Muonic Molecules and Nucleon-Deuteron Capture*

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Variational wave functions have been computed for all S states of muonic molecules involving hydrogen isotopes. Results for the $p\mu d$ ground state have been used to compare nucleon-deuteron capture cross sections with $p\mu d$ fusion rates. Using our molecular wave functions and the results of muonic-molecule experiments, we estimate the low-energy S-wave cross section for the reaction $D(p,\gamma)He^3$. Our result is in disagreement with measurements in the tens-of-kilovolts region, but is consistent with thermal neutron-deuteron capture data when Coulomb effects are taken into account by means of the usual penetration factor. Finally, we reexamine the usual assumption that proton-deuteron capture occurs only from the doublet state and show how the doublet and quartet matrix elements could both be determined from a new measurement of the total yield of gamma rays from $p\mu d$ fusion in liquid hydrogen.

I. INTRODUCTION AND SUMMARY

MUONIC molecules are systems like $p\mu p$, $p\mu d$, etc., in which a negative muon (μ) binds together two nuclei, each of which is a proton (p), deuteron (d), or triton (t). Such systems were first studied theoretically by Frank[1] and experimentally by Alvarez *et al.*[2] in connection with the phenomenon of muon-catalyzed fusion, in which a muon stopping in liquid hydrogen initiates the following sequence of reactions:

$$\begin{aligned} &\mu+p\to p\mu\,,\\ &p\mu+d\to d\mu+p\,,\\ &d\mu+p\to p\mu d\,,\\ &p\mu d\to He^3\mu+\gamma \quad\text{or}\quad He^3+\mu\,. \end{aligned} \tag{1}$$

Other experiments on muonic molecules have involved the production of $p\mu p$ and $d\mu d$, fusion[3] in $d\mu d$, and weak capture[4] of the muon by a proton in $p\mu p$. Also, weak capture has recently been observed[5] in $p\mu d$ and $He^3\mu$ (the $He^3\mu$ being produced by fusion in $p\mu d$).

Theoretical work on muonic molecules has included many approximate solutions of the Schrödinger equations for these systems. In the past, wave functions have been computed for all muonic molecules using various adiabatic approximations,[6–11] whereas variational calculations[12–18] have apparently been limited to molecules with identical nuclei. Section II of the present paper includes variational calculations of the bound S states of all muonic molecules. Only the results for $p\mu d$ are used in subsequent sections, but some of the other states are of interest for computing various molecular,[19] nuclear,[19] and weak-capture[4,5] rates not discussed in this paper. [*Note added in proof.* A. A. Frost *et al.*—J. Chem. Phys. **41**, 482 (1964)—have used a generalization of the method of C. L. Pekeris—Phys. Rev. **112**, 1649 (1958)—to obtain 2874 eV for the $p\mu d$ ground state.]

For each of our variational functions, we may perform an integration over the muon coordinates to obtain a pseudo wave function $G(r_{12})$, which depends only on the distance between the two nuclei. In Sec. II, we calculate $|G(0)|^2$ for the $p\mu d$ ground state. In Sec. III, using $|G(0)|^2$ and the results of muonic-molecule experiments, we estimate the low-energy S-wave cross section σ_{sp} for the reaction $D(p,\gamma)He^3$. For 25-keV protons, our estimate of σ_{sp} is in disagreement with the measured[20] value by a factor of 3. This discrepancy, and the kind of work needed to resolve it, are discussed in Sec. III.

We find that the spin-averaged square of the matrix element that appears in our estimate of σ_{sp} is roughly equal to the corresponding quantity for the mirror process $D(n,\gamma)H^3$. The matrix elements for proton capture are defined in such a way that the Coulomb penetration factor appears as a multiplying factor in the expression for σ_{sp}. This result is expected on the basis of certain assumptions, discussed in Sec. III, which, in turn, rest on the approximate equality of the absolute

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values of the exchange moments of the nuclei H^3 and He^3.

It is assumed in Sec. III that the fusion rate from the quartet state of proton and deuteron is negligible compared to the doublet rate. This assumption, which is now widely used,[3,10,19,21] is re-examined in Sec. IV. There we point out that certain results which have been obtained for thermal neutron-deuteron capture apply directly in our case. In particular, an orthogonality relation[22,23] shows that the quartet rate is expected to be of the same order of magnitude as the doublet rate.

Since there is no theoretical justification for ignoring the quartet rate, we use the present data[21] on the time distribution and total yield of gamma rays to determine a range of possible values of the quartet rate. We also show how a more accurate measurement of the total yield would determine both rates.

II. S STATES OF MUONIC MOLECULES

Variational Trial Functions and Matrix Elements

In this section, we find approximate solutions to the spinless, nonrelativistic Schrödinger equation for each of the muonic molecules $p\mu p$, $p\mu d$, $p\mu t$, $d\mu d$, $d\mu t$, and $t\mu t$. Since we are only concerned with S states, the only coordinates of interest are the interparticle distances r_1, r_2 (from nuclei 1 and 2 to the muon), and r_{12} (between the two nuclei). For each state, we construct a wave function

$$\psi=\sum_{i=1}^{N} X_i|A_i,B_i,C_i\rangle, \tag{2}$$

where the X_i are numerical coefficients of basis functions $|A_i,B_i,C_i\rangle$ that have the form

$$|A,B,C\rangle\equiv\exp[-S(Ar_1+Br_2+Cr_{12})]. \tag{3}$$

The scale factor S is positive, and never varies during a calculation. Once the $3N$ parameters A_i, B_i, C_i have been chosen, the next step is to compute the matrix elements of the Hamiltonian and of unity:

$$\begin{aligned} H_{ij}&=\langle A_iB_iC_i|H|A_jB_jC_j\rangle, \\ U_{ij}&=\langle A_iB_iC_i|1|A_jB_jC_j\rangle. \end{aligned} \tag{4}$$

These integrals can be evaluated exactly, as rational functions of the six arguments A_i, B_i, C_i, A_j, B_j, C_j. The functional dependence is given in Appendix A, in a form which is convenient for numerical work. From Appendix A, we see that the integrals converge, if and only if A_i+B_i, A_i+C_i, and B_i+C_i are all positive.

To find the energy level E, and the linear parameters X_i, we solve the matrix eigenvalue problem

$$H_{ij}X_j=-EU_{ij}X_j, \tag{5}$$

where H and U are real, symmetric N by N matrices, and U is positive definite. A computer program has been written to evaluate the matrix elements and solve Eq. (5). The program has been run on an IBM 7090 for various choices of the A_i, B_i, C_i. The resultant energy levels and certain other quantities (see below) are presented in Table I. Our energy levels agree with the most accurate adiabatic[9,19] and variational[16] calculations of other authors to within a few electron volts, for the ground states. For the vibrationally excited states, our levels differ from those of other authors[9,19] by 7–10 eV.

TABLE I. Energy levels and muon-nucleus overlaps of muonic molecules.

