$$g_{p,\xi} = + \frac{r_n p_-}{4} \frac{dp_+}{dr_n} (E_3 - E_1) [C_1(\Sigma) - C_1(\Delta)],$$

$$g_{\xi,p} = \frac{r_n p_+}{4} \frac{dp_-}{dr_n} (E_3 - E_1) [C_1(\Sigma) - C_1(\Delta)],$$

$$g_{q,\xi} = -\frac{r_n p_-}{4} \frac{dq_+}{dr_n} (E_2 - E_0) [C_2(\Sigma) - C_2(\Delta)],$$

$$g_{\xi,q} = -\frac{r_n p_+}{4} \frac{dq_-}{dr_n} (E_2 - E_0) [C_2(\Sigma) + C_2(\Delta)],$$

$$g_{A,\eta} = \frac{1}{A_+} \frac{dA_+}{dr_n} \frac{q_-}{2} E_1 [C_0(\Sigma) + C_0(\Delta) - C_2(\Sigma) - C_2(\Delta)],$$

$$g_{\eta,A} = \frac{1}{A_-} \frac{dA_-}{dr_n} \frac{q_+}{2} E_1 [C_0(\Sigma) - C_0(\Delta) - C_2(\Sigma) + C_2(\Delta)],$$

$$g_{p,\eta} = -\frac{r_n q_-}{4} \frac{dp_+}{dr_n} E_1 [C_0(\Sigma) + C_0(\Delta) - C_2(\Sigma) - C_2(\Delta)],$$
  

$$g_{\eta,p} = -\frac{r_n q_+}{4} \frac{dp_-}{dr_n} E_2 [C_0(\Sigma) - C_0(\Delta) - C_2(\Sigma) + C_2(\Delta)],$$
  

$$g_{q,\eta} = \frac{r_n q_-}{4} \frac{dq_+}{dr_n} E_1 [C_1(\Sigma) + C_1(\Delta) - C_3(\Sigma) - C_3(\Delta)],$$
  

$$g_{\eta,q} = \frac{r_n q_+}{4} \frac{dq_-}{dr_n} E_1 [C_1(\Sigma) + C_1(\Delta) - C_3(\Sigma) - C_3(\Delta)].$$

Again, for  $r < r_c$ , modifications such as have already been pointed out must be made:  $(1/A_{-})(dA_{-}/dr_{n})$  is replaced by  $(1/A_{-})(dA_{-}/dr_{n}) - (1/r_{n})$ ,  $E_{n}$  by  $E_{n+1}$ , and  $p_{-}E_n$  by  $p_{-}E_{n+1} - (2/r_n)E_n$  in these expressions.

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# µ-Mesonic Molecules. II. Molecular-Ion Formation and Nuclear Catalysis\*

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The methods developed in the preceding paper are applied to the study of the behavior of  $\mu$  mesons in liquid hydrogen. Numerically evaluated energy eigenvalues for the bound states of the various molecular-ion configurations are presented. Phase shifts and cross sections for the scattering of mesonic atoms from hydrogen and deuterium are given. It is shown that in the neighborhood of 0.2 ev the scattering of  $(d\mu)$  atoms from protons exhibits a Ramsauer-Townsend effect with an anomalously small cross section occurring in this region. The existence of this effect provides an explanation for the appearance of "gaps" in the experimental observation of the catalytic process. The rate of exchange of mesons from protons to deuterons in pure deuterium is calculated along with the rates of formation of the  $(p\mu p)^+$ ,  $(p\mu d)^+$ , and  $(d\mu d)^+$  molecular ions. It is shown that the predominant mechanism for the formation of the molecular ions is dipole electron ejection. These results are shown to be in agreement with available experimental data. A semiphenomenological treatment of the (pd) nuclear reaction is also given. A rough estimate of the  $\gamma$ -emission process indicates that the dominant mode of emission is from the singlet proton spin states.

