Nonadiabatic coupled-rearrangement-channel approach to muonic molecules

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A new variational approach to muonic molecules is proposed. The total three-body wave function is expanded in terms of basis functions spanned over the three rearrangement channels in the Jacobian coordinate system. Energy levels of the $dt\mu$ molecule are calculated with a high accuracy and a short computation time. For the weakly bound state with $J=v=1$, which is a key to the muon-catalyzed *d-t* fusion, the calculated energy ε_{11} is better than the literature data. With the use of the most up-to-date, 1986 CODATA-recommended [E. R. Cohen and B. N. Taylor, CODATA Bull. 63 (1986)] values of physical constants, we obtained $\varepsilon_{11}=-0.660\,264$ eV with 2662 basis functions and $\varepsilon_{11}(\infty)=-0.660\,30\pm000\,02$ eV by extrapolation.

I. INTRODUCTION

In connection with muon-catalyzed fusion, energy levels of muonic molecules have extensively been studied with various calculational methods.[1] It is known[2] that formation of the $dt\mu$ molecule via the resonance reaction $(t\mu)_{1s}+D_2\rightarrow[(dt\mu)_{J=v=1}dee]^*$ is a key to the most interesting *d-t* fusion. The $dt\mu$ molecular state with $J=v=1$ is very weakly bound, by only ~0.66 eV, with respect to the $(t\mu)_{1s}$-*d* threshold. The calculated rate of the resonance reaction is very sensitive[3] to the energy of this state; required accuracy of the calculated energy is $\sim10^{-3}$ eV which is to be compared with the total three-body binding energy of 2.7 keV.

For the nonrelativistic, Coulomb three-body Hamiltonian of the $dt\mu$ system, most recent variational calculations[4-7] which took more than one thousand basis functions give the energy of the weakly bound state more accurately than the adiabatic nonvariational calculations.[8,9] The large-size variational calculations of Refs. 4 and 5 and those of Refs. 6 and 7 utilized basis functions of molecular type in spheroidal coordinates and of generalized Hylleraas type, respectively.

In the present paper we propose a new variational approach to muonic molecules in which all the three rearrangement channels are explicitly employed with the use of their *Jacobian coordinates* (Fig. 1). The reason for taking such an approach is as follows: The fact that the state with $J=v=1$ lies by only ~0.66 eV below the $(t\mu)_{1s}$-*d* breakup threshold suggests that the three-body wave function of the state has a large component corresponding to the $(t\mu)_{1s}$-*d* configuration. Similarly, the $(d\mu)_{1s}$-*t* configuration is likely to be important because the state is located slightly (48 eV) below the $(d\mu)_{1s}$-*t* threshold [cf. the $(t\mu)_{2s,2p}$-*d* threshold appearing 2 keV higher]. The (dt)-μ component is also necessary for determining the wave function precisely in the region of the nuclear fusion. Large-size variational calculations can suffer from a very large linear dependence between the basis functions employed, but the basis functions spanned over the three rearrangement channels are expected to give a not exceedingly large linear dependence and therefore to result in a high accuracy and a short computation time. Furthermore, the framework of the Jacobian coordinate system for the rearrangement channels is found[10] to be particularly suited for a nonadiabatic description of scattering processes such as $(d\mu)_{1s}+t\rightarrow(t\mu)_{1s}+d$ with the correct boundary condition.

II. METHOD

Formulation of our method is presented with the $dt\mu$ system as an example. The three channels $(t\mu)$-*d*, $(d\mu)$-*t*, and (dt)-μ are referred to as channels $c=1$, 2, and 3, respectively, and their Jacobi coordinates $(\mathbf{r}_c,\mathbf{R}_c)$ are defined as in Fig. 1. We introduce the reduced masses (m_c,M_c) associated with the coordinates (r_c,R_c): $m_1^{-1}=m_t^{-1}+m_\mu^{-1}$, $M_1^{-1}=m_d^{-1}+(m_t+m_\mu)^{-1}$, $m_2^{-1}=m_d^{-1}+m_\mu^{-1}$, $M_2^{-1}=m_t^{-1}+(m_d+m_\mu)^{-1}$, and $m_3^{-1}=m_d^{-1}+m_t^{-1}$, $M_3^{-1}=m_\mu^{-1}+(m_d+m_t)^{-1}$, where m_μ, m_d, and m_t are masses of μ^-, *d*, and *t*, respectively.

