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Effects of the finite size of the ion $(dd\mu)^+$ on the energy levels of the molecules $(dd\mu)e$ and $(dd\mu)dee$

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The energy shift due to the finite size of the pseudonucleus $(dd\mu)_{11}^{+}$ in the molecules $(dd\mu)_{11}e$ and $(dd\mu)_{11}dee$, the subscripts indicating the first excited state with total angular momentum of one unit, is of importance in the theoretical estimation of the rate of d-d fusion catalyzed by negative muons. The energy shift in the molecule $(dd\mu)_{11}e$ is calculated using perturbation theory up to second order. The finite-size shift is found to be 1.46 meV. This is significantly larger than the value of 0.7 meV for this energy shift calculated by Bakalov [Muon Catalyzed Fusion **3**, 321 (1988)] by a method similar to the present method; recently found excellent agreement of theory with experimental results for the formation rate of the molecule $(dd\mu)_{11}dee$ was based on Bakalov's value with some modifications. The results of a direct calculation of the finite-size energy shifts in $(dd\mu)_{11}dee$ using first-order perturbation theory are presented. The contribution from the quadrupole component of the $(dd\mu)_{11}$ charge distribution, which is not taken into account in the conventional scaling procedure based on the finite-size energy shifts of $(dd\mu)_{11}e$, is found to be of the order of 1 meV and to depend on the angular-momentum states of $(dd\mu)_{11}dee$. Sources of uncertainty in the current theoretical estimates are also discussed. [S1050-2947(97)01210-9]

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I. INTRODUCTION

The molecule $(dd\mu)_{11}dee$ plays a key role in the process of muon-catalyzed fusion in D_2 (see [1–3] for recent general reviews). This molecule may be produced by collision of a D_2 molecule with a muonic deuterium atom $(d\mu)_{1s}$, in which the muon is in the $1s$ orbital,

$$(d\mu)_{1s}+[\mathrm{D}_2]_{\nu_iJ_i}\rightarrow[(dd\mu)_{11}dee]_{\nu_fJ_f}, \tag{1}$$

where the labels ν_i, J_i specify the initial vibrational and rotational quantum numbers of the D_2 molecule and ν_f, J_f specify the vibrational and rotational quantum numbers of the molecule in which the small three-body $(dd\mu)_{11}$ system plays the role of a pseudonucleus. The 11 subscript on this three-body system indicates that it is in the first excited state $(v=1)$ with total angular momentum J of one unit. The ground electronic state of the D_2 molecule is almost unchanged in reaction (1).

Since reaction (1) is a resonance process (in which the right-hand side is a resonance state), the rate for molecular formation by this mechanism is sensitive to the precise energy levels of the molecules involved. For energy matching of the initial and final states, the following condition must be satisfied to within an accuracy of the order of the width of the resonance:

$$E_{(d\mu)_{1s}}+E_{[D_2]_{\nu_iJ_i}}+E^{KE}=E_{[(dd\mu)_{11}dee]_{\nu_fJ_f}}, \tag{2}$$

where E_x is the energy of the appropriate system x and E^{KE} is the kinetic energy of the relative motion of the muonic atom and the D_2 molecule before collision. The width of the resonance state is broadened due to several effects [4–6] including back decay into the initial channel, nuclear fusion in $dd\mu$, Auger decay of the complex $[(dd\mu)_{11}dee]_{\nu_fJ_f}$, and collisional processes. Calculations of molecular formation rates indicate that all energies in Eq. (2) should be defined to better than about 1 meV in order make contact with experiment [7].

The $(dd\mu)_{11}$ system is small compared to the dimensions of the whole $(dd\mu)_{11}dee$ molecule [8] and it is therefore convenient to break down the energy $E_{[(dd\mu)_{11}dee]_{\nu_fJ_f}}$ into a contribution $E_{11}^{dd\mu}$ from the isolated $(dd\mu)_{11}$ system and a contribution $E_{\nu_fJ_f}^{Ndee}$ from the hydrogenlike molecule $Ndee$ in which the fictitious nucleus N has a mass equal to the combined mass of a deuteron, triton, and muon:

$$E_{[(dd\mu)_{11}dee]_{\nu_fJ_f}}=E_{11}^{dd\mu}+E_{\nu_fJ_f}^{Ndee}+E^{FS}. \tag{3}$$

The quantity E^{FS}, referred to here as the finite-size energy shift, represents the energy arising from the interaction of the extended $(dd\mu)_{11}$ charge distribution with the extra deuteron and two electrons in $(dd\mu)_{11}dee$. The binding energy of

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$(dd\mu)_{11}$, $E_{(d\mu)_{1s}} - E^{dd\mu}_{11}$, has been calculated to be 1966.4 meV in the lowest hyperfine level [9–11]. The energies $E^{Ndee}_{\nu_f J_f}$ have also been calculated accurately for various states $(\nu_f,\ J_f)$ of interest [12]. However, the energy E^{FS} remains to be accurately determined.