System	Vibrational level v	Energy level E (eV)	Muon-nucleus overlaps[a] γ_p	γ_d	γ_t
$p\mu p$	0	2780.8	0.568	...	...
$p\mu d$	0	2883.8	0.507	0.641	...
$p\mu t$	0	2924.1	0.490	...	0.663
$d\mu d$	0	2987.6	...	0.585	...
$d\mu d$	1	2698.2	...	0.5036	...
$d\mu t$	0	3029.1	...	0.569	0.610
$d\mu t$	1	2741.7	...	0.377	0.635
$t\mu t$	0	3072.5	...	...	0.595
$t\mu t$	1	2786.3	...	...	0.516

[a] The overlap γ_N is the ratio of the muon density at a particular nucleus of type N in the molecule, to the muon density at the nucleus of an $N\mu$ atom.

Muon-Nucleus Overlaps

For each S state, we define P_1 and P_2 to be the probability densities for the muon to be in the vicinity of nuclei 1 and 2, respectively. From Eqs. (2) and (3), the following simple formulas are obtained:

$$\begin{aligned} P_1&=\sum_{i=1}^{N}\sum_{j=1} X_iX_j\int_0^\infty \exp[-S(B_i+B_j+C_i+C_j)r]4\pi r^2dr, \\ P_2&=\sum_{i=1}^{N}\sum_{j=1} X_iX_j\int_0^\infty \exp[-S(A_i+A_j+C_i+C_j)r]4\pi r^2dr. \end{aligned} \tag{6}$$

If nucleus 1 (2) is a proton, then the overlap γ_p is defined, as usual,[16,17] to be the ratio of P_1 (P_2) to the muon density at the nucleus of a $p\mu$ atom. γ_d and γ_t are defined similarly with respect to $d\mu$ and $t\mu$ atoms. Equations (6) have been evaluated, and the overlaps computed, for all states listed in Table I. Our value of γ_p for the $p\mu p$ ground state is within 1% of that obtained by Wessel and Phillipson.[16] This close agreement is evidence that our computer program is running correctly.

Hyperfine Structure of the $p\mu d$ Ground State

Zel'dovich and Gershtein have calculated the hyperfine levels in $p\mu d$, assuming that each nucleus interacts with the muon through a contact term in the Hamil-

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tonian.[19] They used a molecular wave function such that, in our notation, $\gamma_p=\gamma_d=\frac{1}{2}$. Since the contact terms for the proton and deuteron are proportional to γ_p and γ_d, respectively, we multiply these terms by $2\gamma_p$ and $2\gamma_d$. Otherwise our treatment is identical to theirs. We do not repeat their derivation, but merely quote results.

Using the values of the overlaps in Table I, we obtain the following energies for the $J=2$ level, the $J=0$ level, and the two $J=1$ levels:

$$\epsilon_2=0.022 \text{ eV}; \qquad \epsilon_0=-0.0088 \text{ eV};$$
$$\epsilon_1=-0.040 \text{ eV}; \quad \epsilon_1'=0.0065 \text{ eV}.$$

The rate of fusion from each level depends on the probability that the proton and deuteron are in a doublet state. This probability is zero for ϵ_2, unity for ϵ_0, α for ϵ_1', and β for ϵ_1, where α and β depend on the values assumed for the overlaps. We obtain $\alpha=0.923$, $\beta=0.077$, whereas the values obtained in Ref. 19 were $\alpha=0.82$, $\beta=0.18$.

Probability Density P_3 in $p\mu d$

The rate of fusion of a muonic molecule by a particular process is proportional to one or the other of two quantities which can be computed from the molecular wave function, depending on the nature of the fusion process. For $p\mu d$, the radiative rate is proportional to the probability per unit volume P_3 that the two nuclei are together, while the nonradiative rate is proportional to $|\psi(0,0,0)|^2$, the probability per unit volume squared that all three particles are together. Our wave functions are not sufficiently accurate to give a reliable value for $|\psi(0,0,0)|^2$, so we shall not deal with this quantity in the present paper.

We have computed P_3 from various of our wave functions, by means of an expression similar to (6). The values so obtained have all been unreasonably large, and have tended to decrease, but not converge, as the number of terms in the wave function increased. A reliable value for P_3 could have been obtained by merely increasing the number of terms in our wave functions, but we determined P_3 by an alternate method (described below), which was more economical in terms of computer time.

Two-Body Pseudo Wave Function

For a given wave function $\psi(r_1,r_2,r_{12})$, we define the pseudo wave function $G(r_{12})$ as follows:

$$G(r_{12})=\left[\int\int \psi^2 d\tau_\mu\right]^{1/2}. \qquad (7)$$

Here the integration is over r_1 and r_2, and $d\tau_\mu$ is the two-center volume element for fixed r_{12}. Thus the probability that the internuclear distance lies between r_{12} and $r_{12}+dr_{12}$ is equal to $4\pi r_{12}^2 G^2(r_{12})$. The integration in (7)

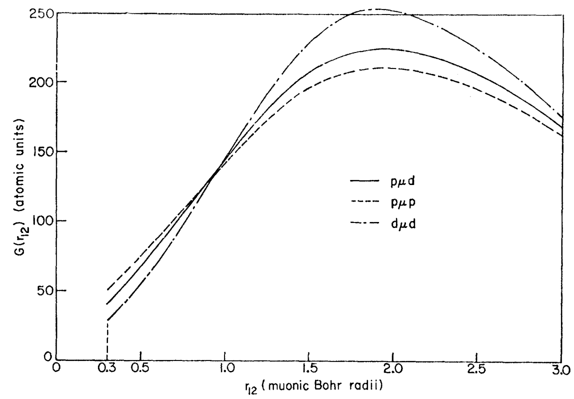

FIG. 1. Plots of the pseudo wave function $G(r_{12})$ for the relative coordinate of the two nuclei in the $p\mu p$, $p\mu d$, and $d\mu d$ ground states. These curves are remarkably straight for $r_{12}\lesssim 0.7$. In this respect they resemble zero-energy, S-wave Coulomb wave functions—which is not surprising, since for small values of r_{12} the Coulomb potential between the two nuclei is the dominant potential term in the Hamiltonian.

is carried out in detail in Appendix B, for ψ given by Eqs. (2) and (3); results for some of our wave functions are plotted in Figs. 1–3.

