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Series of resonance states of muonic molecules

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Two infinite series of resonance states of muonic molecular ions $(pp\mu)^+$ and their isotopes converging to the dissociation limit $n=2$ are theoretically studied within nonrelativistic quantum mechanics. These resonance states are supported by the attractive long-range dipole potential originating from the linear Stark effect and behaving as R^{-2}. The positions ϵ_v $(v=0,1,2,\ldots)$ of the resonances relative to the dissociation threshold satisfy a simple power law $\epsilon_v = A\alpha^v$ for high v, the constant α being easily calculable in terms only of the total angular momentum and the masses of the nuclei and the muon. The size of a muonic molecule in a state ϵ_v is $\sim 10^{-8}$ cm$/[4\sqrt{|\epsilon_v \text{ (eV)}|}]$, which is comparable to (or even larger than) the size of ground-state electronic molecules if the resonance lies within a fraction of an electron volt from the dissociation threshold. The theory is useful in the classification of resonances in muonic molecules. The systems $(dt\mu)^+$ and $(dd\mu)^+$ are analyzed as examples.

I. INTRODUCTION

The d-t and d-d fusion catalyzed by muons without the need of high-temperature plasmas has been the subject of increasing experimental and theoretical interest in these years. It has been established experimentally that a single muon can catalyze d-t fusion about 150 times on the average during its lifetime in a dense mixture of deuterium and tritium under certain conditions.[1-5] In pure deuterium (without tritium), less efficient muon-catalyzed d-d fusion occurs. The clue to the mechanism of the catalysis is the formation of muonic molecular ions $(dt\mu)^+$ or $(dd\mu)^+$,[1,6,7] which are essentially Coulomb three-body systems similar to the hydrogen molecular ions $H_2{}^+$, $D_2{}^+$, HD^+, etc. The equilibrium internuclear distances R_e of the muonic molecules are smaller than the R_e of the electronic molecules by a factor of about 207. This greatly reduces the Coulomb barrier against nuclear fusion in muonic molecules compared with the barrier for electronic molecules, thus enhancing the intramolecular fusion rate by, for example, 95 orders of magnitude in going from DT^+ to $(dt\mu)^+$.[8] The high intramolecular fusion rates have stimulated extensive theoretical studies of bound states of muonic molecules.[7,9-12] Particular attention has been paid to weakly bound states, since they are considered to be formed efficiently in thermal collisions

$$d + t\mu \rightarrow (dt\mu)^+ + \Delta E \tag{1a}$$

or

$$d + d\mu \rightarrow (dd\mu)^+ + \Delta E \tag{1b}$$

between a deuteron in a deuterium and a $t\mu$ (or $d\mu$) atom in the ground $1s$ state. This is because the excess energy ΔE is small for weakly bound states of the muonic molecule, and is efficiently absorbed by the vibrational-rotational motion of a hydrogenlike electronic molecule $d^+(dt\mu)^+e^-e^-$ [or $d^+(dd\mu)^+e^-e^-$], in which the muonic molecular ion $(dt\mu)^+$ [or $(dd\mu)^+$] has a much smaller size than the electron orbitals and plays a role of a "pseudonucleus" in the electronic molecule.

Most $d\mu$ atoms and many $t\mu$ atoms are formed initially in highly excited states and are then deexcited down to lower and lower states. In a D_2-T_2 mixture, $t\mu$ atoms in the lower states are also formed in the muon-transfer processes

$$t + d\mu \rightarrow d + t\mu \ , \tag{2}$$

which appear to occur with a higher rate for excited states than for the ground state.[13] Therefore, many $t\mu$ and $d\mu$ atoms are expected to be in the metastable $2s$ state for quite a long time. An interesting question to be posed, then, is whether processes (1) occur with a high rate when the $t\mu$ (or $d\mu$) atom is in the $2s$ or $2p$ state. If they do, they may play an important role in the muon-catalyzed fusion.

For processes (1) with $t\mu$ $(n=2)$ or $d\mu$ $(n=2)$ to occur appreciably, the excess energy ΔE must be small enough to be transferred efficiently to some degrees of freedom of motion, such as the vibrational-rotational motion of an electronic molecule, just as in processes (1) with ground-state atoms. In other words, the $(dt\mu)^+$ or $(dd\mu)^+$ molecule must have a level or levels lying close to the dissociation limit $d+t\mu$ $(n=2)$ or $d+d\mu$ $(n=2)$. Such levels are autodissociating or resonance states, because they are embedded in the continuum above the dissociation limit $d+t\mu$ $(n=1)$ or $d+d\mu$ $(n=1)$.

The purpose of this paper is to discuss the possibility of the existence of resonance states of muonic molecules that might be relevant to muon-catalyzed fusion. In fact, an infinite number of resonance levels that converge to the dissociation limit $n=2$ exist within the framework of the nonrelativistic quantum mechanics. This is shown in Sec. II in the Born-Oppenheimer approximation and in

Sec. III without this approximation. Then, in Sec. IV, the theory is applied to analyze the recent results of Ref. 14 on resonance states of the $(dd\mu)^+$ and $(dt\mu)^+$ molecules. The nonrelativistic quantum mechanics is not completely satisfactory for studying the resonance levels of real muonic molecules that lie close to the dissociation limit $n=2$, and the corrections that must be taken into account are discussed in Sec. V.

Resonance states of the $(dt\mu)^+$ molecule are also studied in Ref. 15. These states are reported to lie several electron volts above the $d\mu$ $(n=1)$ threshold, i.e., far below the $t\mu$ $(n=2)$ threshold, and appear to be formed by a mechanism different from that discussed in this paper.

II. BORN-OPPENHEIMER APPROXIMATION

The Born-Oppenheimer approximation is quite good for low-lying electronic states, but is numerically inaccurate for muonic molecules. Nevertheless, it is sometimes useful as a zeroth-order approximation for understanding the basic physics of muonic systems,[8] provided that one is well aware of its limitation. For the purpose of the present paper the inspection of the vibrational-rotational levels (v,N) of electronic states of $H_2{}^+$ is instructive. In the Born-Oppenheimer approximation each potential-energy curve $V_\mu(R)$ of the muonic molecules $(pp\mu)^+$, $(pd\mu)^+$, $(dd\mu)^+$, $(dt\mu)^+$, etc., as a function of the internuclear distance R is equivalent to a potential-energy curve $V_e(R)$ of the electronic molecule $H_2{}^+$, if the muon atomic units are used for the former and the electron atomic units are used for the latter. In other words the two potentials are related to each other by

$$V_\mu(R)=\frac{m_\mu}{m_e}V_e\left[\frac{m_\mu}{m_e}R\right] \tag{3}$$

in terms of the muon-to-electron mass ratio $m_\mu/m_e=206.768\,26$.[16] The absolute value of a muonic potential is larger than the corresponding electronic potential by the factor m_μ/m_e, and the range is smaller by m_e/m_μ. Since the strength of a potential is roughly proportional to the absolute value times the square of the range, muonic potentials are much weaker than electronic potentials. This explains the fact that much less vibrational levels are supported by the ground $1s\sigma_g$ muonic potential than by the corresponding electronic potential.