The nonrelativistic three-body Hamiltonian assumed in the present paper is

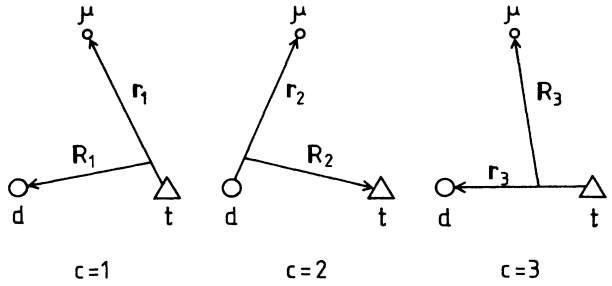

FIG. 1. Three arrangement channels of the $dt\mu$ system and their Jacobian coordinates.

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$$H=-\frac{\hbar^2}{2m_c}\nabla^2_{\mathbf{r}_c}-\frac{\hbar^2}{2M_c}\nabla^2_{\mathbf{R}_c}-\frac{e^2}{r_1}-\frac{e^2}{r_2}+\frac{e^2}{r_3}\ , \tag{1}$$

where $c=1$, 2, or 3. The total wave function with J and its z component M, Ψ_{JM}, may be expanded in terms of basis functions spanned over the three channels:

$$\Psi_{JM}=\sum_{c=1}^{3}\sum_{i_c l_c I_c L_c} A^{(c)}_{i_c l_c I_c L_c}\phi^{(c)}_{i_c l_c}(r_c)\chi^{(c)}_{I_c L_c}(R_c) \times[Y_{l_c}(\hat{\mathbf{r}}_c)Y_{L_c}(\hat{\mathbf{R}}_c)]_{JM}\ . \tag{2}$$

Here l_c (L_c) stands for the angular momentum of the relative motion associated with the coordinate $\mathbf{r}_c$ ($\mathbf{R}_c$), and the bracket $[\ ]_{JM}$ represents the vector coupling of the two spherical harmonics. In Eq. (2), l_c and L_c are restricted as $0\le l_c\le l_c^{\max}$, $|J-l_c|\le L_c\le J+l_c$, and $(-1)^{l_c+L_c}=(-1)^J$. The numbers i_c and I_c specify the radial dependences of $\phi^{(c)}_{i_c l_c}(r_c)$ and $\chi^{(c)}_{I_c L_c}(R_c)$, respectively (see below). The expansion coefficients and the eigenenergies corresponding to Ψ_{JM} may be determined by the Rayleigh-Ritz variational method as usual.

In the present method muon is treated on an equal footing with the two nuclei since the muon mass is not much smaller than the nuclear masses. For the three-body system, the expression (2) must be the most general form of the expansion with a finite number of basis functions. In principle, Ψ_{JM} may be expanded in terms of the complete set of *single*-channel basis functions, but this would need, in actual calculations, up to large l_c and L_c and generate a large linear dependence between the basis functions. Our idea is that we take the three-channel expansion and expect l_c and L_c to remain rather small.

The form of the radial functions ϕ and χ is taken as (the index c omitted)

$$\begin{aligned}&\phi_{il}(r)=r^l\exp[-(r/\bar{r}_i)^2]\ ,\\ &\bar{r}_i=\bar{r}_1 a^{(i-1)}\quad(i=1-n)\ ,\\ &\chi_{IL}(R)=R^L\exp[-R/\bar{R}_I)^2]\ ,\\ &\bar{R}_I=\bar{R}_1 A^{(I-1)}\quad(I=1-N)\ .\end{aligned} \tag{3}$$

The geometrical progressions for $\{\bar{r}_i\}$ and $\{\bar{R}_I\}$ are found to be useful in optimizing the ranges with a small number of free parameters. The nonlinear parameters $(n,\bar{r}_1,\bar{r}_n)$ and $(N,\bar{R}_1,\bar{R}_N)$ are chosen for each c and (l_c,L_c); $\bar{r}_n$ and $\bar{R}_N$ are employed instead of the ratios a and A.