Although $G(r_{12})$ is not a wave function in the sense of a solution of a Schrödinger equation, Figs. 1 to 3 indicate that G behaves very much like the absolute value of an adiabatic[6] wave function for the nuclear motion. The dependence of G on the reduced mass[24] of the two nuclei is just what one would expect in the

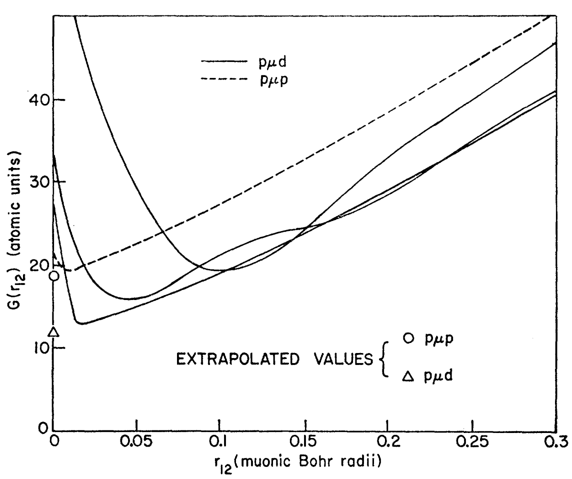

FIG. 2. Continuation of $p\mu p$ and $p\mu d$ curves of Fig. 1, for $r_{12}\lesssim 0.3$. Three curves are drawn for $p\mu d$, illustrating the convergence with increasing N in Eq. (2). The height and width of the spikes at $r_{12}=0$ decrease as ψ becomes more accurate. We assume the spikes are unphysical, and extrapolate to $r_{12}=0$.

[24] In fact, to an accuracy of about 1% the G curves for $p\mu d$ depend *only* on the reduced mass. This was checked by substituting $2m_pm_d/(m_p+m_d)$ for m_p and m_d in the Schrödinger equation for ψ. Authors whose computer programs are limited to the case of identical nuclei can therefore find P_3 for $p\mu d$ (and probably for $p\mu t$ and $d\mu t$ as well).

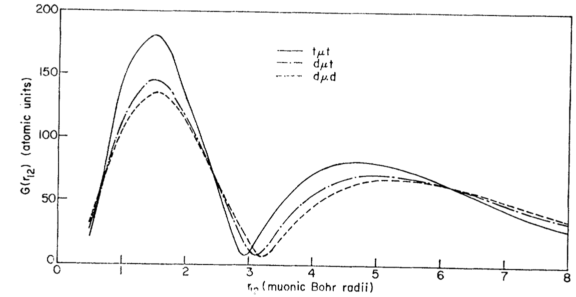

FIG. 3. Plots of $G(r_{12})$ for the first (and only) vibrationally excited S states. These states are just barely bound; hence the curves extend out many muonic Bohr radii, and die down at a rate which goes roughly like the square root of the dissociation energy. (This energy is greater for $d\mu d$ than for $d\mu t$, hence their curves will cross at some large value of r_{12}.)

Born-Oppenheimer approximation: As the reduced mass is increased, the curves (i) tend to have sharper maxima and minima, (ii) become more concentrated near the equilibrium point $r_{12}=2$, and (iii) fall off more rapidly as $r_{12}\rightarrow 0$ or ∞.

Each curve in Fig. 2 has a spike at $r_{12}=0$. The unreasonable values of P_3 mentioned above are the values of $|G(0)|^2$ at the tops of the spikes. We therefore replace them by the extrapolated values shown in Fig. 2. For $p\mu d$, we obtain $|G(0)|^2=[12 \text{ atomic units (a.u.)}]^2=10^{27}$ cm^{-3}, which we shall use in the next section to relate the $p\mu d$ fusion rate to the S-wave cross section for the reaction $D(p,\gamma)He^3$.

III. CAPTURE CROSS SECTIONS AND $p\mu d$ FUSION RATES

The last of Reactions (1) has two modes: radiative and nonradiative. We shall assume throughout this section that both kinds of fusion occur only when the proton and deuteron are in the doublet spin state. This is strictly true for the nonradiative process,[10] but open to question (see Sec. IV) in the radiative case.

Let λ_r and λ_{nr} be the radiative and nonradiative rates from the doublet pd state in $p\mu d$. The total doublet fusion rate is then $\lambda_f=\lambda_r+\lambda_{nr}$. The radiative, nonradiative, and total rates for a given hyperfine state are obtained by multiplying the doublet rates by the probability (see Sec. II) that the proton and deuteron are in the doublet state. We shall only discuss data taken at low deuteron concentration. In this case, the hyperfine levels are statistically[21] populated, and the average radiative, nonradiative, or total rate is just one third of the corresponding doublet rate.

$p\mu d$ Fusion Rates and the Time Distribution of Gamma Rays

When a muon comes to rest in liquid hydrogen, the rate of formation of $p\mu d$ is rapid compared to λ_f, provided that the deuterium concentration is larger than a few parts per million. The time distribution of gamma rays[21] from $p\mu d$ fusion then looks like a parent-daughter decay curve. We are interested in the behavior of this curve in the region 1 μsec$\leq t \leq 7$ μsec, which makes t large compared to the rise time. We may therefore neglect the rise time and assume that a free muon becomes a $p\mu d$ instantaneously, with statistical probabilities 5/12, $\frac{1}{4}$, $\frac{1}{4}$, $\frac{1}{12}$ for occupying the four hyperfine levels.

Let A, B, C, D be the populations of the hyperfine levels (see Sec. II) as a function of time t. The initial populations are statistical: $A(0)=5$, $B(0)=3$, $C(0)=3$, $D(0)=1$. Each state is decaying at its fusion rate plus the decay rate $\lambda_0\equiv 0.455$ μsec^{-1} of the muon,[25] but free muons are being produced at the rate $\lambda_{nr}(D+\alpha C+\beta B)$, and these free muons may be regarded as becoming $p\mu d$'s instantaneously. The populations therefore satisfy the following differential equations:

$$\begin{aligned}\dot{A}(t)&=(5/12)\lambda_{nr}(D+\alpha C+\beta B)-\lambda_0 A\,,\\ \dot{B}(t)&=(3/12)\lambda_{nr}(D+\alpha C+\beta B)-(\lambda_0+\beta\lambda_f)B\,,\\ \dot{C}(t)&=(3/12)\lambda_{nr}(D+\alpha C+\beta B)-(\lambda_0+\alpha\lambda_f)C\,,\\ \dot{D}(t)&=\tfrac{1}{12}\lambda_{nr}(D+\alpha C+\beta B)-(\lambda_0+\lambda_f)D\,.\end{aligned}\quad(8)$$

Substituting the initial populations into the last three equations above, and using the fact that $\alpha+\beta=1$, we obtain

$$\begin{aligned}\dot{B}(0)&=(\eta-3\beta)\lambda_f-3\lambda_0\,,\\ \dot{C}(0)&=(\eta-3\alpha)\lambda_f-3\lambda_0\,,\\ \dot{D}(0)&=(\tfrac{1}{3}\eta-1)\lambda_f-\lambda_0\,,\end{aligned}\quad(9)$$

where $\eta=\lambda_{nr}/\lambda_f$ is the fraction of fusion events in which the muon is ejected.