Our present concern is about possible muonic molecular states lying close to the dissociation limit $n=2$. Therefore, attention is paid to those $H_2{}^+$ potential curves that correlate with the $2s$ and $2p$ states of the H atom in the separated-atom limit $R\rightarrow\infty$;[17,18] see Fig. 1. There are six of them, of which, two repulsive curves $2s\sigma_g$ and $3p\sigma_u$ behave asymptotically as $V_e(R)\sim -0.125+3/R^2$ a.u., and another repulsive curve $3d\pi_g$ as $V_e(R)\sim -0.125+O(R^{-3})$ a.u. The other three curves, namely, the $2p\pi_u$, $3d\sigma_g$, and $4f\sigma_u$ potential energy curves are attractive and support vibrational levels.

The $2p\pi_u$ curve has a weak maximum at $R\simeq 26$ a.u., behaves asymptotically as $V_e(R)\sim -0.125+6/R^3$ a.u., and correlates with the separated-atom state H$(2p)$. For a rotational-angular-momentum quantum number N of unity, this potential supports a resonance state above the dissociation limit -0.125 a.u. and 12 vibrational states below it.[19] For the $(pp\mu)^+$, $(pd\mu)^+$, $(pt\mu)^+$, $(dd\mu)^+$, $(dt\mu)^+$, and $(tt\mu)^+$ molecules this potential supports one even-parity bound state with $N=1$ and one odd-parity vibrational state with $N=0$ [except for $(tt\mu)^+$, for which there are two vibrational states with $N=0$] in the Born-Oppenheimer approximation.[20] Recent nonadiabatic, completely three-body calculations also show that each of these molecules has one even-parity bound state with a total-angular-momentum quantum number J of unity.[14] This bound state is interpreted to be associated with the adiabatic $2p\pi_u$ potential.

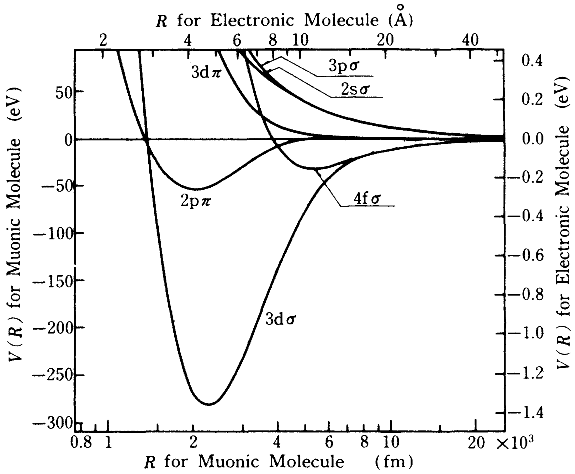

FIG. 1. The adiabatic potential energies, including the internuclear Coulomb repulsion, of the $H_2{}^+$ molecule and its isotopes converging to the dissociation limit $n=2$. The energy scale shown on the right-hand side and the internuclear-distance scale shown on top are for electronic molecules, and the scales on the left-hand side and on the bottom are for muonic molecules. The energies are measured relative to the dissociation limit $n=2$. For homonuclear molecules the $3d\sigma$, $3d\pi$, and $2s\sigma$ states are gerade states and the $2p\pi$, $4f\sigma$, and $3p\sigma$ states are ungerade states.

The $3d\sigma_g$ and $4f\sigma_u$ curves have no maximum, behave asymptotically as $V_e(R)\sim -0.125-3/R^2$ a.u., and correlate with linear combinations of the separated-atom states H$(2s)$ and H$(2p)$.[21] For muonic molecules these potentials behave asymptotically as

$$V_\mu(R)\sim -0.125\frac{m_\mu}{m_e}-3\frac{m_e}{m_\mu}R^{-2} \tag{4}$$

in atomic units according to Eq. (3).

A particle having a mass m and an angular-momentum quantum number l has an infinite number of bound states in a centrally symmetric potential that decays asymptotically as $-(\hbar^2/2m)\gamma r^{-2}$, if $\gamma>(l+\frac{1}{2})^2$.[22] This holds true regardless of the form of the potential at smaller distances; only the asymptotic behavior is essential. It follows from this that the $3d\sigma_g$ and $4f\sigma_u$ curves of $H_2{}^+$

TABLE I. Critical angular momentum J_c (or N_c) such that, for $J \le J_c$ (or for $N \le N_c$), there exist an infinite number of resonance states below the dissociation threshold $n=2$. BO: Born-Oppenheimer approximation, for which N and N_c are rotational-angular-momentum quantum numbers. Three-body: asymptotic full three-body treatment of Sec. III, for which J and J_c are total-angular-momentum quantum numbers.

	BO N_c	Three-body Dissociation limit	J_c
$(pp\mu)^+$	4	$p+p\mu$ $(n=2)$	5
$(pd\mu)^+$	5	$p+d\mu$ $(n=2)$	5
		$d+p\mu$ $(n=2)$	6
$(pt\mu)^+$	5	$p+t\mu$ $(n=2)$	6
		$t+p\mu$ $(n=2)$	6
$(dd\mu)^+$	6	$d+d\mu$ $(n=2)$	7
$(dt\mu)^+$	7	$d+t\mu$ $(n=2)$	7
		$t+d\mu$ $(n=2)$	7
$(tt\mu)^+$	8	$t+t\mu$ $(n=2)$	8
$(ppe^-)^+$	73	$p+H$ $(n=2)$	73
$(pde^-)^+$	85	$p+D$ $(n=2)$	85
		$d+H$ $(n=2)$	85
$(pte^-)^+$	90	$p+T$ $(n=2)$	90
		$t+H$ $(n=2)$	90
$(dde^-)^+$	104	$d+D$ $(n=2)$	104
$(dte^-)^+$	114	$d+T$ $(n=2)$	114
		$t+D$ $(n=2)$	114
$(tte^-)^+$	127	$t+T$ $(n=2)$	127

support an infinite number of vibrational levels for any $N \le 73$.[21] For muonic molecules in the Born-Oppenheimer approximation these two potential curves support an infinite number of vibrational levels only for smaller N, e.g., for $N \le 4$ for $(pp\mu)^+$; see Table I for the critical values of N for the six muonic molecules and for six electronic molecules. Recent nonadiabatic three-body calculations indicate that the $(dd\mu)^+$ and $(dt\mu)^+$ molecules have about ten levels with $J=0$ and about ten with $J=1$ associated with the adiabatic potentials $3d\sigma_g$ and $4f\sigma_u$.[14] These levels are discussed in Sec. IV.