In a recent, detailed comparison of experiment and theory for $(dd\mu)_{11}dee$ [7], a theoretical formula for the molecular formation rate was fitted to the experimentally determined formation rate with the energy $\epsilon_{11} = E^{dd\mu}_{11} + E^{FS} - E_{(d\mu)_{1s}}$ as a fitted parameter. This yielded $\epsilon_{11} = -1966.1 \pm 0.2$ meV. This was compared with a theoretical value of -1966.2 meV calculated assuming a value $E^{FS} = 0.24$ meV. This remarkable agreement between experiment and theory needs to be carefully examined since the theoretical value of E^{FS} used in this comparison was based on an approximate scaling procedure applied to a directly calculated literature value [13]. The aim of this paper is therefore to address the question of the finite-size shift in this system.

A proper treatment of the energy levels of the $(dd\mu)_{11}dee$ molecule would involve solving the wave equation for a six-body system with particles in three different mass ranges. At the present moment such a calculation appears not to be tractable. A possible approach to calculate finite-size shifts is to employ perturbation theory with the sum of the Hamiltonians $H^{dd\mu}$ for $dd\mu$ and H^{Ndee} for $Ndee$ as the unperturbed Hamiltonian and with the first two terms on the right-hand side of Eq. (3) as the unperturbed energy. However, a direct application of perturbation theory to $(dd\mu)_{11}dee$ is extremely difficult beyond a first-order treatment because of the requirement to perform a summation over excited molecular states, including states that lie in the continuum. Thus the approach that has been adopted to obtain energy shifts used in fits of experimental formation rates is to consider first a simpler system $(dd\mu)_{11}e$ in which the finite-size effect can be treated more easily and more accurately. The electron in this system is assumed to be in its ground state in the zeroth-order approximation so that its perturbation energies may be related to those of $(dd\mu)_{11}dee$; see the discussion in Sec. IV A.

In Sec. II the finite-size energy shift in $(dd\mu)_{11}e$ is calculated using perturbation theory up to second order. In Sec. III first-order perturbation theory is applied to the six-body system $(dd\mu)_{11}dee$ in order to study the effect of terms in the perturbation interaction that are qualitatively different from those that appear in the case of the $(dd\mu)_{11}e$ system and that therefore have not been considered in the conventional scaling procedure. Section IV is devoted to a discussion of the validity of various theoretical assumptions made in the present work and in the literature.

II. THE MOLECULE $(dd\mu)_{11}e$

The method used here to calculate the finite-size energy shift E^{FS} in $(dd\mu)_{11}e$ is the same as that used in a previous calculation of E^{FS} in $(dt\mu)_{11}e$ and has been described in detail in Ref. [14]. Only a brief summary will be presented here.

The zeroth-order wave functions are products of the wave functions $\Phi^{dd\mu}_{JvM}$ for the isolated three-body $dd\mu$ system and the wave functions ψ^e_{nlm} for the two-body atomic system consisting of an electron and a nucleus N, whose Hamiltonian is H^{Ne} with eigenvalues E^{Ne}_{nl}. Here n, l, and m are the principal, azimuthal, and magnetic quantum numbers for the two-body system, while M is the z component of the total angular momentum in the three-body $dd\mu$ system. The eigenfunctions $\Phi^{dd\mu}_{JvM}$ and ψ^e_{nlm} satisfy equations

$$H^{dd\mu}\Phi^{dd\mu}_{JvM} = E^{dd\mu}_{Jv}\Phi^{dd\mu}_{JvM} \tag{4}$$

and

$$H^{Ne}\psi^e_{nlm} = E^{Ne}_{nl}\psi^e_{nlm}. \tag{5}$$

The perturbation interaction is

$$V = -\frac{1}{|\mathbf{r}_{d_1} - \mathbf{r}|} - \frac{1}{|\mathbf{r}_{d_2} - \mathbf{r}|} + \frac{1}{|\mathbf{r}_\mu - \mathbf{r}|} + \frac{1}{r}, \tag{6}$$

where $\mathbf{r}_{d_1}$, $\mathbf{r}_{d_2}$, $\mathbf{r}_\mu$, and $\mathbf{r}$ are the coordinates of the two deuterons, the muon, and the electron, respectively. The coordinate origin is chosen to be the center of mass of the $dd\mu$ system. Up to second order, E^{FS} may be written as