Use of Gaussian tails, rather than exponential ones, makes the transformation between the three sets of the Jacobian coordinates very simple, and therefore closed forms of the energy- and norm-matrix elements are obtained straightforwardly. The "fast" damping of the Gaussian tails is not a serious problem since we can make $\bar{r}_n$ and $\bar{R}_N$ much longer than the muonic molecular size. It is to be noted that Gaussian-type basis functions have often been utilized in variational calculations of atomic and molecular problems,[11] but the present functions spanned over the three rearrangement channels with the use of the Jacobian coordinate system are quite different from them.

The $(t\mu)_{1s}$-d and $(d\mu)_{1s}$-t configurations mentioned above are taken into account as follows: A satisfactorily accurate expansion of the 1s wave function $\Phi_{1s}(r)$ of the $t\mu$ atom is obtained (even at $r\simeq 0$) by diagonalizing the t-μ sub-Hamiltonian with a set of basis functions $\{\phi_{il}(r);\ l=0,\ i=1-n\}$ of Eq. (3) with $n\sim 20$, $\bar{r}_1\sim(0.005-0.01)a_\mu$, and $\bar{r}_n\sim(5-10)a_\mu$, where $a_\mu=\hbar^2/m_\mu e^2\simeq 2.56\times10^{-11}$ cm; the same is true for $\Phi_{1s}(r)$ of the $d\mu$ atom. Therefore, taking those basis functions as $\phi^{(c)}_{i_c l_c}(r_c)$ ($l_c=0,c=1,2$), we can automatically incorporate the $(t\mu)_{1s}$-d and $(d\mu)_{1s}$-t configurations of the $\Phi_{1s}(r_c)\chi_{JM}(R_c)$ type into the total wave function when the Hamiltonian (1) is diagonalized.

III. CALCULATIONS

As for physical constants, we take two sets: set I, $m_\mu=206.7686m_e$, $m_d=3670.481m_e$, $m_t=5496.918m_e$, and $R_y=m_e e^4/2\hbar^2=13.605\,804$ eV (Ref. 12); these were often used in the literature;[5] set II, $m_\mu=206.768\,262m_e$, $m_d=3670.483\,014m_e$, and $R_y=13.605\,698\,1$ eV which are taken from the new, 1986 CODATA-recommended values[13] of physical constants; and $m_t=5496.921\,58m_e$ with m_e from Ref. 13 and m_t from Ref. 14 (since Ref. 13 refers to Ref. 14 for mass of nucleon and deuteron). The calculated energy of a state with J and v is denoted by ε_{Jv} which is measured with respect to the $(t\mu)_{1s}$-d threshold energy, $-(m_1/m_e)R_y$. The number of the basis functions is presented by N_b.

All the numerical calculation was performed in double precision (14~16 decimal digits). For typical sets of the nonlinear variational parameters, the energy-matrix elements were compared with those calculated in quadrupole precision (31–33 decimal digits), and error was found to be less than 10^{-8} eV in all the elements. Diagonalization of the energy and norm matrices was not ill conditioned even for $N_b\sim 2600$, and error in the process was checked to be less than 10^{-7} eV. Thus, the whole computation can be performed with a vector processor which works in double precision. The computation time is then very short. When $N_b=2000$ ($J=1$), the time for calculating all the matrix elements is about 30 sec and that for the eigenvalues and eigenvectors of the lowest-lying ten states is about 100 sec on a FACOM VP-200 computer.

Such a short computation time enables us to optimize the nonlinear variational parameters very carefully. For $J=1$, 15 cases of N_b were chosen. For each case of N_b the optimization was made so as to obtain the best value of ε_{11}; the total number of such trial-and-error calculations for those 15 cases of N_b was about 100. Figure 2 shows the best values of ε_{11} versus N_b thus obtained; some of them are listed in Table I together with ε_{10} which was simultaneously given by the diagonalization;[16] here, we took set I of the physical constants. As seen in Table I, the energy $\varepsilon_{11}=-0.660\,104$ eV ($N_b=2662$) is better than the literature data.[17] The dependence of ε_{11} on N_b in Fig. 2 is very smooth, which shows a high accuracy of the present calculation. The distribution of ε_{11} versus N_b is well followed by the dashed curve which is given by the