III. ASYMPTOTIC FULL THREE-BODY PROBLEM

We now consider the full three-body problem without the Born-Oppenheimer approximation. The region of the configuration space that is most crucial to our arguments is the asymptotic region where particle 3 with positive charge $+e$ and mass M_3 lies far from the atom consisting of particle 1 with charge $+e$ and mass M_1 and particle 2 with charge $-e$ and mass M_2; $-e$ is the charge of an electron. We define a set of Jacobi coordinates $(\mathbf{r},\mathbf{R})$, $\mathbf{r}$ being the position vector of particle 2 relative to particle 1, and $\mathbf{R}$ being that of particle 3 relative to the center of mass of the atom (1,2).

Let $\mathbf{J}\hbar$ and $M\hbar$ be the total angular momentum and its z component. To describe the angular part of the wave function of the three-body system we form eigenfunctions $\mathcal{Y}_{ll'}^{(JM)}(\hat{\mathbf{r}},\hat{\mathbf{R}})$ of $(\mathbf{J}\hbar)^2$ and $M\hbar$ with eigenvalues $J(J+1)\hbar^2$ and $M\hbar$ from spherical harmonics $Y_{lm}(\hat{\mathbf{r}})$ and $Y_{l'm'}(\hat{\mathbf{R}})$. The space-part wave function $\Psi^{(JM)}(1,2,3)$ of the three-body system may be expanded in terms of these angular functions and the radial wave functions $\psi_{nl}(r)$ of the atom (1,2) as

$$\Psi^{(JM)}(1,2,3)=\sum_{n,l,l'} F_{nll'}^{(J)}(R)\psi_{nl}(r)\mathcal{Y}_{ll'}^{(JM)}(\hat{\mathbf{r}},\hat{\mathbf{R}}) \ . \tag{5}$$

For energies close to the level $n=2$ of atom (1,2), the only channels in expansion (5) that are important in the asymptotic or large-R region are those with $n=2$, namely, channel 1 with $(n,l,l')=(2,0,J)$, channel 2 with $(2,1,J-1)$, and channel 3 with $(2,1,J+1)$.[23] (Channel 2 vanishes for $J=0$, leaving only two important channels.) Substituting Eq. (5) with only these three channels into the Schrödinger equation for the three-body system, projecting the Schrödinger equation onto the angular functions and the atomic radial wave functions, expanding the potential matrix in terms of inverse powers of R, and retaining only the leading term, we obtain three coupled equations

$$\frac{\hbar^2}{2\mu_{3,12}}\left[\frac{d^2}{dR^2}+k^2\right]\mathbf{F}=\frac{\hbar^2}{2\mu_{3,12}}\frac{\Lambda^2}{R^2}\mathbf{F} \quad (\text{for } R \ge a) \tag{6}$$

with

$$\Lambda^2=\begin{bmatrix} J(J+1) & \beta\sqrt{J/(2J+1)} & -\beta\sqrt{(J+1)/(2J+1)} \\ \beta\sqrt{J/(2J+1)} & J(J-1) & 0 \\ -\beta\sqrt{(J+1)/(2J+1)} & 0 & (J+1)(J+2) \end{bmatrix} , \tag{7}$$

$$\beta=\frac{6\mu_{3,12}}{\mu_{12}} , \tag{8}$$

$$\mu_{3,12}=\frac{M_3(M_1+M_2)}{M_1+M_2+M_3} , \tag{9}$$

and

$$\mu_{12}=\frac{M_1M_2}{M_1+M_2} , \tag{10}$$

where $(\hbar k)^2/2\mu_{3,12}$ $(=\epsilon)$ is the energy of the three-body system relative to the level $n=2$ of atom (1,2), and a is some large distance at and beyond which the terms in the potential of higher order than R^{-2} are negligible. This is straightforward extension of the formulation for electron-hydrogen scattering developed in Ref. 23.

The diagonal elements of Λ^2 represent the usual centrifugal potential. The off-diagonal elements represent the dipole potential that couples the $2s$ channel with the $2p$ channels. This long-range coupling potential arises from the leading term in the asymptotic expansion of the sum of the positive Coulomb potential $e^2/|\mathbf{r}_1-\mathbf{r}_3|$ between particles 1 and 3 and the negative Coulomb potential $-e^2/|\mathbf{r}_2-\mathbf{r}_3|$ between particles 2 and 3.

Equation (6) may be diagonalized by a unitary transformation of the channels. This leaves three uncoupled single-channel equations with potentials $\lambda_i(\lambda_i+1)/R^2$ $(i=1,2,3)$ in units of $\hbar^2/2\mu_{3,12}$, where[23]

$$\begin{aligned}\lambda_1(\lambda_1+1)&=J(J+1)\ ,\\ \lambda_2(\lambda_2+1)&=J^2+J+1-(2J+1)x\ ,\\ \lambda_3(\lambda_3+1)&=J^2+J+1+(2J+1)x\ ,\end{aligned} \tag{11}$$

or

$$\begin{aligned}\lambda_1&=J\ ,\\ \lambda_2&=-\tfrac{1}{2}+[(J-\tfrac{1}{2})^2-(2J+1)(x-1)]^{1/2}\ ,\\ \lambda_3&=-\tfrac{1}{2}+[(J+\tfrac{3}{2})^2+(2J+1)(x-1)]^{1/2}\ ,\end{aligned} \tag{12}$$

with

$$x=\left[\left[\frac{\beta}{2J+1}\right]^2+1\right]^{1/2}\ . \tag{13}$$

For the $\mathrm{H_2}^+$ ion with $J=0$ the asymptotic potential

$$(\hbar^2/2\mu_{3,12})\lambda_i(\lambda_i+1)R^{-2}$$

is 0 for $i=1$, $-[3+O(\delta)]R^{-2}$ a.u. for $i=2$, and $[3+O(\delta)]R^{-2}$ a.u. for $i=3$, where δ is the electron-to-proton mass ratio m_e/m_p, in agreement with the consequences of the Born-Oppenheimer approximation explained in Sec. II. In particular, the transformed channel $i=2$ with the gerade property corresponds to the adiabatic potential curve $3d\sigma_g$, and that with the ungerade property to the potential $4f\sigma_u$.