$$E^{FS} \simeq \Delta E^{(1)} + \Delta E^{(2)}, \tag{7}$$

with the first-order energy given by

$$\Delta E^{(1)} = \langle \psi^e_{1s}\Phi^{dd\mu}_{110}|V|\Phi^{dd\mu}_{110}\psi^e_{1s}\rangle \tag{8}$$

and the real part of the second-order energy by

$$\Delta E^{(2)} = \sum_{J,l} \mathrm{P}\sum\!\!\!\!\!\!\int_{v,n}{}' \sum_{M,m}{}' \frac{|\langle \psi^e_{nlm}\Phi^{dd\mu}_{JvM}|V|\Phi^{dd\mu}_{110}\psi^e_{1s}\rangle|^2}{E^{dd\mu}_{11} + E^{Ne}_{1s} - E^{dd\mu}_{Jv} - E^{Ne}_{nl}}, \tag{9}$$

where the summation-integration is over the bound and continuum electronic and $dd\mu$ states and P stands for the principal value of the integral over the electron energy. The primes on the summations indicate that the combination of the $dd\mu$ state with $J=1$ and $v=1$ and the $1s$ electronic state is to be excluded from the summation. In Eqs. (8) and (9) a representative value of zero has been chosen for M for the state $(J,v)=(1,1)$ since the results are independent of M.

The $dd\mu$ wave functions are written in terms of three sets of Jacobi coordinates $\{\mathbf{r}_c, \mathbf{R}_c\}_c$ $(c=1, 2, 3)$ that describe the positions of the three particles. These wave functions are expressed as sums of products of radial functions $\varphi_{Jv\lambda L}(r_c, R_c)$ and angular functions in these coordinates [15]:

$$\Phi^{dd\mu}_{JvM} = \sum_c \sum_{\lambda,L} \varphi_{Jv\lambda,L}(r_c, R_c)[Y_\lambda(\hat{\mathbf{r}}_c) \times Y_L(\hat{\mathbf{R}}_c)]_{JM}, \tag{10}$$

where the quantity in square brackets is the tensor product of two spherical harmonics.

It is convenient to expand the operator V in a multipole expansion and to perform the integrations over the coordinates of the deuteron, triton, and muon first, before performing the integration over the electron coordinate. The product of two $dd\mu$ wave functions and the interaction potential integrated over the $dd\mu$ coordinates, involved in Eq. (9), can then be written as

$$\langle\Phi^{dd\mu}_{JvM}|V|\Phi^{dd\mu}_{110}\rangle=\sqrt{4\pi}\sum_{\alpha} U^{(\alpha)}_{Jv}(r)\times\sum_{\beta=-\alpha}^{\alpha}\langle 10\alpha\beta|JM\rangle Y^{*}_{\alpha\beta}(\hat{\mathbf{r}})+\delta_{J1}\delta_{v1}\delta_{M0}r^{-1}, \tag{11}$$

where $\langle 10\alpha\beta|JM\rangle$ is a Clebsch-Gordan coefficient and $U^{(\alpha)}_{Jv}(r)$, the αth multipole potential, is given by

$$U^{(\alpha)}_{Jv}(r)=\sum_{p} q_p \sum_{\lambda,L\lambda',L'} \sqrt{4\pi}\frac{(2L+1)\tilde{J}\tilde{L}\tilde{\lambda}}{(2\alpha+1)\tilde{L}'\tilde{\lambda}'}(-1)^{L+\lambda}\times\langle\alpha 0L0|L'0\rangle W(JL1\lambda';\lambda\alpha)\times\sum_{c,c'}\int \varphi_{Jv\lambda L}(r_c,R_c)r_<^{\alpha}r_>^{-(\alpha+1)}\varphi_{11\lambda'L'}\times(r'_c,R'_c)r_c^2dr_cR_c^2dR_c. \tag{12}$$

Here the subscript p denotes a particle that is one of the two deuterons or the muon in $dd\mu$. This particle has a charge q_p and is located at a position $\mathbf{s}_p$. The symbol $\tilde{y}$ stands for the quantity $y(y+1)$ and $W(JL1\lambda';\lambda\alpha)$ is a Racah coefficient. The quantities $r_<$ and $r_>$ are the smaller or greater of the two radial coordinates s_p and r. The summation is to be taken over the three particles p in the $dd\mu$ system. In terms of the potentials $U^{(\alpha)}_{Jv}(r)$ defined in this way, the first-order perturbation energy can then be written as