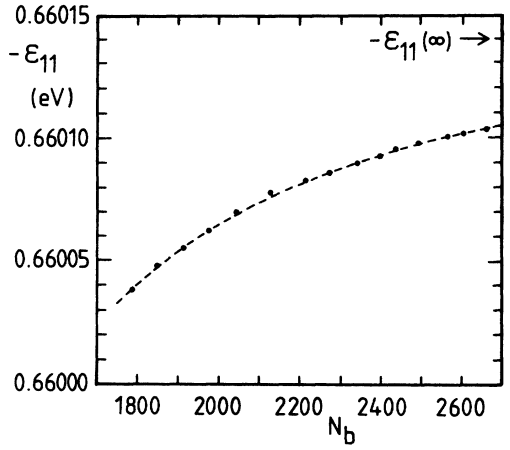

FIG. 2. Convergence of the calculated energy ε_{11} of the $dt\mu$ molecule with respect to the number of the basis functions N_b. The dashed curve corresponds to the function $\varepsilon_{11}(N_b)=\varepsilon_{11}(\infty)+CN_b^{-\alpha}$ with $\varepsilon_{11}(\infty)=-0.660\,14$ eV, $C=285\,00$ eV, and $\alpha=2.638$. Set I of the physical constants are used [for set II, all the dots and the dashed curve are to be shifted upward by 0.000 160 eV giving $\varepsilon_{11}(\infty)=-0.660\,30$ eV].

function[5,15] $\varepsilon_{11}(N_b)=\varepsilon_{11}(\infty)+CN_b^{-\alpha}$ with $\varepsilon_{11}(\infty)=-0.660\,14$ eV, $C=385\,00$ eV, and $\alpha=2.638$. Error in the estimation of $\varepsilon_{11}(\infty)$ is considered to be $\pm 0.000\,02$ eV.

In order to examine the role of the individual rearrangement channels, we made three *two*-channel calculations in which one of the $(t\mu)$-d, $(d\mu)$-t, and (dt)-μ channels is omitted from the full three-channel calculation with $N_b=2662$ in Table I, and we obtained $(\varepsilon_{11},\varepsilon_{10})=(+48.405,-188.546)$ eV, $(+0.370,-176.451)$ eV, and $(-0.646,-232.454)$ eV, respectively. It is interesting to see that the third calculation with the $(t\mu)$-d and $(d\mu)$-t channels can almost reproduce the energy of the full-channel calculation both for the weakly bound state and for the deeply bound one, although the least important (dt)-μ channel cannot be neglected to accomplish the required accuracy (10^{-3} eV) of ε_{11} and to determine precisely the wave function in the nuclear fusion region.

As far as the roles of l_c and L_c are concerned in the case of $J=1$ $(v=0,1)$, the angular momenta $(l_c,L_c)=(0,1)$ for $c=1$ and 2 are the most important, and the large angular momenta become the less important quickly, as expected. Contribution of the basis functions with $(l_c,L_c)=(3,4)$ and (4,3) $(c=1\sim 3)$ to the energy ε_{11} is roughly 5×10^{-5} eV in the case of $N_b=2662$ (Table I), and the role of the angular momenta larger than them is regarded to be included in the extrapolated value $\varepsilon_{11}(\infty)$.

The energies ε_{Jv} of the states with $J=0$ and 2 of the $dt\mu$ molecule were obtained, for set I of the physical constants, as $\varepsilon_{00}=-319.139\,606$ eV and $\varepsilon_{01}=-34.834\,372$ eV with $N_b=1442$ and $\varepsilon_{20}=-102.643\,337$ eV with $N_b=1566$. The values of ε_{Jv} are in agreement with those in Refs. 4–7 within 10^{-3} eV (ε_{20} is not given there). The calculated energies with the use of the most up-to-date set of the physical constants, set II, are listed in Table II.

IV. CONCLUDING REMARKS

The nonadiabatic coupled-rearrangement-channels method proposed here has been shown to be suited for describing the $dt\mu$ molecule with satisfactorily high accuracy.[18] Since the computation time is very short, an extensive optimization of the nonlinear variational parame-

TABLE I. Calculated energies ε_{11} and ε_{10} of the $dt\mu$ molecule for different numbers of the basis functions N_b. The numbers in parentheses are given by extrapolation. Set I of the physical constants is used. Some of literature data are also listed.