The condition for the existence of an infinite number of states in the potential

$$(\hbar^2/2\mu_{3,12})\lambda_i(\lambda_i+1)R^{-2}$$

is

$$(\lambda_i+\tfrac{1}{2})^2<0 \tag{14}$$

according to the rule mentioned in the last paragraph of Sec. II. This condition is satisfied by channel $i=2$, if

$$\beta^2>(J^2+3J+\tfrac{9}{4})(J^2-J+\tfrac{1}{4})\ . \tag{15}$$

This is consistent with the result of the Born-Oppenheimer approximation that the adiabatic potentials $3d\sigma_g$ and $4f\sigma_u$ of $\mathrm{H_2}^+$ support an infinite number of vibrational levels for any $N\le 73$, because

$$\beta^2=(3m_p/m_e)^2[1+O(\delta)]\simeq 3.03\times 10^7\ .$$

For the muonic molecules, β^2 is much smaller. Thus, the critical J at and below which the muonic molecule $(pp\mu)^+$, for example, has an infinite number of states below the threshold $n=2$ is five in the three-channel approximation of Eq. (6). Table I includes the critical J for six muonic and six electronic molecules.

The wave functions $G(R)$ of these states in channel $i=2$ satisfy the single-channel Schrödinger equation

$$\left[\frac{d^2}{dR^2}-\kappa^2\right]G(R)=\frac{\lambda_2(\lambda_2+1)}{R^2}G(R)\quad (R\ge a)\ , \tag{16}$$

where the binding energy $|\epsilon|$ has been expressed as $(\hbar\kappa)^2/2\mu_{3,12}$. The mock angular momentum λ_2 may be written as $-\frac{1}{2}+i\nu$ with a real constant ν according to the condition (14). The physical solution to Eq. (16) is that linear combination of two linearly independent solutions which decays to zero at infinity, and is expressible as

$$G(R)=\text{const}\times\sqrt{R}\,K_{i\nu}(\kappa R)\quad (R\ge a) \tag{17}$$

in terms of a modified Bessel function $K_{i\nu}$ of the second kind and of imaginary order.

Now we consider the $\mathcal{R}$ matrix at $R=a$ to obtain the energy eigenvalues ϵ_v, and hence κ_v^2, and the corresponding wave functions $G_v(R)$. Here, v is an integer to label the full three-body states that converge to the dissociation limit $n=2$, and has the meaning of the vibrational quantum number if M_2 is much smaller than M_1 and M_3. In the following we sometimes refer to v as the vibrational quantum number and the corresponding state a vibrational state for convenience, even when the Born-Oppenheimer approximation breaks down. The $\mathcal{R}$ matrix is defined by the full three-body wave function in the inner region $R\le a$. Because only the long-range potentials behaving as $\sim R^{-2}$ are important near $R=a$ (and beyond) in the energy region of interest, and because of the uncoupling discussed above (which is valid also for the inner-region wave function close to $R=a$), we consider only the diagonal element $\mathcal{R}_{22}(\kappa_v;a)$ of the $\mathcal{R}$ matrix for the transformed channel $i=2$. For smooth connection of the inner-region wave function to the outer-region wave function $G_v(R)$, the $\mathcal{R}$-matrix element $\mathcal{R}_{22}(\kappa_v;a)$ must satisfy the relation

$$\mathcal{R}_{22}(\kappa_v;a)=G_v(a)/[aG_v'(a)]\ . \tag{18}$$

It follows from the power-series expansion of the modified Bessel function that Eq. (18) may be rewritten as

$$[\mathcal{R}_{22}(\kappa_v;a)]^{-1}=\tfrac{1}{2}+\frac{\sum_m p_v^{2m}\mathrm{Im}[(2m+i\nu)F_{vm}]}{\sum_m p_v^{2m}\mathrm{Im}F_{vm}}\ , \tag{19}$$

where

$$p_v=(\kappa_v a)/2\ , \tag{20}$$

$$F_{vm}=\frac{\exp[i(\nu\ln p_v-\eta)]}{m!|\Gamma(m+1+i\nu)|}\ , \tag{21}$$

$\Gamma(m+1+i\nu)$ is a Γ function, η is its phase, and the summation in Eq. (19) over m is taken from zero to infinity.

Because we are considering only a small region of energy close to the level $n=2$ of atom (1,2), we may neglect the dependence of the $\mathcal{R}$ matrix on the energy; in general, the $\mathcal{R}$ matrix varies smoothly across the threshold for a new channel. For Eq. (19) to be unchanged with v for small p_v, it must satisfy the relation

$$-\nu \ln p_v = v\pi + \text{const} . \qquad (22)$$

In other words, the binding energies $|\epsilon_v|$ must satisfy the relation

$$\ln|\epsilon_v| = -2v\pi/\nu + \text{const} . \qquad (23)$$

Therefore, the level positions $E_{n=2}+\epsilon_v$ converge to the $n=2$ dissociation limit $E_{n=2}$ as $v \to \infty$ as

$$E_{n=2}+\epsilon_v = E_{n=2} + A\alpha^v , \qquad (24a)$$

where

$$\alpha = \exp[-2\pi/\nu] < 1 . \qquad (24b)$$

IV. RESONANCES IN MUONIC MOLECULES

When the coupling with the open $1s$ channel is taken into account, the bound vibrational states discussed in the preceding section change into resonance states. Their energy positions and the dominant part of the wave functions, however, are hardly affected by this coupling for the following reasons. This coupling is extremely weak, since the spacing between the levels $n=1$ and 2 of atom (1,2) is large, for example, ~ 2 keV for muonic atoms $p\mu$, $d\mu$, and $t\mu$. The coupling remains weak relative to $|\epsilon_v|$ even for high v; the probability distribution of the wave function $G_v(R)$ is concentrated close to the classical turning point R_x, which is proportional to $|\epsilon_v|^{-1/2}$ (see the last paragraph of this section), so that the strength of the $1s$-$2p$ coupling potential behaving asymptotically as $\sim R^{-2}$ is effectively proportional to $|\epsilon_v|$. The $1s$-$2s$ coupling potential is of short range and is negligible for high v.

These considerations justify application of relation (24) to the infinity of resonances lying below the level $n=2$. This result is consistent with the result of Ref. 24, which discusses the resonances in scattering of electrons by hydrogen atoms by studying the dependence of the K matrix on the scattering energy. The theory developed so far in this paper is a generalization of the theory of Ref. 24 for arbitrary masses of the three particles by use of the knowledge of the wave functions themselves. It also follows from a generalization of Ref. 24 that the width of the resonance decreases with v with the same ratio $\exp[-2\pi/\nu]$ as the resonance position, and that neighboring resonances do not overlap for any high v.

For homonuclear molecules (1,2,3) there are two infinite series of resonances converging to the same limit, namely, the gerade series (which is supported by the adiabatic $3d\sigma_g$ potential if M_2 is much smaller than M_1 and M_3) and the ungerade series (supported by the $4f\sigma_u$ potential if $M_2 \ll M_1, M_3$). These two series are uncoupled to each other because of the well-defined gerade-ungerade property.