$$\Delta E^{(1)}=\int \phi^2_{1s}[U^{(0)}_{11}(r)+r^{-1}]r^2dr, \tag{13}$$

where ϕ_{nl} is the radial component of the electronic wave function ψ^{e}_{nlm}. The second-order energy is

$$\Delta E^{(2)}=\frac{1}{3}\sum_{J,l}(2J+1)\mathrm{P}\times\sum\!\!\!\!\!\!\int_{v,n}\frac{\int\phi_{nl}[U^{(l)}_{Jv}(r)+\delta_{J1}\delta_{v1}\delta_{l0}r^{-1}]\phi_{1s}r^2dr}{E^{dd\mu}_{11}+E^{Ne}_{1s}-E^{dd\mu}_{Jv}-E^{Ne}_{nl}}. \tag{14}$$

The $dd\mu$ wave functions were calculated using a variational method [15] with 1789 basis functions giving a binding energy $E_{(d\mu)_{1s}}-E^{dd\mu}_{11}$ of 1974.8 meV. The contributions to the second-order energy from different states (J,v), after summation over all electronic states with a given l value, are shown in Table I. The results for the first- and second-order perturbation energies and their sum are summarized in Table II and compared with the corresponding results reported in Ref. [14] for the analogous $(dt\mu)_{11}e$ system.

A previous calculation of E^{FS} yielded a value of 1.0 ± 0.3 meV for $(dd\mu)_{11}dee$ after scaling the result for $(dd\mu)_{11}e$ by a factor of 1.45 [13]. This factor is the ratio of the electron density at the nucleus N in the atom Ne and in the molecule $Ndee$. Thus the value of E^{FS} for $(dd\mu)_{11}e$ implied by this result is 0.7 ± 0.2 meV as compared to the present result of 1.46 meV. Since only brief details of these calculations are given in Ref. [13], it is difficult to ascertain the source of the difference between the present calculations and those of Ref. [13]. The scaling procedure is discussed in more detail in Sec. IV A below.

TABLE I. Contributions to the second-order perturbation energy $\Delta E^{(2)}(J,v,l)$ for $(dd\mu)_{11}e$ from different intermediate $(dd\mu)_{Jv}$ states for a given electron angular momentum l, after summation over all bound and continuum electronic states for this value of l.

J	v	l	$\Delta E^{(2)}(J,v,l)$ (meV)
0	0	1	0.000
0	1	1	0.000
0	$v\geqslant 2$	1	-3.361
1	0	0	0.000
1	1	0	-0.154
1	$v\geqslant 2$	0	-0.252
1	0	2	0.000
1	$v\geqslant 1$	2	-0.064
2	0	1	0.000
2	$v\geqslant 1$	1	-6.177
2	0	3	0.000
2	$v\geqslant 1$	3	-0.008
3	$v\geqslant 0$	2	-0.097
4	$v\geqslant 0$	3	0.000
5	$v\geqslant 0$	4	0.000
	Total $\Delta E^{(2)}$		-10.113

III. THE MOLECULE $(dd\mu)_{11}dee$

We now consider the case of a molecule $(dd\mu)_{11}dee$. We note that one needs to use an eigenfunction of the square of the total angular momentum and its z component (with the corresponding quantum numbers denoted by J_t and M_t), rather than just the product of a $dd\mu$ wave function and a wave function of $Ndee$. Up to first order, the total energy of this state may be written in the form of Eq. (3) with the finite-size energy E^{FS} given as

$$E^{FS}\simeq\Delta E^{(1)}_{e,M}+\Delta E^{(1)}_{e,Q}+\Delta E^{(1)}_{d,M}+\Delta E^{(1)}_{d,Q} \tag{15}$$

up to the quadrupole terms in the interaction potential [16,17]. Here $\Delta E^{(1)}_{e,M}$ and $\Delta E^{(1)}_{e,Q}$ are the contributions to the first-order energy arising from the interaction of the electron distribution with the $(dd\mu)_{11}$ system and the subscripts M

TABLE II. Contributions to the finite-size energy shift in $(dd\mu)_{11}e$ and $(dt\mu)_{11}e$. $\Delta E^{(1)}$ and $\Delta E^{(2)}$ are the first- and second-order energies, while E^{FS} is the total finite-size energy shift up to second order. All energies are in meV.

Energy	$(dd\mu)_{11}e$	$(dt\mu)_{11}e$
$\Delta E^{(1)}$	11.577	18.253
$\Delta E^{(2)}$	-10.113	-17.752
E^{FS}	1.46	0.50

and Q denote the contributions from the monopole and quadrupole terms in the multipole expansion of the interaction operator. Similarly, $\Delta E^{(1)}_{d,M}$ and $\Delta E^{(1)}_{d,Q}$ are the contributions to the first-order energy arising from the interaction of the third deuteron with the $(dd\mu)_{11}$ system.