The rate at which gamma rays are being produced is (except for a constant of proportionality)

$$n_\gamma(t)=D+\alpha C+\beta B\,.\quad(10)$$

As mentioned before, this function behaves like the tail of a parent-daughter decay curve. The slope of the curve is

$$\dot{n}_\gamma(t)=\dot{D}+\alpha\dot{C}+\beta\dot{B}\,,\quad(11)$$

and the apparent rate of decay (the rate of depletion of the daughter product) is minus the ratio of slope to value:

$$\lambda_c(t)=-\dot{n}_\gamma(t)/n_\gamma(t)\,.\quad(12)$$

From (9), (10), (11), (12), and the initial populations, we obtain

$$\lambda_c(0)=\lambda_0+\{\tfrac{1}{4}[1+3(\alpha^2+\beta^2)]-\tfrac{1}{3}\eta\}\lambda_f\,.\quad(13)$$

The experimental value $\lambda_{fx}=0.305\pm0.010$ μsec^{-1} was determined[21] from $n_\gamma(t)$ by using the values $\alpha=0.86$, $\beta=0.14$, and $\eta=0.16$. As a first approximation, we shall

[25] We neglect the rate of capture of the muon by the nuclei, since the capture rate in this case is three orders of magnitude smaller than the fusion and decay rates.

TABLE II. Dependence of $p\mu d$ fusion rates and yields on α, β, and η, assuming no quartet capture.

	$p\mu d$ fusion rates from the doublet pd state			First generation yield of fusions	Total yield of gamma rays
	Non-radiative	Radiative	Total		
λ_{fx}	λ_{nr}	λ_r	λ_f	Y_1	Y_γ
	(i) $\alpha=0.86$, $\beta=0.14$, $\eta=0.15$				
0.295	0.044	0.250	0.294	0.130	0.113
0.305[a]	0.046	0.258	0.304	0.133	0.115
0.315	0.047	0.267	0.314	0.136	0.118
	(ii) $\alpha=0.86$, $\beta=0.14$, $\eta=0.19$				
0.295	0.057	0.242	0.299	0.132	0.109
0.305[a]	0.059	0.250	0.309	0.135	0.112
0.315	0.061	0.258	0.319	0.138	0.114
	(iii) $\alpha=0.923$, $\beta=0.077$, $\eta=0.15$				
0.295[a]	0.040	0.228	0.268	0.118	0.102
0.305	0.042	0.235	0.277	0.121	0.105
0.315	0.043	0.243	0.286	0.124	0.107
	(iv) $\alpha=0.923$, $\beta=0.077$, $\eta=0.19$				
0.295[a]	0.052	0.221	0.272	0.120	0.099
0.305	0.053	0.228	0.281	0.122	0.101
0.315	0.055	0.235	0.291	0.125	0.104

[a] Interpolated values of λ_{fx} (see text). λ_{fx} determines the ratio of slope to value of $n\gamma(t)$ at $t=0$.

assume that $\lambda_c(0)$ is an experimentally known quantity, related to the above values of λ_{fx}, α, β, and η by Eq. (13). We may then solve Eq. (13) to determine what λ_f must be for other values of α, β, and η:

$$\lambda_f=0.76607\lambda_{fx}/[\tfrac{1}{4}+\tfrac{3}{4}(\alpha^2+\beta^2)-\tfrac{1}{3}\eta]. \quad (14)$$

Equation (14) is evaluated in Table II for various values of α, β, η, and λ_{fx}. For each row with $\lambda_{fx}=0.295$ or 0.315 (the lower and upper limits of experimental error) in Table II, we have integrated equations (8) numerically to find the populations at $t=3$ μsec, and then obtained $\lambda_c(3)$ from (8), (10), (11), and (12). To improve on our first approximation, we have found by interpolation the value of λ_{fx} which makes $\lambda_c(3)$ the same as it would be for the case $\lambda_{fx}=0.305$, $\alpha=0.86$, $\alpha=0.14$, $\eta=0.16$. The interpolated values are given in Table II. Since they lie within the experimental bounds, we conclude that our first approximation is accurate enough to deal with the available data.

$p\mu d$ Fusion Yields and an Estimate of λ_r

From the known[21] molecular rates it is easily shown that, at 1% deuterium concentration, the probability for a free muon to form a $p\mu d$ molecule is 0.9123. Multiplying this quantity by the probability that a $p\mu d$ molecule will undergo fusion, we obtain the first generation yield, i.e., the probability that a muon will catalyze at least one fusion reaction:

$$Y_1=0.9123\left\{\frac{1}{4}\frac{\alpha\lambda_f}{\alpha\lambda_f+\lambda_0}+\frac{1}{4}\frac{\beta\lambda_f}{\beta\lambda_f+\lambda_0}+\frac{1}{12}\frac{\lambda_f}{\lambda_f+\lambda_0}\right\}. \quad (15)$$

Let Y_N be the probability that a muon will catalyze at least N fusions. Then

$$Y_N=Y_{N-1}\eta Y_1=Y_1(\eta Y_1)^{N-1}. \quad (16)$$

The total yield of gamma rays per muon is[26] $(1-\eta)$ times the expected number of fusions per muon:

$$Y_\gamma=(1-\eta)(Y_1+Y_2+Y_3+\cdots)$$
$$=(1-\eta)Y_1/(1-\eta Y_1). \quad (17)$$

The yields Y_1 and Y_γ are evaluated for each row of Table II.

The first generation yield of muons (at 1% deuterium concentration) is[21,27] $\eta Y_1=0.0264\pm0.0035$. By comparing this value with the product of η and column 5 of Table II, we find that

$$\eta=0.22\pm0.03;\quad Y_\gamma=0.099^{+0}_{-0.005}. \quad (18)$$

This is in contrast to the measured[21] value $Y_\gamma=0.14 \pm0.024$. If it can be said with certainty that Y_γ is greater than 10%, then from (18) we must conclude that the assumption of no quartet capture is inconsistent with experiment.

In Table II, the value for λ_r corresponding to (17) is 0.216 ± 0.009 μsec^{-1}.

The S-Wave Cross Section for $D(p,\gamma)He^3$

The S- and P-wave cross sections σ_s, σ_p for the reaction $D(p,\gamma)He^3$ have been measured[20] for proton energies as low as 24 keV in the lab system. σ_p is the larger and more accurately known of the two, and can be used to compute the fraction of λ_r which comes from the relative P wave of the proton and deuteron in $p\mu d$. We have not yet accomplished this, but our preliminary results are in agreement with other authors,[10] who find that the P-wave contribution is a small part of λ_r. We shall therefore ignore P-wave effects for the remainder of this section, and assume that λ_r comes entirely from the S wave. Following Ref. 21, we assume that an energy-independent reaction constant R accounts for both σ_s and λ_r:

$$\sigma_s=[2\pi\eta/(e^{2\pi\eta}-1)]R/v;$$
$$\tfrac{1}{3}\lambda_r=R|G(0)|^2. \quad (19)$$

Here v is the speed of the proton with respect to the deuteron, and $\eta=e^2/\hbar v$ is the dimensionless variable which enters into the Coulomb barrier penetration factor (in brackets) in Eqs. (19). The factor $\frac{1}{3}$ is the probability that the pd system is in the doublet state.