For heteronuclear molecules (1,2,3), for which no rigorous gerade-ungerade property exists, there are two closely spaced dissociation limits $3+(1,2)$ ($n=2$) and $1+(2,3)$ ($n=2$). If $M_1 > M_3$, the former lies below the latter. The spacing between the two dissociation limits is 33.677 eV for $(pd\mu)^+$, 45.687 eV for $(pt\mu)^+$, and 12.010 eV for $(dt\mu)^+$. There exists an infinite series of resonances converging to each dissociation limit. The lower and upper series may sometimes overlap in terms of energy. However, they are almost uncoupled to each other, because, for high-lying resonance states, the effective internuclear distance is so large that the overlap between the wave functions of the two different arrangement channels is negligible. It is instructive to note that, if M_2 is much smaller than M_1 and M_3, the resonances belonging to the lower series are supported by the (almost gerade) adiabatic $3d\sigma$ potential, and those belonging to the upper series are supported by the (almost ungerade) adiabatic $4f\sigma$ potential. Even when the Born-Oppenheimer approximation is inaccurate, as with the muonic molecules, an approximate gerade-ungerade property is still expected to be preserved with the inner (or small-R) part of the wave functions for resonances lying not too close to their series limit. This is because the difference between M_1 and M_3 has a relatively weak effect on the wave function when particle 2 interacts strongly with *both* nuclei having the same charge. Therefore, a resonance having a dominant gerade character, if any, may be classified into the lower series, and that having a dominant ungerade character into the upper series.

Table II shows the values of the parameters $E_{n=2}$ and

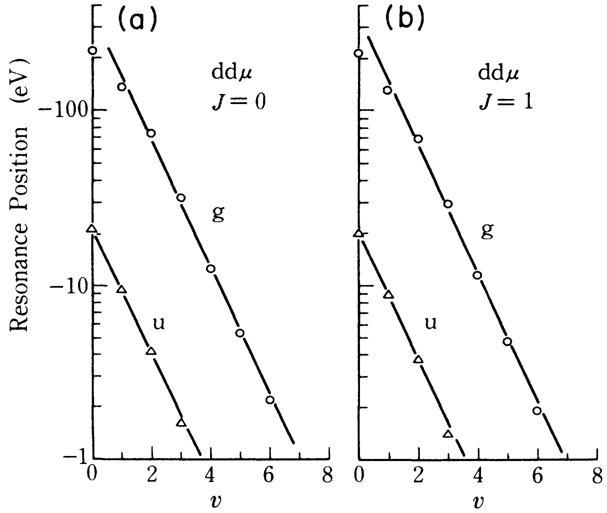

FIG. 2. Resonance energies of the $(dd\mu)^+$ molecules relative to the level $n=2$ of the $d\mu$ atom. v, "vibrational" quantum number; J, total-angular-momentum quantum number; g, gerade; u, ungerade; ○ and △, results of the full three-body calculations of Ref. 14; ——, Eq. (24) with the parameters given in Table II.

α in Eq. (24a) for six muonic molecules calculated with the masses of particles taken from Refs. 16 and 25. This table also includes the coefficient of the long-range potential proportional to R^{-2} both in the Born-Oppenheimer approximation and in the asymptotic three-body formulation. Even for the lightest isotope $(pp\mu)^+$ the adiabatic potentials relative to the adiabatic dissociation limit are seen to be a fairly good approximation to the asymptotic potential in the channel $i=2$ relative to its dissociation limit.

Equation (24a) with the values of α given in Table II may be tested with the results of Ref. 14, and the constant A may be determined. In this reference full three-body Hamiltonian matrices are calculated mainly for the $(dd\mu)^+$ and $(dt\mu)^+$ systems using the spheroidal coordinates. The eigenvalues of Hamiltonian matrices of some different sizes are compared, and stable ones are chosen in a manner similar to the stabilization method that was devised to calculate resonance energies in atomic physics.[26,27] The eigenvalues chosen in this manner are assumed to represent resonance positions.

The positions of all the gerade resonance levels of the $(dd\mu)^+$ molecule with $J=0$ and 1 calculated in Ref. 14 are plotted in Fig. 2. As for the ungerade resonances, the lowest one with $J=1$ is reported to have a dominant π_u character and to be associated with the $2p\pi_u$ potential curve, whereas the other ones that are calculated are reported to have a dominant σ_u character. This strongly suggests that only the latter resonances belong to the series in the channel $i=2$ that converges to the dissociation limit $n=2$. Therefore, the plot of ungerade resonances in Fig. 2 excludes the lowest resonance with $J=1$ and adopts renumbered v.

Each straight line in Fig. 2 has a slope determined by the value of α defined by Eq. (24b) and given in Table II. Only the height is adjusted to fit each line to the resonance positions. The constant A in Eq. (24a) determined in this way is included in Table II. In this fitting procedure the deeply bound resonances are neglected, because they are considered not to satisfy the approxima-

TABLE II. Parameters for the asymptotic potentials $E_{n=2}+C/R^2$ and for the resonance positions $E_{n=2}+\epsilon_v=E_{n=2}+A\alpha^v$ for muonic molecules.

System	Dissociation limit	$E_{n=2}$[a] (eV)	Angular momentum J or N	C BO[b] (eV Å^2)	C Three-body[c] (eV Å^2)	α	A gerade[d] (eV)	A ungerade[d] (eV)
$(pp\mu)^+$	$p+p\mu$ $(n=2)$	−632.124	0	−0.1106	−0.1191	0.3175		
			1	−0.1023	−0.1118	0.3057		
			2	−0.0857	−0.0970	0.2800		
$(pd\mu)^+$	$p+d\mu$ $(n=2)$	−665.800	0	−0.1106	−0.1138	0.3557		
			1	−0.1043	−0.1080	0.3461		
			2	−0.0919	− 0.0964	0.3251		
$(pd\mu)^+$	$d+p\mu$ $(n=2)$	−632.124	0	−0.1106	−0.1201	0.3754		
			1	−0.1043	−0.1146	0.3667		
			2	−0.0919	−0.1035	0.3479		
$(pt\mu)^+$	$p+t\mu$ $(n=2)$	−677.811	0	−0.1106	−0.1120	0.3730		
			1	−0.1050	−0.1068	0.3641		
			2	−0.0939	−0.0963	0.3451		
$(pt\mu)^+$	$t+p\mu$ $(n=2)$	−632.124	0	−0.1106	−0.1205	0.3993		
			1	−0.1050	−0.1156	0.3917		
			2	−0.0939	−0.1058	0.3753		
$(dd\mu)^+$	$d+d\mu$ $(n=2)$	−665.800	0	−0.1106	−0.1148	0.4337	−360	−21.7
			1	−0.1064	−0.1109	0.4274	−345	−20.6
			2	−0.0981	−0.1031	0.4140		
$(dt\mu)^+$	$d+t\mu$ $(n=2)$	−677.811	0	−0.1106	−0.1130	0.4614	−375	
			1	−0.1071	−0.1097	0.4561	−360	
			2	−0.1002	−0.1031	0.4449		
$(dt\mu)^+$	$t+d\mu$ $(n=2)$	−665.800	0	−0.1106	−0.1151	0.4679		(−23.9)[e]
			1	−0.1071	−0.1119	0.4628		(−23.0)[e]
			2	−0.1002	−0.1054	0.4520		
$(tt\mu)^+$	$t+t\mu$ $(n=2)$	−677.811	0	−0.1106	−0.1134	0.5018		
			1	−0.1078	−0.1107	0.4977		
			2	−0.1022	−0.1054	0.4891		

