as suggested by the lower values of calculated energies, due to the use of a much larger number of terms in the trial function. In other cases, based again on the consideration of the calculated binding energies for finite N, we consider the present results more reliable and probably the most accurate to date. For $tt\mu$, our extra-

polated value of 362.9095 eV is in the middle of the interval between the extrapolated values of Ref. 4 (362.904 eV) and Ref. 2 (362.95 eV). Surprisingly enough, our extrapolated values for $dd\mu^*$ and $tt\mu^*$ are smaller than those reported by other authors,[2–4] the difference for $dd\mu^*$ being as large as about 60 meV.[10] However, since our calculated total energies for finite N are considerably lower than those of other authors and seem to converge more rapidly, our extrapolated values also seem to be more accurate.

However, in spite of the achieved numerical accuracy, the physical relevance of our results as well as of those of other authors is limited by the physical approximations adopted, the neglect of the vacuum polarization correction being presumably the most important. An attempt to include it in the calculations is presently underway.[11]

Note added in proof. As we have discovered recently, a direct comparison of the results at this level of accuracy is somewhat difficult due to different values of particle masses assumed by different authors. For instance, Frolov and Efros[4] assume larger values for m_t and m_μ than we do. Consequently, if we used their values, our binding energies would be still larger, i.e., still better than those of Ref. 4. For comparison, we have recalculated the binding energy of the $pp\mu$ molecule with $m_\mu = 206.769 m_e$ (as in Ref. 4), and with $N=140$ we have obtained $-E=253.152\,58$ eV, which is better than the corresponding result of Efros and Frolov (253.152 40 eV).

ACKNOWLEDGMENT

*Permanent address: Department of Theoretical Chemistry, Jagiellonian University, ul. Karasia 3, 30-060 Cracow, Poland.

[1] L. Bracci and G. Fiorentini, Phys. Rep. A **30**, 2138 (1982).

[2] A. K. Bhatia and R. J. Drachman, Phys. Rev. A **30**, 2138 (1984).

[3] Ch.-Y. Hu, Phys. Rev. A **32**, 1245 (1985).

[4] A. M. Frolov and V. D. Efros, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 499 (1984) [JETP Lett. **39**, 544 (1984)]; J. Phys. B **18**, L265 (1985).

[5] A. J. Thakkar and V. H. Smith, Jr., Phys. Rev. A **15**, 1, 16 (1977).

[6] A. J. Thakkar, Ph.D. thesis, Queen's University, 1976.

[7] P. Petelenz and V. H. Smith, Jr., Int. J. Quant. Chem. Symp. **14**, 83 (1980).

[8] S. A. Alexander, H. J. Monkhorst, and K. Szalewicz, J. Chem. Phys. **85**, 5821 (1986).

[9] Our binding energy is defined as the total energy, calculated with respect to the energy of the hydrogenlike atom of the heavier isotope present in the molecule at hand, and taken with reversed sign.

[10] One should bear in mind that the calculated energies have been obtained from a variational treatment, but the extrapolated energies have not. Therefore, the extrapolated values do not provide upper bounds for the actual value of the total energy and the lower extrapolated value need not necessarily be the more accurate one.

[11] P. Petelenz and V. H. Smith, Jr., Phys. Rev. A **35**, 4055 (1987).