The electron monopole contribution $\Delta E^{(1)}_{e,M}$ is given by

$$\Delta E^{(1)}_{e,M} = \sum_{i=1}^{2} \langle \psi^{\nu}_{\mathrm{vib}}(R)\,\psi_e(\mathbf{r}_1,\mathbf{r}_2;R) | [U^{(0)}_{11}(r_i) + r_i^{-1}] \times | \psi_e(\mathbf{r}_1,\mathbf{r}_2;R)\,\psi^{\nu}_{\mathrm{vib}}(R)\rangle, \tag{16}$$

where $\psi_e(\mathbf{r}_1,\mathbf{r}_2;R)$ is the electronic wave function for the ground state of the molecule $Ndee$ (which is the same as the ground-state wave function of the hydrogen molecule H_2 in the Born-Oppenheimer approximation) at internuclear separation R and $\psi^{\nu}_{\mathrm{vib}}(R)$ is the vibrational wave function for this compound molecule. The electronic quadrupole contribution $\Delta E^{(1)}_{e,Q}$ is given by

$$\begin{aligned}\Delta E^{(1)}_{e,Q} = {} & \sqrt{4\pi} \sum_{i=1}^{2} \sum_{M,M_r} \sum_{M',M'_r} \sum_{\beta} \langle JMJ_rM_r | J_tM_t\rangle \\ & \times \langle JM'J_rM'_r | J_tM_t\rangle \langle JM'2\beta | JM\rangle \\ & \times \langle \psi^{J_rM_r}_{\mathrm{rot}}(\hat{\mathbf{R}})\,\psi^{\nu}_{\mathrm{vib}}(R)\,\psi_e(\mathbf{r}_1,\mathbf{r}_2;R) | \;\; U^{(2)}_{11} \\ & \times (r_i)\,Y^{*}_{2\beta}(\hat{\mathbf{r}}_i) | \psi_e(\mathbf{r}_1,\mathbf{r}_2;R)\,\psi^{\nu}_{\mathrm{vib}}(R)\,\psi^{J_rM'_r}_{\mathrm{rot}}(\hat{\mathbf{R}})\rangle,\end{aligned} \tag{17}$$

where $\psi^{J_rM_r}_{\mathrm{rot}}$ is the wave function for a rotational state (J_r,M_r) of the molecule $Ndee$. After performing the integration over $\hat{\mathbf{R}}$ and carrying out the summations over magnetic quantum numbers, this reduces to

$$\begin{aligned}\Delta E^{(1)}_{e,Q} = {} & (4\pi)^{3/2} A(J_t,J_r,J) \\ & \times \sum_{i=1}^{2} \int [\psi^{\nu}_{\mathrm{vib}}(R)]^2 [\psi_e(\mathbf{r}_1,\mathbf{r}_2;R)]^2 U^{(2)}_{11} \\ & \times (r_i)\,Y^{*}_{2\beta}(\hat{\mathbf{r}}_i)\,d\mathbf{r}_1 d\mathbf{r}_2 R^2 dR,\end{aligned} \tag{18}$$

with the factor $A(J_t,J_r,J)$ given by

$$\begin{aligned}A(J_t,J_r,J) = {} & (-1)^{J+J_r-J_t} [5(2J+1)(2J_r+1)]^{1/2} (4\pi)^{-1/2} \\ & \times \langle J020 | J_r0\rangle\, W(JJJ_rJ_r;2J_t).\end{aligned} \tag{19}$$

The monopole and quadrupole contributions arising from the presence of the third deuteron are given by the expressions

$$\Delta E^{(1)}_{d,M} = -\int [\psi^{\nu}_{\mathrm{vib}}(R)]^2 [U^{(0)}_{11}(R) + R^{-1}] R^2 dR \tag{20}$$

and

$$\Delta E^{(1)}_{d,Q} = -(4\pi)^{1/2} A(J_t,J_r,J) \int [\psi^{\nu}_{\mathrm{vib}}(R)]^2 U^{(2)}_{11}(R) R^2 dR. \tag{21}$$

TABLE III. Contributions to the finite-size energy shift in the molecules $(dd\mu)_{11}dee$ and $(dt\mu)_{11}dee$. Columns headed I refer to calculations using a fixed-nuclei Hartree-Fock approximation for the electronic wave function. Columns headed II refer to calculations using an accurate 54-term James-Coolidge wave function and accurate vibrational wave functions. The factor $A = A(J_t,J_r,J)$ is defined in the text and numerical values are given in Table IV. All energies are in meV.