[a] The atomic unit of energy is 27.211 396 eV (Ref. 16). The masses of p, d, t, and μ are taken from Refs. 16 and 25.

[b] Sum of the asymptotic form (4) of the adiabatic potential and the centrifugal potential with the angular momentum $N\hbar$ for the relative motion between nuclei 1 and 3.

[c] Asymptotic form of the potential in the transformed channel $i=2$ with the total angular momentum $J\hbar$.

[d] Determined by fitting to the resonance energies given in Ref. 14.

[e] Determined from a single data point as is explained in the text.

tion made in Sec. III. In particular, they are supported by that part of the potential which is weaker than the dipole potential behaving as $\sim R^{-2}$, so that the data points representing their positions lie below the straight lines in Fig. 2. In fact, the straight lines reproduce well even the resonances as far as $\sim 10^2$ eV from the series limit, although the semilogarithmic plot gives an impression of better agreement than a linear plot. The position of the calculated highest-lying (largest-v) resonance in each series has not completely converged.[14] Unconverged eigenvalues are known empirically to lie often, but not always, above the true resonance positions, the more in error for the higher resonances, although there is no upper-bound principle for resonance energies of Ref. 14. This empirical rule is consistent with the result that the calculated resonance positions close to the series limit lie slightly below the straight lines in Fig. 2.

In plotting the resonances in the $(dt\mu)^+$ molecule information on the approximate symmetry property of the wave functions is particularly useful. Among the nine reported resonances with $J=0$ the sixth [lying at 11.41 eV below the level $t\mu$ $(n=2)$, or at 23.42 eV below the level $d\mu$ $(n=2)$] has a large ungerade component, whereas the other resonances have a large gerade component except for the highest resonance, which exhibits strong g-u mixing.[14] This suggests the following. First, the sixth resonance is associated with the adiabatic $4f\sigma$ potential, and belongs to the upper series of resonances converging to the limit $d\mu$ $(n=2)$. Second, all the other resonances but the highest are associated with the adiabatic $3d\sigma$ potential, and belong to the lower series converging to the limit $t\mu$ $(n=2)$. Third, appropriate classification of the highest calculated resonance is unknown from the wave function only. On the temporary assumption that it also belongs to the lower series, all the calculated members of this series are plotted in Fig. 3(a) just like the plot in Fig. 2. A straight line with a slope determined by α in Table II accurately reproduces most resonance positions including the highest. This justifies the assumed classification. The constant A is determined by fitting the straight line to the calculated resonance positions.

Only a single resonance that appears to belong to the upper series is known. Therefore, rather than testing Eq. (24) on it, we may assume this resonance to be the lowest member of the upper series satisfying Eq. (24), and may examine if it is reasonable. We note that the low-v resonances in the lower series of the $(dt\mu)^+$ molecule have values of ϵ_v similar to, but slightly lower than, those of the gerade resonances of the $(dd\mu)^+$ molecule with the same v. We expect similarity between the upper series of the $(dt\mu)^+$ molecule and the ungerade series of the $(dd\mu)^+$ molecule. Indeed, the value -23.42 eV relative to the level $d\mu$ $(n=2)$ for the $(dt\mu)^+$ resonance in question is close to and slightly lower than the values -21.16 eV $(J=0)$ and -20.12 eV $(J=1)$ for the lowest ungerade $(dd\mu)^+$ resonances. We also note that all the ungerade resonances in Fig. 2 of the $(dd\mu)^+$ molecule with $J=0$ and 1 including the lowest satisfy Eq. (24) well, but that the best A values estimated are slightly (by about 0.5 eV) lower than ϵ_0. This suggests a preliminary estimate of A of -23.9 eV for the upper series of the $(dt\mu)^+$ molecule. Then the second member of this series would be at 11.2 eV below the level $d\mu$ $(n=2)$, or at 0.8 eV *above* the level $t\mu$ $(n=2)$, which is in the region of energy where stable eigenvalues of the Hamiltonian matrices are not carefully searched for in Ref. 14. This must be the reason why only one resonance is found to have a large ungerade component. The highest calculated resonance with strong u-g mixing lies far from any value satisfying Eq. (24) for the upper series.

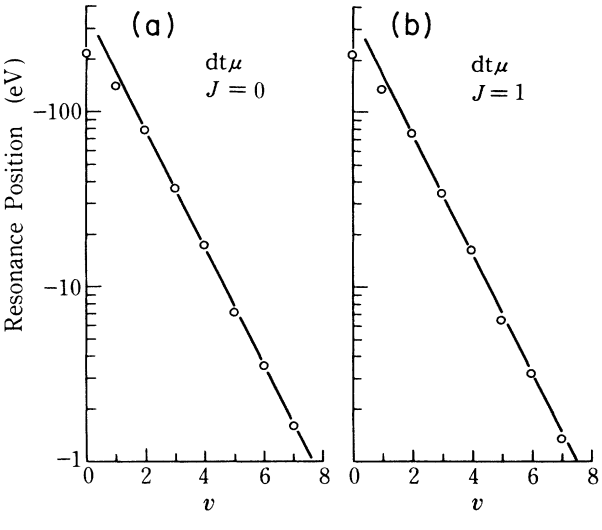

FIG. 3. Resonance energies of the $(dt\mu)^+$ molecules relative to the level $n=2$ of the $t\mu$ atom. v, "vibrational" quantum number; J, total-angular-momentum quantum number; ○, results of the full three-body calculations of Ref. 14; ——, Eq. (24) with the parameters given in Table II.