#### I. INTRODUCTION

HE theoretical possibility that  $\mu$  mesons could greatly enhance the nuclear fusion of hydrogen and deuterium so that it might be observed experimentally was first suggested by Frank<sup>1</sup> and later was estimated by Zeldovich.<sup>2</sup> Later, this fusion was in fact observed by Alvarez et al. in a liquid-hydrogen bubble chamber in a process in which the energy of fusion was given to the  $\mu$  meson.<sup>3</sup> Still later,  $\gamma$  rays from

the reaction were detected by Ashmore et al. with counters, again in liquid hydrogen.<sup>4</sup> The process has been further investigated theoretically by Tackson<sup>5</sup> and by Skyrme,<sup>6</sup> who have also given phenomenological descriptions in which the reaction is assumed to proceed through the following steps:

(a) A fast  $\mu$  meson is rapidly slowed down and captured to form a hydrogen  $(p\mu)$  or deuterium  $(d\mu)$ mesonic atom. (Because the experiments have been carried out with much more hydrogen than deuterium, it is much more likely that hydrogen mesonic atoms are formed.)

(b) The neutral mesonic hydrogen atom will then move about with thermal energy, colliding with the atoms of the liquid. It may then undergo elastic scatter-

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<sup>&</sup>lt;sup>2</sup> Ya. B. Zeldovitch, Doklady Akad. Nauk. S.S.S.R. 95, 493

<sup>(1954).
\*</sup> L. W. Alvarez, H. Bradner, F. S. Crawford, Jr., J. A. Crawford, P. Falk-Vairant, M. L. Good, J. D. Gow, A. H. Rosenfeld, F. Solmitz, L. Stevenson, H. K. Ticho, and R. D. Tripp, Phys. Rev. 105, 1127 (1957).

<sup>&</sup>lt;sup>4</sup> Ashmore, Nordhagen, Strauch, and Townes, Proc. Phys. Soc. (London) 71, 161 (1958). <sup>5</sup> J. D. Jackson, Phys. Rev. 106, 330 (1957).

<sup>&</sup>lt;sup>6</sup> T. H. R. Skyrme, Phil. Mag. 2, 910 (1957).

ing, exchange of the  $\mu$  meson from a proton to a deuteron, or formation of either a  $(p\mu d)^+$  or a  $(p\mu p)^+$ molecular ion. In either case of molecular-ion formation, no further configurational change is likely to occur because the ion carries a positive charge. It is expected that the exchange from proton to deuteron is much more likely than the  $(p\mu d)^+$  formation. Because of the difference in the reduced mass of the meson in the  $(p\mu)$ and  $(d\mu)$  atoms, the exchange of the meson from proton to deuteron will release about 135 ev; the inverse process therefore does not occur at thermal energies.

(c) The  $(d\mu)$  atom formed may in turn form either a  $(d\mu\phi)^+$  or a  $(d\mu d)^+$  molecular ion, with  $(d\mu\phi)^+$  generally more likely because of the greater abundance of hydrogen in the chamber.

(d) Because of the close proximity of the two nuclei in these molecular ions, nuclear fusion can occur in a time comparable with that of the lifetime of the meson.

(e) In some of these reactions, which normally proceed via  $\gamma$  emission, the  $\gamma$  ray may be internally converted with the ejection of a fast  $\mu$  meson. This  $\mu$  meson is then free to repeat the cycle, thus playing the role of a catalyst for the reaction.

In all of the above steps there is of course a competition with the natural decay of the meson. The observed fusion probabilities indicate, however, that the various steps in the process have rates that are comparable to or greater than the natural decay rate of the meson. In the experiments of Alvarez et al., the number of rejuvenated mesons produced per incident meson was measured after adding various amounts of deuterium to the hydrogen.<sup>3</sup> It was found that for small amounts of deuterium the fraction of mesons rejuvenated increased with increasing concentration, but that for concentrations larger than a few percent no further appreciable increase occurred. In addition, it was found that many events were observed in which the point of production of the fast meson was a considerable distance from the end of the incident meson track. It was also observed that the number of long gaps diminished with increasing deuterium concentration. These gaps were difficult to understand on the basis of the above scheme, and a more refined calculation of the phenomena involved seemed necessary.