Elimination of R from (19) yields a relationship between σ_s, λ_r, and $|G(0)|^2$, which can be used to calculate any one of them, given the other two. Using the above estimate of λ_r and the value of $|G(0)|^2$ from Sec. II, we obtain an estimate $\sigma_s=0.39\times10^{-32}$ cm² for 25-keV

[26] Contrary to appearance, this result agrees with Eq. (A13) of Ref. 21; since what is called η there is λ_{nr}/λ_r.
[27] M. Schiff, Nuovo Cimento 22, 66 (1961).

protons, in contrast to the measured[20] value: $(1.3\pm0.3)\times10^{-32}$ cm^2. Because of this discrepancy, either an independent calculation of $|G(0)|^2$ or a more precise measurement of σ_s would be desirable. Previous estimates[19] of $|G(0)|^2$ only make the discrepancy worse.

Comparison of D(p,γ)He^3 and D(n,γ)H^3

The reaction constant R can be written as the product $k(\frac{1}{3}M_d^2+\frac{2}{3}M_q^2)Q^3$, where Q is the energy of the gamma ray, k is a constant (which need not concern us here), and M_d, M_q are nuclear matrix elements for the magnetic dipole transition[28] from the doublet and quartet p-d states to He^3. Substituting the above expression for R in Eq. (19), and writing a similar equation for D(n,γ)H^3, we have

$$\sigma_{sp}=[2\pi\eta/(e^{2\pi\eta}-1)]k(\tfrac{1}{3}M_{dp}^2+\tfrac{2}{3}M_{qp}^2)Q_p^3/v_p, \quad (20)$$

$$\sigma_{sn}=k(\tfrac{1}{3}M_{dn}^2+\tfrac{2}{3}M_{qn}^2)Q_n^3/v_n, \quad (21)$$

where the subscripts p, n refer to the nucleon being captured by d. Eliminating k from these equations and using the measured[29] value $\sigma_{sn}=0.60\pm0.05$ mb for thermal ($v_n=2.2\times10^5$ cm/sec) neutron capture, we obtain a relationship between the M's and σ_{sp}. For our estimate of σ_{sp} above, we obtain approximately

$$\tfrac{1}{3}M_{dn}^2+\tfrac{2}{3}M_{qn}^2=\tfrac{1}{3}M_{dp}^2+\tfrac{2}{3}M_{qp}^2. \quad (22)$$

For the measured value[20] of σ_{sp}, the right-hand side of (22) is about three times as large as the left-hand side.

By assuming that the magnetic dipole operator is of isovector type[23] and neglecting Coulomb effects other than the penetration factor, one finds that $M_{dn}^2=M_{dp}^2$ and $M_{qn}^2=M_{qp}^2$, which is equivalent to (22) plus the statement that

$$M_{qn}^2/M_{dn}^2=M_{qp}^2/M_{dp}^2. \quad (23)$$

In principle, it is possible to test (23) experimentally. The left-hand side could be determined by a measurement of σ_{ns} using polarized neutrons and a polarized deuterium target, but this would be a very difficult experiment.[30] The right-hand side would also be very difficult to determine from a scattering experiment but can be found more easily from the time distribution and absolute yield of gamma rays from $p\mu d$ fusion, as we shall show in the next section.

IV. CAPTURE FROM THE QUARTET STATE

In the last section we saw that M_{dn}^2, M_{qn}^2, M_{dp}^2, M_{qp}^2 are quantities that can be determined experimentally, although only the combination $\frac{1}{3}M_{dn}^2+\frac{2}{3}M_{qn}^2$ is presently known with any confidence. The purpose of this section is to show how one can determine not only the statistical combination $\frac{1}{3}M_{dp}^2+\frac{2}{3}M_{qp}^2$, but also the ratio $\epsilon=2M_{qp}^2/M_{dp}^2$ from experiments on $p\mu d$ fusion. This has not been done before, since a theoretical argument[10] to the effect that $\epsilon\ll1$ has been generally accepted.[3,21] The argument is based on an estimate of M_{dp} which ignores the orthogonality of the initial and final states, whereas for M_{dn} it is well known[22,23] that this orthogonality relation must be used to obtain meaningful results. The two cases, n and p capture by d, are equivalent in this respect. In either case, after the spin-isospin indices have been summed over, the doublet and quartet matrix elements are remarkably similar in form (see Ref. 23). The question of which is larger depends on the details of the space dependence of the initial and final states, and perhaps of the magnetic dipole operator as well. In any event, it seems unlikely that one of the matrix elements should be orders of magnitude smaller than the other.

Fusion Rates and Time Distribution for Arbitrary ϵ

The rates from the doublet pd state in $p\mu d$, which were called λ_r, λ_{nr}, and λ_f in the last section, shall hereinafter be denoted by d_r, d_{nr}, and d_f. The corresponding quartet rates are q_r, q_{nr}, and q_f. We generalize the previous results by finding the various rates and yields for an arbitrary value of the parameter

$$\epsilon=2M_{qp}^2/M_{dp}^2=2q_r/d_r. \quad (24)$$

By definition, we have $d_f=d_r+d_{nr}$ and $q_f=q_r+q_{nr}$. But we have[10] $q_{nr}=0$ by the electric monopole ($E0$) selection rules; hence $q_f=q_r$. Thus for a given value of ϵ we have no more degrees of freedom than we did before.

As in Sec. III, we take $A(0)=5$, $B(0)=C(0)=3$, and $D(0)=1$ for the initial populations of the four $p\mu d$ hyperfine states. The differential equations are

$$\begin{aligned}\dot{A}(t)&=(5/12)d_{nr}(\beta B+\alpha C+D)-(q_f+\lambda_0)A,\\ \dot{B}(t)&=\tfrac{1}{4}d_{nr}(\beta B+\alpha C+D)-(\alpha q_f+\beta d_f+\lambda_0)B,\\ \dot{C}(t)&=\tfrac{1}{4}d_{nr}(\beta B+\alpha C+D)-(\beta q_f+\alpha d_f+\lambda_0)C,\\ \dot{D}(t)&=\tfrac{1}{12}d_{nr}(\beta B+\alpha C+D)-(d_f+\lambda_0)D.\end{aligned} \quad (25)$$

Substitution of the initial populations into (25) yields

$$\begin{aligned}\dot{A}(0)&=5(\tfrac{1}{3}d_{nr}-q_f-\lambda_0),\\ \dot{B}(0)&=d_{nr}-3(\alpha q_f+\beta d_f+\lambda_0),\\ \dot{C}(0)&=d_{nr}-3(\beta q_f+\alpha d_f+\lambda_0),\\ \dot{D}(0)&=\tfrac{1}{3}d_{nr}-(d_f+\lambda_0).\end{aligned} \quad (26)$$

The fusion rate at time t is

$$n_\gamma(t)=q_rA+(\alpha q_r+\beta d_r)B+(\beta q_r+\alpha d_r)C+d_rD. \quad (27)$$

From (26), (27), the initial populations, and the fact that $\alpha+\beta=1$, we obtain

$$n_\gamma(0)=8q_r+4d_r, \quad (28)$$

[28] The electric quadrupole contribution has been shown to be negligible: N. Austern, Phys. Rev. 85, 147 (1951). See also Ref. 10.
[29] F. T. Jurney and H. T. Motz (unpublished).
[30] M. R. Yearian (private communication).