Ten resonances are found in Ref. 14 for the $(dt\mu)^+$ system with $J=1$. Among them the fifth, lying at 19.16 eV below the level $t\mu$ $(n=2)$, has a large π_u component and is strongly related to the even-parity bound state associated with the $2p\pi_u$ adiabatic potential. The other nine resonances may be analyzed in a manner similar to the analysis for $J=0$. The seventh resonance, lying at 10.51 eV below the level $t\mu$ $(n=2)$, has a large ungerade component. Being close to ϵ_0 of the other three σ_u-type series of the $(dd\mu)^+$ and $(dt\mu)^+$ systems, the level position -22.52 eV relative to the series limit $d\mu$ $(n=2)$ is reasonable as the lowest member of the upper series. A value of A of -23.0 eV is estimated in the same way as for $J=0$. This leads to an estimate of the second resonance level of -10.6 eV relative to $d\mu$ $(n=2)$, or $+1.4$ eV relative to $t\mu$ $(n=2)$, lying outside of the energy region where stable eigenvalues are carefully searched for. The eight resonances in the lower series are plotted in Fig. 3(b), which is similar to Fig. 3(a).

Once A and α are determined for a particular series, the positions of high-lying resonances follow immediately from Eq. (24), and the corresponding channel wave functions $G_v(R)$ are easily calculable from Eq. (17). Figure 4 shows examples of the $(dt\mu)^+$ resonances in the lower series with $J=0$. The maximum amplitude of each wave function is normalized to a constant. The resonance posi-

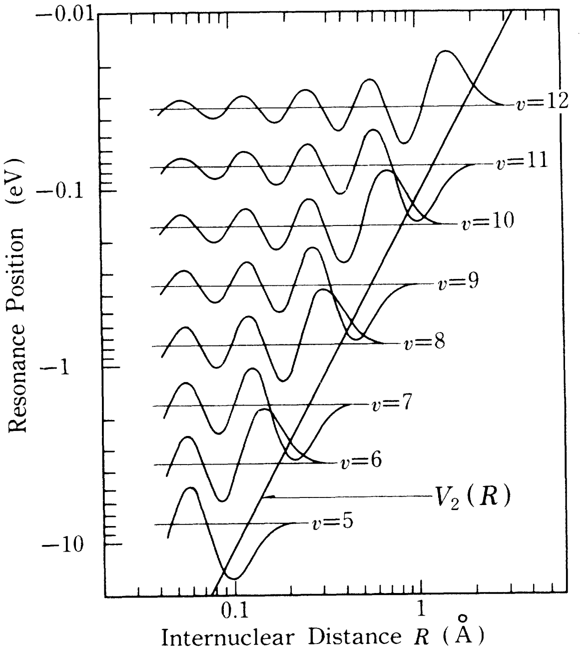

FIG. 4. Resonance levels ϵ_v of the $(dt\mu)^+$ molecule relative to the level $n=2$ of the $t\mu$ atom and their channel wave functions $G_v(R)$. The total angular momentum is zero and the "vibrational" quantum number v is from 5 to 12. Each horizontal line represents both a resonance position and the internuclear-distance axis for the plot of $G_v(R)$; the values of $G_v(R)$ are plotted to a linear scale. $V_2(R)$ is the asymptotic potential $(\hbar^2/2\mu_{3,12})\lambda_2(\lambda_2+1)R^{-2}$ for $G_v(R)$.

tions have been determined in Sec. III on the assumption of a constant logarithmic derivative (independent of the energy) at $R=a$. Figure 4 clearly illustrates that, for any R larger than a certain fixed distance and far from the classical turning point R_x, the channel wave functions for all high-lying resonances are practically the same within a constant factor and have almost the same logarithmic derivatives. The wavelengths of each wave function increase rapidly as R increases towards R_x, and the peak closest to R_x has the largest probability distribution; note the logarithmic scale for R. The position of this peak may be regarded as the size of the muonic molecule. This size R_v of a state at ϵ_v is about 20% less than R_x, or $0.8\sqrt{C/\epsilon_v}$, where C is given in Table II. Reference to this table indicates that the size is about

$$R_v\ (\text{cm}) \sim 10^{-8}/[4\sqrt{|\epsilon_v\ (\text{eV})|}] \qquad (25)$$

for all resonance states of muonic molecules in the table.

V. DISCUSSION

The theory developed in Sec. III not only proves the presence of two infinite series of resonances converging to the level $n=2$ of muonic atoms but also provides simple and useful formulas such as Eq. (24) for the resonance positions, Eq. (17) for the channel wave functions, and Eq. (25) for the size of the muonic molecules. The theory has also been found useful in the classification of resonance states by associating them with particular adiabatic potential-energy curves. The applications of the theory to the $(dd\mu)^+$ and $(dt\mu)^+$ molecules in Sec. IV have proved the reliability of both the present theory and the resonance energies of Ref. 14.

The resonances discussed in this paper may have an influence on various aspects of muon-related physics. In the study of this influence two peculiar properties of these resonances, especially high-lying resonances, must be kept in mind. One is the extremely large size of the muonic molecules. Figure 4 shows that the size of the $(dt\mu)^+$ molecules in resonance states lying within about an electron volt from the series limit is a few tenths of an angstrom or larger. Equation (25) says that the same is also true for other muonic molecules with small J. Therefore, a muonic molecular ion in any of these high-lying resonance states is not small enough to be regarded as a "pseudonucleus" in an electronic molecule. This is a property different even from that of the weakly bound muonic molecular ions briefly mentioned in Sec. I. The difference is due to the much longer range of the potentials in the channels $n=2$ than the potential in the channel $n=1$.

The other peculiar property that is absent from bound muonic molecules concerns the strong effects of the corrections to be made on the nonrelativistic theory. To discuss this we stress again that the results of Secs. III and IV are consequences of the long-range dipole potential proportional to R^{-2}. The mathematical origin of this potential has been explained in Sec. III. Its physical origin is the linear Stark effect, which follows from the perturbation theory for degenerate states. The degeneracy of the $2s$ and $2p$ levels, which are coupled by the dipole operator, is essential for the theory of Sec. III. Such degeneracy occurs only for two-body hydrogenlike atoms and not for atoms with more than two particles. Therefore, infinite series of resonances never occur for four-body or larger systems below a limit of dissociation into a nonhydrogenic neutral atom and an ion (or any charged particle); the optical potential of the longest range in this channel is the dipole polarization potential asymptotically proportional to R^{-4}.

We may expect that the three-body muonic molecules $(pp\mu)^+$, $(pd\mu)^+$, $(pt\mu)^+$, $(dd\mu)^+$, $(dt\mu)^+$, and $(tt\mu)^+$ have infinite series of resonances produced by the same mechanism as has been discussed so far and converging to the threshold of each principal quantum number n of a muonic hydrogenlike atom.