The experiment of Ashmore et al. yielded information on the time distribution of the  $\gamma$  rays produced, as measured from the time of injection of the initiating meson into the chamber.<sup>4</sup> These authors made an analysis of this distribution which gave the number of  $\gamma$  rays per incident meson. This number, together with the above result for the number of fast mesons per incident meson, can be used to obtain a value for the internal conversion coefficient in the process.

Because these experiments created considerable interest, and since all of the previous theoretical investiga-

tions7 were essentially qualitative in nature and the explanation of the gaps was not completely clear, we felt that quantitative calculations of all the steps in the process would be worthwhile in further understanding of these experiments.

We have carried out such calculations based on a variational approximation to the wave function of the three-body system. This approximation is described in the preceeding paper which will henceforth be referred to as I.<sup>8</sup> The solutions of the equations developed therein were obtained numerically with the use of an IBM-701 digital computer. In this manner we obtain the eigenvalues of the energy for bound states, and the elasticand exchange-scattering cross sections for the free states. From the calculated wave functions, we have determined the rates of molecular-ion formation in the  $(p\mu p)^+$ ,  $(d\mu d)^+$ , and  $(p\mu d)^+$  systems using perturbation theory. In addition, we have made a more detailed investigation into the nuclear processes involved in the  $(p\mu d)^+$  reaction and have obtained a somewhat different time dependence for the  $\gamma$  rays than was obtained by Ashmore et al. using the previous phenomenological theory. Specifically, we have found that the nuclear reactions will take place predominantly from the singlet spin states of the two protons involved and not from the triplet states, and that the reactions will proceed from various spin channels quite independently. Our re-analysis of the data of Ashmore et al. has then led us to a considerably lower value than they had obtained for the number of  $\gamma$  rays per incident  $\mu$  meson.

## II. SOLUTION OF THE BOUND STATE AND SCATTERING EQUATIONS

# A. Numerical Technique

Solutions to the radial differential equations for the nuclear wave functions, Eqs. (2) of I, were obtained numerically by the use of the procedure of Milne<sup>9</sup> with a four-point integration formula. In this method the solution is advanced to successive grid points by first obtaining predicted values for the first derivative,  $y_n'$ , of the wave function,  $y_n$ , at the new grid point by the use of the "open" formula:

$$y_{n+1}{}^{\prime(P)} = y_{n-3}{}^{\prime} + \frac{4}{3}\Delta r (2y_{n}{}^{\prime\prime} - y_{n-1}{}^{\prime\prime} + 2y_{n-2}{}^{\prime\prime}),$$

where  $r_n = r_0 + n\Delta r$ ,  $\Delta r$  is the spacing of the grid points, and  $y_n''$  is the second derivative of the wave function.

(1960), preceding paper.

<sup>9</sup> James B. Scarborough, Numerical Mathematical Analysis (Johns Hopkins Press, Baltimore, 1950), second edition, p. 293.

<sup>&</sup>lt;sup>7</sup> Gallone, Prosperi, and Scotti, Nuovo cimento 6, 168 (1957); Hagashi, Nakano, Nishida, Suakane, and Yamaguchi, Progr. Theoret. Phys. (Kyoto) 17, 615 (1957); H. Marschall and Th. Schmidt, Z. Physik 150, 293 (1958); M. Schmizu, Y. Mizumo and T. Izuyama, Progr. Theoret. Phys. (Kyoto) 20, 777 (1958) have treated parts of this problem using approximations which are similar in nature to methods presented here. Note added in proof.---More recently, calculations on collision processes involving mu mesic atoms were reported by T.-Y. Wu at the 12th Annual Gaseous Electronics Conference held at the National Bureau of Standards, Washington, D. C., October 14–16, 1959. <sup>8</sup> S. Cohen, D. L. Judd, and R. J. Riddell, Phys. Rev. **119**, 384