$$\dot{n}_\gamma(0)=5q_r(\tfrac{1}{3}d_{nr}-q_f)+(\alpha q_r+\beta d_r)[d_{nr}-3(\alpha q_f+\beta d_f)] +(\beta q_r+\alpha d_r)[d_{nr}-3(\beta q_f+\alpha d_f)] +d_r(\tfrac{1}{3}d_{nr}-d_f)-\lambda_0(8q_r+4d_r). \quad (29)$$

Substituting q_r for q_f and d_r+d_{nr} for d_f in (28) and (29), and using (24) to eliminate q_r, we obtain

$$n_\gamma(0)=4d_r(1+\epsilon), \quad (30)$$

$$\dot{n}_\gamma(0)=-4d_r[Rd_r+Sd_{nr}+\lambda_0(1+\epsilon)], \quad (31)$$

where

$$R=\tfrac{5}{16}\epsilon^2+\frac{3}{4}\left[\left(\alpha\frac{\epsilon}{2}+\beta\right)^2+\left(\beta\frac{\epsilon}{2}+\alpha\right)^2\right]+\tfrac{1}{4},$$
$$S=-\tfrac{1}{3}(1+\epsilon)+\frac{3}{4}\left[\beta\left(\alpha\frac{\epsilon}{2}+\beta\right)+\alpha\left(\beta\frac{\epsilon}{2}+\alpha\right)\right]+\tfrac{1}{4}. \quad (32)$$

Substitution of (13) into (12) yields

$$\lambda_c(0)=\lambda_0+0.76607\lambda_{fx}. \quad (33)$$

Equation (11) defines $\lambda_c(t)$, and (33) defines λ_{fx} in terms of $\lambda_c(0)$. From (11), (30), (31), and (33), we obtain

$$Rd_r+Sd_{nr}=0.76607\lambda_{fx}(1+\epsilon). \quad (34)$$

Equation (34) is a linear relationship between d_r and d_{nr}, for fixed α, β, ϵ, and λ_{fx}. In order to determine d_r and d_{nr} we need one more equation, which we get by fixing the value of η. Experimentally, η is defined to be the fraction of fusion events which go by the nonradiative mode. If $\epsilon\neq 0$, then we no longer have $\eta=d_{nr}/(d_r+d_{nr})$ as we did in Sec. III, but rather

$$\eta=P_{nr}/(P_r+P_{nr}), \quad (35)$$

where P_r and P_{nr} are the probabilities that a $p\mu d$ will undergo fusion by the radiative and nonradiative modes, respectively:

$$P_r=\frac{1}{12}\frac{d_r}{d_f+\lambda_0}+\frac{1}{4}\left\{\frac{\alpha d_r+\beta q_r}{\alpha d_f+\beta q_f+\lambda_0}+\frac{\beta d_r+\alpha q_r}{\beta d_f+\alpha q_f+\lambda_0}\right\}+\frac{5}{12}\frac{q_r}{q_f+\lambda_0}, \quad (36)$$

$$P_{nr}=\frac{1}{12}\frac{d_{nr}}{d_f+\lambda_0}+\frac{1}{4}\left\{\frac{\alpha d_{nr}}{\alpha d_f+\beta q_f+\lambda_0}+\frac{\beta d_{nr}}{\beta d_f+\alpha q_f+\lambda_0}\right\}.$$

The needed equation is obtained by substituting (36) into (35) and eliminating $q_r=q_f$ by means of (24). However, these substitutions lead to a quartic equation for d_r (or d_{nr}), which is difficult to solve exactly. An easier approach is to make the approximation $\alpha=1$, $\beta=0$ in (36), so that we only have to solve a quadratic equation in d_r. We shall see that once we have found the solution in this approximation, the exact solution can be found by an iterative scheme.

Solution for the Fusion Rates

The problem formulated above is as follows: α, β, ϵ, λ_{fx}, and η are constants. First find d_r, d_{nr}, and q_r in the approximation that

$$P_r=\frac{1}{3}\frac{d_r}{d_r+d_{nr}+\lambda_0}+\frac{2}{3}\frac{q_r}{q_r+\lambda_0},$$
$$P_{nr}=\frac{1}{3}\frac{d_{nr}}{d_r+d_{nr}+\lambda_0}, \quad (37)$$

which comes from setting $\alpha=1$, $\beta=0$ in (36). Then use the approximate solution to find the exact one.

Introducing the auxiliary quantities $\theta=(1-\eta)/\eta$ and $z=q_r/(q_r+\lambda_0)$, rewriting (35) as $\theta=P_r/P_{nr}$, and substituting (37) into this latter equation, we obtain

$$(\theta-2z)d_{nr}=d_r+2z(d_r+\lambda_0). \quad (38)$$

From (24) and the definition of z, we have $2z=\epsilon d_r/(\lambda_0+\frac{1}{2}\epsilon d_r)$. Substituting this expression into (38), we obtain

$$[\theta(\lambda_0+\tfrac{1}{2}\epsilon d_r)-\epsilon d_r]d_{nr}=d_r(\lambda_0+\tfrac{1}{2}\epsilon d_r)+\epsilon d_r(d_r+\lambda_0). \quad (39)$$

Solving (39) for d_{nr} and substituting the resulting expression into (34), we obtain a quadratic equation for d_r:

$$ad_r^2+bd_r+c=0,$$

where

$$a=-\tfrac{3}{2}S\epsilon-R\epsilon(\tfrac{1}{2}\theta-1),$$
$$b=-S\lambda_0(1+\epsilon)-R\theta\lambda_0+\epsilon(1+\epsilon)(\tfrac{1}{2}\theta-1)(0.76607\lambda_{fx}), \quad (40)$$
$$c=(1+\epsilon)(0.76607\lambda_{fx})\theta\lambda_0.$$

The correct solution is the one which reduces to $d_r=-c/b$ when $\epsilon=0$:

$$d_r=2c/[-b+(b^2-4ac)^{1/2}]. \quad (41)$$

To summarize, the approximate solution is found as follows: R and S are found from (32), and $\theta=(1-\eta)/\eta$ is evaluated. Then a, b, and c are computed from (40), and d_r is evaluated from (41). Finally, the other rates (d_{nr} and q_r) are obtained from d_r by solving (34) and (24).

The exact solution is found as follows: The approximate solution is found by the method described above, but with η replaced by an arbitrary number η'. The values of d_r, d_{nr}, and q_r so obtained are used to calculate a different value of η (call it η'') by means of (35) and (36). Then η' is replaced by $\eta'+\eta-\eta''$. The whole process is repeated several times, until η'' is very close to η. At this point we have (very nearly) the exact solution, because the rates satisfy both (34) and (very nearly) (35), where P_r and P_{nr} are given by the exact expressions (36) instead of the approximations (37).

We have computed the exact solution for the same values of α, β, η, and λ_{fx} as appear in Table II, but for $\epsilon=0.1$, 0.2, and 0.3. For each set of values of α, β, η, and

TABLE III. Dependence of σ_s and Y_γ on α, β, η, and ϵ.