We have considered purely nonrelativistic Schrödinger equation. For accurate description, however, we must take into account the relativistic corrections, the nuclear effects, and the quantum-electrodynamic corrections.[28] Then, the degenerate levels with $n=2$ of the hydrogenlike muonic atoms $p\mu$, $d\mu$, and $t\mu$ split into six or seven nondegenerate levels, of which the two $2s_{1/2}$ metastable levels lie lower than the $2p$ levels by $\Delta E_2 \simeq 0.2$ eV due mainly to the vacuum polarization effect. Consequently, the asymptotic form of the optical potential is now proportional to R^{-4}, and there exist only a finite number of resonances. For intermediate R for which the potential is much stronger than ΔE_2, however, the level spacings are negligible and the potential in the transformed channel $i=2$ still behaves as R^{-2}. For this reason resonances satisfying the inequality $|\epsilon_v| \gg \Delta E_2$ in the nonrelativistic approximation are affected only little by the level splitting. Nonrelativistic resonances with $|\epsilon_v|$ of the same order of magnitude as ΔE_2 may be strongly influenced by the relativistic and quantum-electrodynamic effects. As for nonrelativistic resonances lying within about ΔE_2 from the dissociation limit, the mechanism of supporting them is now absent, and calculations of the conventional types based on the Schrödinger equation do not even serve as a starting point of perturbative approaches to these high-lying resonances in general. These considerations cast doubt on the existence of muonic molecular resonances with $|\epsilon_v| \lesssim 0.2$ eV. Probably there exist resonances within about an electron volt from the dissociation limit $n=2$, or within a few vibrational quanta of an electronic "hydrogen-isotope molecule," e.g., $d^+(dt\mu)^+e^-e^-$ or $d^+(dd\mu)^+e^-e^-$ for which the vibrational quantum is about 0.3 eV. If so, these resonance states may be formed with a high rate in processes (1), and may play an important role in muon-catalyzed fusion. A definitive proof of their existence has to await calculations of three-body resonances including the relativistic and quantum-electrodynamic effects. Furthermore, for studying their relevance to muon-catalyzed fusion, six-body treatment of the electronic molecules, rather than the approximation to regard them as pseudo-hydrogen-isotope molecules, is necessary because of the large size of the $(dt\mu)^+$ and $(dd\mu)^+$ ions.

Note added in proof. Equation (18) is valid when the off-diagonal elements of the $\mathcal{R}$ matrix are negligible. When they are not, a quantity varying slowly with energy must be added to the left-hand side of Eq. (18). Then, the left-hand side of Eq. (19) must be modified accordingly, but all the following equations remain unchanged.

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[1] S. E. Jones, Nature **321**, 127 (1986).

[2] S. E. Jones, A. N. Anderson, A. J. Caffrey, J. B. Walter, K. D. Watts, J. N. Bradbury, P. A. M. Gram, M. Leon, H. R. Maltrud, and M. A. Paciotti, Phys. Rev. Lett. **51**, 1757 (1983).

[3] W. H. Breunlich, M. Cargnelli, P. Kammel, J. Marton, P. Pawlek, J. Werner, J. Zmeskal, K. M. Crowe, J. Kurck, A. Janett, C. Petitjean, R. H. Sherman, H. Bossy, and W. Neumann, Phys. Rev. Lett. **53**, 1137 (1984).

[4] S. E. Jones, A. N. Anderson, A. J. Caffrey, C. DeW. Van Siclen, K. D. Watts, J. N. Bradbury, J. S. Cohen, P. A. M. Gram, M. Leon, H. R. Maltrud, and M. A. Paciotti, Phys. Rev. Lett. **56**, 588 (1986).

[5] W. H. Breunlich, M. Cargnelli, P. Kammel, J. Marton, N. Naegele, P. Pawlek, A. Scrinzi, J. Werner, J. Zmeskal, J. Bistirlich, K. M. Crowe, M. Justice, J. Kurck, C. Petitjean, R. H. Sherman, H. Bossy, H. Daniel, F. J. Hartmann, W. Neumann, and G. Schmidt, Phys. Rev. Lett. **58**, 329 (1987).

[6] E. A. Vesman, Pis'ma Zh. Eksp. Teor. Fiz. **5**, 113 (1967) [JETP Lett. **5**, 91 (1967)].

[7] L. I. Ponomarev, in *Atomic Physics 10,* edited by H. Narumi and I. Shimamura (Elsevier, Amsterdam, 1987), p. 197.

[8] I. Shimamura, Prog. Theor. Phys. (Kyoto) **82**, 304 (1989).

[9] S. Hara, T. Ishihara, and N. Toshima, Muon Catalyzed Fusion **1**, 277 (1987).

[10] C. Y. Hu, Phys. Rev. A **36**, 4135 (1987); **36**, 5420 (1987).

[11] M. Kamimura, Phys. Rev. A **38**, 621 (1988).

[12] S. E. Haywood, H. J. Monkhorst, and K. Szalewicz, Phys. Rev. A **37**, 3393 (1988); **39**, 1634 (1989).

[13] L. I. Menshikov and L. I. Ponomarev, Z. Phys. D **2**, 1 (1986).

[14] S. Hara and T. Ishihara, Phys. Rev. A **40**, 4232 (1989); and private communication.

[15] P. Froelich and K. Szalewicz, Muon Catalyzed Fusion **3**, 345 (1988); Phys. Lett. A **129**, 321 (1988).

[16] E. R. Cohen and B. N. Taylor, CODATA Bull. **63**, 1 (1986); J. Res. Natl. Bur. Stand. **92**, 85 (1987).

[17] D. R. Bates and R. H. G. Reid, Adv. Atom. Mol. Phys. **4**, 13 (1968).

[18] M. M. Madison and J. M. Peek, At. Data **2**, 171 (1971).

[19] C. L. Beckel, M. Shafi, and J. M. Peek, Phys. Rev. **59**, 5288 (1973).

[20] B. P. Carter, Phys. Rev. **173**, 55 (1968).

[21] M. Shafi and C. L. Beckel, Phys. Rev. **59**, 5294 (1973).

[22] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics,* 3rd ed. (Pergamon, Oxford, 1977), Sec. 35.

[23] M. J. Seaton, Proc. Phys. Soc. **77**, 174 (1961).

[24] M. Gailitis and R. Damburg, Proc. Phys. Soc. London **82**, 192 (1963).

[25] A. H. Wapstra and G. Audi, Nucl. Phys. **A432**, 1 (1985).

[26] H. S. Taylor, Adv. Chem. Phys. **18**, 91 (1970).

[27] A. U. Hazi and H. S. Taylor, Phys. Rev. A **1**, 1109 (1970); M. F. Fels and A. U. Hazi, *ibid.* **5**, 1236 (1972); H. S. Taylor and A. U. Hazi, *ibid.* **14**, 2071 (1976).

[28] E. Borie and G. A. Rinker, Rev. Mod. Phys. **54**, 67 (1982).