These are then used to obtain predicted values of the wave functions by the use of Simpson's rule:

$$y_{n+1}(P) = y_{n-1} + \frac{1}{3}\Delta r(y_{n+1}'(P) + 4y_n' + y_{n-1}').$$

These predicted values are in turn used in the differential equations in order to calculate predicted values for the second derivatives. From the predicted values, more accurate corrected values for the first derivatives are calculated, again by the use of Simpson's rule (closed formula):

$$y_{n+1}{}^{\prime(C)} = y_{n-1}{}^{\prime} + \frac{1}{3}\Delta r(y_{n+1}{}^{\prime\prime(P)} + 4y_{n}{}^{\prime\prime} + y_{n-1}{}^{\prime\prime}).$$

These corrected values of the first derivatives are assumed to be their true values. Finally,  $y_{n+1}$  and  $y_{n+1}''$ are recalculated by the use of the corrected value of  $y_{n+1}'$ . Starting values for the functions at four grid points are required in order to begin this integration scheme. For small values of r, these values were obtained by use of the power-series expressions for the wave functions given in Sec. II C of I. For this purpose it was necessary to express the various quantities appearing in the differential equation in terms of power series valid near the origin. In general, five terms in the power series were fitted to each of the required functions.

In a classically forbidden region, this numerical integration technique is stable about a solution with monotonically increasing magnitude. If one attempts to obtain a solution with decreasing amplitude, propagated rounding errors will introduce solutions with increasing amplitude, and these will eventually dominate over the desired solution. For this reason, in the case of bound states (and certain free states in the unequal-mass case<sup>10</sup>), it was necessary to calculate the wave functions for large r by integrating in the direction of decreasing r. In these cases, starting values for the wave functions at an arbitrarily chosen large value of r were obtained by the use of the asymptotic behavior of the solutions for  $r \to \infty$ . For the calculations, this large value of r was chosen as  $20a_{\mu}$ , where  $a_{\mu}$  is the Bohr radius in the mesonic hydrogen atom. For values of r less than  $r_0$ , where  $r_0$  is in the vicinity of the classical turning point,

TABLE I. Eigenvalues for various bound states.

System	L	W (ev)	
¢μ	0	2530	
$d\mu$	0	2664	
$(p\mu p)^+$	. 0	2771	
	1	2623	
$(p\mu d)^+$	0	2878	
	1	2754	
$(d\mu d)^+$	0	2986	
	0	2845	
	1	2887	
	2	2746	

<sup>10</sup> For the scattering states in which the energy of the system lies between that of the free  $(p\mu)$  and  $(d\mu)$  atoms, the wave function will generally have an asymptotically decreasing part. It is therefore necessary to follow a similar procedure in such cases.



FIG. 1. Bound-state radial wave functions for the  $(p\mu d)^+$  molecular ion.

the integration was done in an outward direction, while for r larger than  $r_0$ , the inward direction was chosen.

In this numerical work the integration steps were chosen to be  $0.05a_{\mu}$ . In Appendix A we give a discussion of a test problem that was used to check the accuracy of the problem codes. The equal mass and unequal mass cases were coded independently, and the test problem was constructed to convert the two uncoupled differential equations of the equal-mass case to coupled equations similar to those occurring in the unequal-mass case. Aside from rounding errors, the two methods of solution agreed completely, thus providing a good check on the two programs. In addition, the unitarity current developed in I, Sec. II D, was computed at various points and was found to be zero within the rounding errors, as it must be for the regular solutions.

# **B.** Bound States

The bound states of the system were obtained by the use of the variational procedure of I, Sec. IV. As was stated there, the  $(p\mu p)^+$  and  $(d\mu d)^+$  systems involve only the symmetric states, whereas the  $(p\mu d)^+$  case requires the solution of the coupled equations. In Table I we give the computed eigenvalues for the binding energy, W, of all the bound states for various angular momentum, L, for each case. The binding energies given are to be compared to zero for a totally separated system. Also included in this table are the binding energies for the atoms.

In Fig. 1, we give the solutions  $\phi_+$  and  $\phi_-$  for the l=0 and l=1 bound states in the  $(\not p\mu d)^+$  case. In the vicinity of the minimum in the  $V_+$  potential  $(r_n \sim 2)$ ,  $\phi_-$  is considerably smaller than  $\phi_+$ , while as  $r_n$  approaches  $\infty$ ,  $|\phi_-|$  approaches  $|\phi_+|$ ; this results from the greater binding of the meson on the deuteron than on the proton (see I, Sec. III A).