Twice the ratio of the quartet to the doublet radiative rate ϵ	Interpolated value of λ_{fx}	Factor by which σ_s increases	Total yield of gamma rays Y_γ
(i) $\alpha=0.86$, $\beta=0.14$, $\eta=0.15$			
0.1	0.333	1.28	0.145
0.2	0.354	1.55	0.175
0.3	0.365	1.80	0.201
(ii) $\alpha=0.86$, $\beta=0.14$, $\eta=0.19$			
0.1	0.336	1.28	0.141
0.2	0.359	1.57	0.171
0.3	0.373	1.82	0.198
(iii) $\alpha=0.923$, $\beta=0.077$, $\eta=0.15$			
0.1	0.324	1.30	0.131
0.2	0.350	1.62	0.161
0.3	0.367	1.91	0.189
(iv) $\alpha=0.923$, $\beta=0.077$, $\eta=0.19$			
0.1	0.326	1.30	0.127
0.2	0.354	1.63	0.156
0.3	0.374	1.94	0.185

ϵ, we have interpolated λ_{fx} (as in Sec. III) to make λ_c (3 μsec) agree with Ref. 21. The interpolated values of λ_{fx} appear in Table III, along with corresponding values of the total yield Y_γ, and the factor by which $\frac{1}{3}d_r+\frac{2}{3}q_r$ increases over its value at $\epsilon=0$. This latter quantity is also the factor by which the estimate of σ_{sp} in Sec. III increases.

From Table III we see that Y_γ increases rapidly with ϵ, but is relatively insensitive to η. Thus an accurate measurement of Y_γ would yield an accurate value of ϵ, even though η is only known to an accuracy of about 15%. From Table III, we also see that variations in α and β have a greater effect on Y_γ (for fixed ϵ) than variations in η. Thus, in order to determine the relationship between ϵ and Y_γ, it is more important to know the muon-nucleus overlaps in $p\mu d$ (which determine α and β) than to improve the measurement[27] of the yield of rejuvenated muons (from which η is derived).

From the measured[21] value $Y_\gamma=0.14\pm0.024$, we see that $\epsilon\lesssim0.25$, and that our estimate of σ_{sp} in Sec. III should be multiplied by a factor somewhere between 1 and 1.75. In any case, Eq. (22) still holds approximately for our estimate of σ_{sp}.

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APPENDIX A

In this Appendix we compute the matrix elements of the kinetic and potential energy operators (whose sum is the Hamiltonian) and of unity, between two S states $u(r_1,r_2,r_{12})$, $v(r_1,r_2,r_{12})$, each of which is of form[31] (3). The matrix elements of the potential energy and of unity can be written down immediately in terms of the coordinates introduced in Sec. II. In atomic[32] units, we have

$$V=\langle u|(1/r_{12})-(1/r_1)-(1/r_2)|v\rangle,\quad U=\langle u|v\rangle,\quad (A1)$$

where the integration is with respect to the volume element discussed below [see (A7)].

For the kinetic energy, we start with coordinates $\mathbf{r}_{12}$ from nucleus 1 to nucleus 2, and $\mathbf{r}_{12,\mu}$ from the center of mass of 1 and 2 to the muon. The kinetic energy separates in these coordinates; it is[32]

$$\hat{T}=-\frac{1}{2}\left[\left(\frac{1}{m_1}+\frac{1}{m_2}\right)\nabla_{12}{}^2+\left(\frac{1}{m_1+m_2}+\frac{1}{m_\mu}\right)\nabla_{12,\mu}{}^2\right],\quad (A2)$$

where the symbol ∇ with a subscript always means gradient with respect to the corresponding vector. A linear transformation of coordinates yields

$$\hat{T}=-\tfrac{1}{2}[(1/m_1+1/m_\mu)\nabla_1{}^2+(1/m_2+1/m_\mu)\nabla_2{}^2+(2/m_\mu)\nabla_1\cdot\nabla_2],\quad (A3)$$

where $\mathbf{r}_1$, $\mathbf{r}_2$ are vectors from nuclei 1 and 2 to the muon.

To compute the matrix element of $\hat{T}$, we make free use of integration by parts. Our functions fall off exponentially at large distances, so there are no surface terms. We obtain for the matrix element

$$T=\frac{1}{2}\int\int\left\{\left(\frac{1}{m_1}+\frac{1}{m_\mu}\right)(\nabla_1u)\cdot(\nabla_1v)+\left(\frac{1}{m_2}+\frac{1}{m_\mu}\right)(\nabla_2u)\cdot(\nabla_2v)+\frac{1}{m_\mu}[(\nabla_1u)\cdot(\nabla_2v)+(\nabla_1v)\cdot(\nabla_2u)]\right\}d^3r_1d^3r_2.\quad (A4)$$

Introducing unit vectors $\hat{r}_1$, $\hat{r}_2$, $\hat{r}_{12}$ in the directions of $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_1-\mathbf{r}_2$, we write down the identities

$$\nabla_1r_1=\hat{r}_1,\quad \nabla_2r_2=\hat{r}_2,\quad \nabla_1r_{12}=\hat{r}_{12},\quad \nabla_2r_{12}=-\hat{r}_{12}.$$

By the chain rule, when operating on S states, we have

$$\nabla_1=\hat{r}_1\partial_1+\hat{r}_{12}\partial_{12},\quad \nabla_2=\hat{r}_2\partial_2-\hat{r}_{12}\partial_{12},\quad (A5)$$

[31] Integrals of this form can also be evaluated by means of Fourier transforms. See L. I. Schiff, Phys. Rev. **125**, 777, Appendix (1962).
[32] In atomic units, the electronic charge and mass are 1, and the masses of the muon, proton, deuteron, and triton are 206.77, 1836.12, 3670.4, and 5496.8, respectively. Also 1 a.u. = 27.2098 eV.

where ∂_1, ∂_2, ∂_{12} are partial differentiations by r_1, r_2, and r_{12}. From (A4) and (A5), we obtain

$$T=\frac{1}{2}\int\int\left\{\left(\frac{1}{m_1}+\frac{1}{m_\mu}\right)(\partial_1u)(\partial_1v)+\left(\frac{1}{m_2}+\frac{1}{m_\mu}\right)(\partial_2u)(\partial_2v)+\left(\frac{1}{m_1}+\frac{1}{m_2}\right)(\partial_{12}u)(\partial_{12}v)\right.$$
$$+\frac{r_1^2+r_2^2-r_{12}^2}{2m_\mu r_1r_2}[(\partial_1u)(\partial_2v)+(\partial_1v)(\partial_2u)]+\frac{r_1^2+r_{12}^2-r_2^2}{2m_1r_1r_{12}}[(\partial_1u)(\partial_{12}v)+(\partial_1v)(\partial_{12}u)]$$
$$\left.+\frac{r_2^2+r_{12}^2-r_1^2}{2m_2r_2r_{12}}[(\partial_2u)(\partial_{12}v)+(\partial_2v)(\partial_{12}u)]\right\}d^3r_1d^3r_2. \quad \text{(A6)}$$