Energy shift	$(dd\mu)_{11}dee$ I	$(dd\mu)_{11}dee$ II	$(dt\mu)_{11}dee$ I	$(dt\mu)_{11}dee$ II
$\Delta E^{(1)}_{e,M}$	16.41	14.46	25.79	25.22
$\Delta E^{(1)}_{e,Q}$	$3.47A$	$3.09A$	$5.47A$	$5.59A$
$\Delta E^{(1)}_{d,Q}$	$-7.24A$	$-5.97A$	$-11.35A$	$-10.77A$

In Ref. [16] we derived expressions for the electronic contributions $\Delta E_{e,M}$ and $\Delta E_{e,Q}$ for the special case of a Hartree-Fock-type electronic wave function at the equilibrium internuclear separation R_e expanded in a basis set of Gaussian functions centered on the two nuclei. The deuteron contributions $\Delta E_{d,M}$ and $\Delta E_{d,Q}$ were evaluated using both the δ function and accurate vibrational wave functions. In Table III we show, in the column headed I, the results of a calculation for $(dd\mu)_{11}dee$ using the fixed-nuclei Hartree-Fock approximation. In order to check the validity of this approximation (henceforth referred to as approximation I) we have also performed calculations using Eq. (18) with an R-dependent electronic wave function of the hydrogen molecule obtained by Kolos and Wolniewicz with 54-term James-Coolidge-type basis functions [18,19] that has almost converged. The results of this calculation (referred to as approximation II) are also shown in Table III in the column labeled II. For comparison we also give the results of similar calculations for $(dt\mu)_{11}dee$.

Table IV shows values of the coefficient $A(J_t,J_r,J)$ for different molecular states of interest together with the total contributions to the first-order perturbation energy from the quadrupole terms in the perturbation operator calculated using approximation II. The deuteron monopole contributions

TABLE IV. Values of the factor $A(J_t,J_r,J)$ for different molecular states of $(dd\mu)_{11}dee$ and $(dt\mu)_{11}dee$ with total angular momentum J_t and rotational angular momentum J_r. The value of J, the total angular momentum of the three-body $dd\mu$ or $dt\mu$ subsystem, is unity in all cases. Also tabulated is the sum of the contributions to the first-order perturbation energy (in meV) from the quadrupole interactions calculated in approximation II, which is discussed in the text.

			$\Delta E^{(1)}_{e,Q} + \Delta E^{(1)}_{d,Q}$	
J_t	J_r	$A(J_t,J_r,J)$	$(dd\mu)_{11}dee$	$(dt\mu)_{11}dee$
0	1	-0.3989	$+1.24$	$+2.00$
1	0	0.0000	0.00	0.00
1	1	0.1995	-0.62	-1.00
1	2	-0.1995	$+0.62$	$+1.00$
2	1	-0.0399	$+0.12$	$+0.20$
2	2	0.1995	-0.62	-1.00
2	3	-0.1596	$+0.50$	$+0.80$

$\Delta E^{(1)}_{d,M}$ are negligible at the level of 0.1 meV and the electronic monopole term clearly gives the dominant contributions to the perturbation energy for both $(dd\mu)_{11}dee$ and $(dt\mu)_{11}dee$. However, the quadrupole contributions are generally of the order of meV and moreover they depend strongly on the particular molecular state. These conclusions are the same in both approximations I and II, though the simpler approximation I gives results differing roughly at the 10–20 % level from the more accurate results of approximation II. Furthermore, the results for $(dd\mu)_{11}dee$ are seen to be between 50% and 70% of those for $(dt\mu)_{11}dee$.

Quadrupole energy shifts of the order of meV have also been reported by Bakalov and Melezhik [17]. They used a simple, one-parameter product-type electronic wave function and no vibrational or rotational wave function for the $Ndee$ molecule and therefore the dependence of E^{FS} on J_r and J_t was not discussed. Their $dd\mu$ and $dt\mu$ wave functions consisted of two terms. They also estimated energy-level splittings of the order of meV due to the interaction between the particle spins and the quadrupole electric field, which is not considered in the present work.