## C. Scattering States

We have used the methods of I to calculate the wave functions for the free states of the various systems. In the  $\mu$ -meson catalysis experiments, the energies of interest are quite small, however, and it is necessary to consider corrections to these solutions. The energies of

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(1)

interest range from something less than a few hundred electron volts down to thermal energies in liquid hydrogen ( $\sim 1/400$  ev).

The numerical calculations were carried out to a distance  $r_n = 20a_{\mu}$  for several reasons. The potential at this distance is quite small, and it was found that beyond this point it is well represented analytically, therefore analytic corrections for  $r_n > 20a_{\mu}$  could be obtained. In addition, the memory storage in the numerical calculations limited  $r_n$  to a value near  $20a_{\mu}$ . The most important part of the long-range force, varying as  $r_n^{-4}$ , (which is given exactly by second order perturbation theory) has been calculated by Dalgarno and Lewis.<sup>11</sup> They find

$$V(r_n) = -\left(\frac{9}{2\epsilon}\right)r_n^{-4},$$

where  $\epsilon$  is the parameter defined in I. For our variational wave function, one finds that asymptotically  $V \approx -(4/\epsilon)r_n^{-4}$ , which is close to the exact result. To obtain the correction to the phase shift resulting from the wave-function integration from  $r_n = 20a_{\mu}$  to  $\infty$ , we have used the potential obtained by Dalgarno and Lewis. Because the potential is small in this region, we anticipate that the correction to the phase shift will also be small. However, as we are interested in the limit of very small energy, it is not possible to use either the Born or the WKB approximation to obtain estimates of the corrections, because neither is valid in this situation. Rather, we have chosen to use a method of variation of constants to provide the approximate solution desired.

To develop this approximation for S-wave scattering, we define the variables A(r),  $\delta(r)$  by the equations:

 $\psi(r)/r \equiv \phi(r) \equiv A(r) \sin[kr + \delta(r)],$ 

and

$$d\phi(\mathbf{r})/d\mathbf{r} \equiv kA(\mathbf{r}) \cos[k\mathbf{r} + \delta(\mathbf{r})],$$

where k is defined as  $[(W+1-0.25\epsilon)/\epsilon]^{\frac{1}{2}}$ . These equations then imply the constraint:

$$\frac{dA}{dr}\sin[kr+\delta(r)] + A\cos[kr+\delta(r)]\frac{d\delta}{dr} = 0.$$

In addition, the Schrödinger equation,

$$d^2\phi/dr^2+[k^2-V(r)]\phi=0,$$
 leads to

$$\frac{dA}{dr}\cos[kr+\delta(r)] - kA\sin[kr+\delta(r)]\frac{d\delta}{dr}$$

Thus, we obtain

$$\frac{d\delta}{dr} = -\frac{V(r)}{k} \sin^2[kr + \delta(r)], \qquad (2)$$

 $= VA \sin[kr + \delta(r)].$ 

<sup>11</sup> A. Dalgarno and J. T. Lewis, Proc. Roy. Soc. (London) A233, 70 (1956).

and

$$\frac{dA}{dr} = \frac{AV(r)}{2k} \sin\{2[kr + \delta(r)]\}.$$

In our problem, the numerical solution of the equation for  $r_n = 20$  will provide the boundary value for  $\delta(r)$  for r > 20.<sup>12</sup> The true phase shift will then be given by  $\delta = \lim_{r \to \infty} \delta(r)$ . It should be pointed out that this approach is not correct for the  $(p\mu d)^+$  system if exchange is energetically possible, because in this case the asymptotic form is not given correctly by Eq. (1). However, it is only for very small energies that the present corrections are necessary, and in this case the equation for  $\phi_d$  for  $r_n \ge 20$  is essentially uncoupled. Now, we have  $V(r) \approx C/r^4$ , and a posteriori one can show that  $\delta - \delta(20)$  is much less than 1, so that we may write

$$\delta \approx \delta(20) + \frac{C}{k} \int_{20}^{\infty} \frac{\sin^2[kr + \delta(20)]}{r^4} dr,$$
  
or  
$$\delta \approx \delta(20) + \frac{C}{2k} \left\{ \frac{1}{3 \times (20)^3} - (2k)^3 \left[ \cos 2\delta(20) \int_{40k}^{\infty} \frac{\cos u}{u^4} du - \sin 2\delta(20) \int_{40k}^{\infty} \frac{\sin u}{u^4} du \right] \right\}$$