N_b	ε_{11} (eV)	ε_{10} (eV)
1789	−0.660 038	−232.471 435
1848	−0.660 048	−232.471 444
2044	−0.660 070	−232.471 478
2240	−0.660 084	−232.471 492
2438	−0.660 096	−232.471 501
2662	−0.660 104	−232.471 506
∞	(−0.660 14±0.000 02)	(−232.471 53±0.000 02)
	Literature for ε_{11}	
400[a]	−0.607 19 (−0.6554±0.0150)	
844[b]	−0.656	
1102[c]	−0.658 025	
1498[d]	−0.658 87 (−0.663±0.002)	
2084[e]	−0.659 68 (−0.6604±0.0002)	
3063[f]	−0.660 01 (−0.6601±0.0001)	

[a]Reference 15.
[b]Reference 9.
[c]Reference 6.
[d]Reference 4.
[e]Reference 5.
[f]Reference 7.

TABLE II. Calculated energies ε_{Jv} of the $dt\mu$ molecule by the present method with the use of set II, the 1986 CODATA-recommended set, of the physical constants. N_b is the number of the basis functions used.

	ε_{Jv} (eV)	N_b
$J=0,\ v=0$	−319.136 850	1442
$J=0,\ v=1$	−34.834 327	1442
$J=1,\ v=0$	−232.469 703	2662
$J=1,\ v=1$	−0.660 264[a]	2662
$J=2,\ v=0$	−102.642 886	1556

[a] $\varepsilon_{11}(\infty) = -0.660\,30 \pm 0.000\,02$ eV.

ters was performed for the important $J=v=1$ state, and $\varepsilon_{11}(\infty) = -0.660\,30 \pm 0.000\,02$ eV was obtained with the use of the most up-to-date set of the physical constants.

In order to guide the experiments related to ε_{11}, various kinds of corrections ($\gtrsim 10^{-3}$ eV) on ε_{11} are to be considered.[1] With the use of the present wave function for $J=v=1$, correction by the vacuum polarization was estimated[19] to be +0.0167 eV; calculation of the correction due to relativistic kinematics is in progress. Also in progress is a precise three-body calculation in which the diagonalization is made of the Hamiltonian including Coulomb potentials of the finite-size charges and a complex nuclear potential between d and t; the nuclear potential is determined from the $d+t \rightarrow {}^4\mathrm{He}+n$ reaction and therefore fusion rates of the molecular states can be given immediately from the imaginary part of the complex eigenenergies ε_{Jv}.

On account of the explicit use of the Jacobian coordinate system, the present methods are easily extended[10] to a nonadiabatic description of the $(d\mu)_{1s}+t \rightarrow (t\mu)_{1s}+d$ reaction with the correct boundary condition.

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[16] For example, nonlinear variational parameters for $J=1$ at $N_b=1789$ are as follows (lengths are in units of $a_\mu = \hbar^2/m_\mu e^2 \simeq 2.56\times 10^{-11}$ cm): As for the channels $c=1$ and 2, $(n,\bar{r}_1,\bar{r}_n;N,\bar{R}_1,\bar{R}_N) = (21,0.006,6;16,0.8,50)$, (9, 0.2, 4; 10, 1.2, 18), (8, 0.2, 4; 9, 1.2, 16), (7, 0.2, 4; 7, 1.2, 12), (7, 0.2, 4; 8, 1.2, 12), (7, 0.4, 4; 6, 1.2, 12), (4, 0.8, 4; 4, 1.6, 8) for $(l_c,L_c) = (0,1)$, (1,0), (1,2), (2,1), (2,3), (3,2), (3,4), respectively. As for the channel $c=3$, $(n,\bar{r}_1,\bar{r}_n;N,\bar{R}_1,\bar{R}_N) = (11,0.4,6;7,0.4,6)$, (11, 0.4, 6; 10, 0.4, 6), (10, 0.4, 6; 9, 0.4, 6), (8, 0.4, 6; 7, 0.4, 6), (8, 0.4, 6; 7, 0.4, 6), (8, 0.4, 6; 7, 0.4,6) for $(l_c,L_c) = (0,1)$, (1,0), (1,2), (2,1), (2,3), (3,2), respectively. This choice can be a guide for other J and for other molecules with N_b much reduced.

[17] Use of the same constants of Ref. 7 ($m_t = 5496.899 m_e$) gives $\varepsilon_{11} = -0.660\,078$ eV.

[18] The wave functions are available on request as well as the computational code to calculate the energy- and norm-matrix elements.

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