IV. DISCUSSION

A. Scaling according to the electron-density ratio

The calculations of the finite-size shift E^{FS} in the molecule $(dt\mu)_{11}dee$, reported in Ref. [20], involve a scaling of the calculated energy shifts in $(dt\mu)_{11}e$ by a factor of

$$\gamma = \frac{[\text{electron density at a proton in the } H_2 \text{ molecule}]}{[\text{electron density at the proton in the H atom}]}. \tag{22}$$

Later calculations on $(dt\mu)_{11}dee$ and $(dd\mu)_{11}dee$ in Refs. [13] and [21] also involve the same scaling procedure. A value of 1.45 was assigned to this ratio on the basis that it is given approximately by the ratio of the photoionization cross sections of the H_2 molecule and the H atom calculated in Ref. [22]. In fact, since this ratio depends upon the internuclear distance, it seems more appropriate to use a γ value that is averaged over the vibrational state of the molecule according to

$$\overline{\gamma} = \int \gamma [\psi^{\nu}_{\text{vib}}(R)]^2 R^2 dR. \tag{23}$$

Calculating the average in this way using the accurate James-Coolidge H_2 wave function described in Sec. III yields the results shown in the third column of Table V. We have also evaluated electron densities with a δ wave function expanded in Guassians that reproduces 91% of the correlation energy as described in Ref. [23]. This yields the values shown in the fourth column of Table V. The results are given for the $\nu=7$ vibrational state of $(dd\mu)_{11}dee$ and the $\nu=2$ vibrational state of $(dt\mu)_{11}dee$ since these correspond to the dominant resonances for $dd\mu$ and $dt\mu$ formation, respectively. It should be noted that the $\overline{\gamma}$ value for H_2 differs from that implied by the electron density reported in Table IV of [24], for which $\overline{\gamma}=1.299$. The latter value refers to an electron density at a distance of 0.7 a.u. from the midpoint between the nuclei. Taking $\overline{\gamma}=1.45$, as in previous work, appears to overestimate the density by 5% for $(dt\mu)dee$ and 15% for $(dd\mu)dee$.

TABLE V. Table of $\overline{\gamma}$ values for the molecules H_2, $(dd\mu)_{11}dee$, and $(dt\mu)_{11}dee$ in a vibrational state ν. Results in column 3 were calculated with an accurate 54-term James-Coolidge-type (JC) wave function [18,19]. Results in column 4 were calculated with a wave function expanded in Gaussians that reproduces 91% of the correlation energy [23].

		$\overline{\gamma}$	
Molecule	ν	JC	Gaussian basis
H_2	0	1.422	1.280
$(dd\mu)_{11}dee$	7	1.264	1.179
$(dt\mu)_{11}dee$	2	1.380	1.241

As shown in Sec. III, the first-order perturbation energy in $(dd\mu)_{11}dee$ is dominated by the contribution from the monopole term in the multipole expansion of the perturbation energy. This contribution involves an integral of the square of the electronic wave function multiplied by a short-range potential that effectively vanishes for $r > 0.1$ a.u. The short-range behavior of this potential has a consequence that the monopole contribution is only sensitive to the electronic wave function for small radial distances from the pseudonucleus N. This leads to the expectation that the monopole contribution $\Delta E^{(1)}_{e,M}$ in $(dd\mu)_{11}dee$ should be close to the value obtained by multiplying the first-order finite-size correction $\Delta E^{(1)}$ in $(dd\mu)_{11}e$ by the factor $\overline{\gamma}$. Indeed, the monopole contribution is 14.46 meV according to Table III and $\overline{\gamma}\Delta E^{(1)} = 1.264 \times 11.58$ meV$=$14.63 meV according to Tables II and V, and these two values agree within an accuracy of 0.2 meV. Similarly good agreement is found also for $(dt\mu)_{11}dee$: $\Delta E^{(1)}_{e,M} = 25.22$ meV and $\overline{\gamma}\Delta E^{(1)} = 1.380 \times 18.25$ meV $=$ 25.18 meV.

The second-order energy, however, depends on a sum of integrals containing products of electronic wave functions for the ground and excited states of H_2. There is no reason why such integrals should scale by the factor $\overline{\gamma}$ from $(dd\mu)_{11}e$ to $(dd\mu)_{11}dee$ since the factor $\overline{\gamma}$ involves only the ground-state electronic wave function. The method of scaling the sum of the first- and second-order contributions that has been used previously [13,20,21] is thus unjustified.