Integration over the irrelevant coordinates (e.g., three Euler angles) in (A1) and (A6) has the effect of replacing $d^3r_1d^3r_2$ by the volume element

$$d\tau=8\pi^2r_1r_2r_{12}dr_1dr_2dr_{12}. \quad \text{(A7)}$$

Up to this point our treatment has been valid for any bound S states. We now specialize to functions of the form (3):

$$u=\exp[-S(Ar_1+Br_2+Cr_{12})], \quad v=\exp[-S(A'r_1+B'r_2+C'r_{12})]. \quad \text{(A8)}$$

From (A7) and (A8), Eqs. (A1) and (A6) become

$$U=8\pi^2\int r_1r_2r_{12}e^{-S(mr_1+nr_2+pr_{12})}dr_1dr_2dr_{12},$$

$$V=8\pi^2\int(r_1r_2-r_1r_{12}-r_2r_{12})e^{-S(mr_1+nr_2+pr_{12})}dr_1dr_2dr_{12}, \quad \text{(A9)}$$

$$T=4\pi^2S^2\int\left\{\left[\left(\frac{1}{m_1}+\frac{1}{m_\mu}\right)AA'+\left(\frac{1}{m_2}+\frac{1}{m_\mu}\right)BB'+\left(\frac{1}{m_1}+\frac{1}{m_2}\right)CC'\right]r_1r_2r_{12}+\frac{1}{2m_\mu}(AB'+A'B)(r_1^2+r_2^2-r_{12}^2)r_{12}\right.$$
$$\left.+\frac{1}{2m_1}(AC'+A'C)(r_1^2+r_{12}^2-r_2^2)r_2+\frac{1}{2m_2}(BC'+B'C)(r_2^2+r_{12}^2-r_1^2)r_1\right\}e^{-S(mr_1+nr_2+pr_{12})}dr_1dr_2dr_{12},$$

where

$$m=A+A', \quad n=B+B', \quad p=C+C'. \quad \text{(A10)}$$

We introduce an auxiliary function

$$I(m,n,p)=\frac{1}{2}\int\exp(-mr_1-nr_2-pr_{12})dr_1dr_2dr_{12}. \quad \text{(A11)}$$

This integral is most easily evaluated in perimetric coordinates

$$x=\tfrac{1}{2}(r_1+r_2-r_{12}), \quad y=\tfrac{1}{2}(r_1+r_{12}-r_2), \quad z=\tfrac{1}{2}(r_2+r_{12}-r_1),$$

which run independently from 0 to ∞. One obtains

$$I(m,n,p)=1/[(m+n)(m+p)(n+p)]. \quad \text{(A12)}$$

Differentiation of the right-hand side of (A11) by m, n, or p, is equivalent to multiplying the integrand by $-r_1$, $-r_2$, or $-r_{12}$, respectively. By exploiting this fact, we can represent the matrix elements (A9) as linear combinations of derivatives of I:

$$U=-8\pi^2S^6\partial_m\partial_n\partial_pI(m,n,p),$$

$$V=8\pi^2S^5(\partial_m\partial_n-\partial_m\partial_p-\partial_n\partial_p)I(m,n,p),$$

$$T=-4\pi^2S^8\left\{\left[\left(\frac{1}{m_1}+\frac{1}{m_\mu}\right)AA'+\left(\frac{1}{m_2}+\frac{1}{m_\mu}\right)BB'+\left(\frac{1}{m_1}+\frac{1}{m_2}\right)CC'\right]\partial_m\partial_n\partial_p+\frac{1}{2m_\mu}(AB'+A'B)(\partial_m^2+\partial_n^2-\partial_p^2)\partial_p\right.$$
$$\left.+\frac{1}{2m_1}(AC'+A'C)(\partial_m^2+\partial_p^2-\partial_n^2)\partial_n+\frac{1}{2m_2}(BC'+B'C)(\partial_n^2+\partial_p^2-\partial_m^2)\partial_m\right\}I(m,n,p). \quad \text{(A13)}$$

The combination of (A10), (A12), and (A13), yields the matrix elements as rational functions of A, B, C, A', B', C'. From (A12), it is clear that the integral in (A11) converges if and only if $m+n$, $m+p$, and $n+p$ are all positive. For this to be the case, the condition mentioned in Sec. II is clearly sufficient. For the diagonal elements of (3) to be defined, that condition is also necessary.

APPENDIX B

In this Appendix, we evaluate the integral in (7), for ψ of the form indicated by (2) and (3). In elliptic coordinates

$$x=(r_1+r_2)/r_{12},\quad y=(r_1-r_2)/r_{12},\tag{B1}$$

the volume element is

$$d\tau_\mu=\tfrac{1}{4}\pi r_{12}^3(x^2-y^2)dxdy.\tag{B2}$$

From (2), (3), and (B1), we obtain

$$\psi^2=\sum_{i=1}^{N}\sum_{j=1}^{N}X_iX_j\exp[-S(C_i+C_j)r_{12}-(\alpha_{ij}x+\beta_{ij}y)r_{12}],\tag{B3}$$

where

$$\begin{aligned}\alpha_{ij}&=\tfrac{1}{2}S(A_i+A_j+B_i+B_j),\\ \beta_{ij}&=\tfrac{1}{2}S(A_i+A_j-B_i-B_j).\end{aligned}\tag{B4}$$

It is convenient to express G in terms of auxiliary functions which are slightly different from those of Ref. (10):

$$\begin{aligned}P_n(\alpha)&=\int_1^\infty(-x)^ne^{-\alpha x}dx,\\ W_n(\beta)&=\frac{1}{2}\int_{-1}^{1}(-y)^ne^{-\beta y}dy.\end{aligned}\tag{B5}$$

These integrals are most easily evaluated by noticing that P_n is the nth derivative of P_0 with respect to α, and similarly for W_n. We shall need only the following cases:

$$\begin{aligned}P_0(\alpha)&=e^{-\alpha}/\alpha,\\ P_2(\alpha)&=(\alpha^2+2\alpha+2)e^{-\alpha}/\alpha^3,\\ W_0(\beta)&=\sinh\beta/\beta,\\ W_2(\beta)&=[(\beta^2+2)\sinh\beta-2\beta\cosh\beta]/\beta^3.\end{aligned}\tag{B6}$$

Substituting (B2) and (B3) in (7), and then using (B5), we finally obtain

$$\begin{aligned}G(r_{12})=\{&\sum_{i=1}^{N}\sum_{j=1}^{N}X_iX_j\tfrac{1}{2}\pi r_{12}^3\exp[-S(C_i+C_j)r_{12}]\\ &\times[P_2(\alpha_{ij}r_{12})W_0(\beta_{ij}r_{12})-P_0(\alpha_{ij}r_{12})W_2(\beta_{ij}r_{12})]\}^{1/2}.\end{aligned}\tag{B7}$$