B. Scaling according to the results on $(dt\mu)e$

Recently a detailed fit of the experimental values for the effective rate of formation of $(dd\mu)_{11}dee$, $\lambda_F(T)$, was reported [7] using the expression

$$\lambda_F(T)=\lambda_1+\lambda_2\frac{3}{2}kT+\sum_{J_i,S,J_f}\alpha\lambda_{FJ_i,SJ_f}(T;\epsilon_{11})\times\omega_S^{\mathrm{fus}}(J_f;\alpha,\lambda_{\mathrm{fus}}). \qquad (24)$$

Here T is the temperature, k is the Boltzmann constant, λ_1 and λ_2 are constants that determine the rate of nonresonant molecular formation in an approximation linear in T, and $\lambda_{FJ_i,SJ_f}(T;\epsilon_{11})$ is the rate of resonant molecular formation from a $d\mu$ atom in a hyperfine state F and a D_2 molecule in a rotational state J_i yielding a molecule $(dd\mu)_{11}dee$ in a rotational state J_f and with the subsystem $(dd\mu)_{11}$ having spin S. The rate λ_{FJ_i,SJ_f} is proportional to the transition matrix element squared $|V_{if}|^2$ and to the Maxwell distribution of the relative kinetic energy E^{KE} between $d\mu$ and D_2 on the assumption of negligible resonance widths, E^{KE} being related to ϵ_{11} through Eqs. (2) and (3). Thus the rate λ_{FJ_i,SJ_f} depends on ϵ_{11}. The parameter α has been introduced to scale $|V_{if}|^2$ in order to allow for errors in $|V_{if}|^2$. The quantity $\omega_S^{\mathrm{fus}}(J_f;\alpha,\lambda_{\mathrm{fus}})$ is the fusion fraction or the fraction of molecules $(dd\mu)_{11}dee$ that lead to d-d fusion. It depends not only on J_f but also on the effective fusion rate λ_{fus} from the state $(J,v)=(1,1)$ and on the parameter α.

The fitting was carried out using five adjustable parameters λ_1, λ_2, ϵ_{11}, λ_{fus}, and α. The best fit to the data yielded $\epsilon_{11}=-1966.1\pm0.2$ meV, which seems to be in good agreement with a theoretical result $\epsilon_{11}=-1966.2$ meV. However, this excellent agreement should be taken with reservation since, for one thing, this theoretical result involves an uncertainty of about 0.5 meV stemming from the uncertainty in the deuteron polarizability [7,10]. Another source of uncertainty in the theoretical result is the inclusion of an estimated value $E^{FS}_{dd\mu dee}(\mathcal{E})$ of the finite-size shift, which was taken to be 0.24 meV from Table X of Ref. [3]. This result was estimated [25] in the following way from the value of $E^{FS}_{dd\mu dee}(\mathcal{B})=1.0$ meV due to Bakalov [13]:

$$E^{FS}_{dd\mu dee}(\mathcal{E})=E^{FS}_{dd\mu dee}(\mathcal{B})\frac{E^{FS}_{dt\mu dee}(\mathcal{S})}{E^{FS}_{dt\mu dee}(\mathcal{B})}. \qquad (25)$$

Here $E^{FS}_{dt\mu dee}(\mathcal{S})$ is the value (0.29 meV) of the finite-size shift in $(dt\mu)_{11}dee$ calculated by Scrinzi and Szalewicz [21] and $E^{FS}_{dt\mu dee}(\mathcal{B})$ is the value (1.2 meV) of the same shift calculated by Bakalov [13]. This kind of scaling procedure seems to have little justification since it involves the scaling of a theoretical result for one physical quantity $E^{FS}_{dd\mu dee}$ by a large factor (approximately a factor of 4, which demonstrates a large discrepancy between theories) according to a result for a different physical quantity $E^{FS}_{dt\mu dee}$; we note especially that (i) the value of $E^{FS}_{dt\mu dee}(\mathcal{S})$ was obtained by scaling from $E^{FS}_{dt\mu e}$ in Ref. [21] by the use of the procedure discussed and questioned in Sec. IV A, and, furthermore, (ii) the method of Ref. [21] for calculating $E^{FS}_{dt\mu e}$ is inapplicable to homonuclear diatomic molecular ions such as $(dd\mu)_{11}e$ since the method takes advantage of the t-μ distance that is shorter than the d-μ distance.

C. Possibility of cancellation between perturbation energies

The contributions to E^{FS} in $(dd\mu)_{11}dee$ from the quadrupole terms to first order in the perturbation potential have been found to be of order 1 meV. Moreover, this quadrupole contribution, which is absent in the molecule $(dd\mu)_{11}e$, has been found to depend on both the total angular momentum of $(dd\mu)_{11}dee$ and the angular momentum of the molecule $Ndee$. It should be noted that contributions to the first-order perturbation energy from higher multipole terms may partially cancel these quadrupole contributions. However, the degree of such cancellation, which may be state dependent because of the different angular-momentum dependence of the perturbation energies in different orders, remains to be demonstrated in the molecular system $(dd\mu)_{11}dee$. The possibility of incomplete cancellation of the quadrupole terms thus represents an additional limit on the accuracy of the finite-size energy shift in this system.

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