In the limit  $E \rightarrow 0$ , it is possible with the  $r^{-4}$  potential to solve the Schrödinger equation exactly, with the result that we obtain

$$\phi(r) = Ar \sin(C^{\frac{1}{2}}/r + \theta)$$

where  $A, \theta$  are constants. From this, for  $(C/20^2) \ll 1$ , we obtain an approximate value for a, the scattering length,

$$a = a(20) + \frac{C}{3 \times (20)^3} \{a(20)^2 + 3 \times 20[a(20) + 20]\},\$$

where a(20) is the scattering length computed from calculated values of  $\phi$  and  $d\phi/dr_n$  at  $r_n=20$ . Because we have  $a=\lim_{k\to 0}(\delta/k)$ , one finds that the approximate solution of the Schrödinger equation agrees with the exact solution in the limit  $E \to 0$ , and for  $C/(20)^2 \ll 1$ . The latter requirement is necessary in order that we may drop  $\delta(r) - \delta(20)$  in the argument of  $\sin^2[kr + \delta(r)]$ .

In Appendix B we analyze the additional effects on these phase shifts which might be expected because of the presence of electrons. It is shown there that for the energies of interest these contributions are negligible.

In Fig. 2 we have plotted the phase shifts in the  $(p\mu)+p^+$  and  $(d\mu)+d^+$  systems both for the symmetric (+) and antisymmetric (-) wave functions including

<sup>&</sup>lt;sup>12</sup> If one wishes to use this equation for all values of r, it is necessary that  $\delta(r) \to 0$  as  $r \to 0$  if V(r) is well behaved as  $r \to 0$ . For singular V(r), a special treatment is necessary. At discontinuities in V(r),  $\delta(r)$  must be continuous. One obtains an approximate form of the *WKB* expression by setting  $\sin^2(kr+\delta) = \frac{1}{2}$ , which is valid for  $V/E \ll 1$ .

the above correction to the phase shifts. In accordance with the usual convention we have set  $\delta = 0$  for k = 0. For several values of k, an individual phase shift can have the value  $n\pi$ , so that its contribution to the scattering cross section will be zero. However, in general, the second phase shift for these systems will still contribute to the cross section.

The  $(d\mu) + p^+$  system has quite different properties. For small energies only one phase shift is necessary to describe the scattering state. The behavior of this phase shift for small energies is given in Fig. 3 both with and without the asymptotic correction. For small k, the phase without correction is small, leading to a scattering length of only  $\sim 0.8a_{\mu}$ . The correction is important in this case and, as one sees, with the correction the phase shift actually changes sign for  $k \sim 0.02$  (energy  $\simeq 0.2$ ev). This therefore leads to a Ramsauer-Townsend effect<sup>13</sup> in this energy region, and we would therefore expect an extremely small cross section for scattering.



FIG. 2. Phase shifts for s-wave scattering in the  $(p\mu p)^+$  and  $(d\mu d)^+$  systems as a function of k.

The various scattering cross sections as calculated are given in Fig. 4.

We feel that the anomalous behavior of the  $(d\mu) + p^+$ cross section gives the explanation for the gaps that have been observed between the end of a  $\mu$ -meson track and the point at which the nuclear reaction occurs. The interpretation of the gaps is that the meson is slowed down, captured by a proton, and subsequently exchanged to a deuteron as discussed in the introduction. This neutral mesonic atom acquires an energy of about 100 ev in the exchange and is subsequently slowed down to very low energy. However, as it reaches the Ramsauer energy the scattering cross section becomes very small and the atom can travel large distances without effective collisions with the protons of the liquid hydrogen. The only significant collisions for stopping the atom would be those with deuterons, for which



FIG. 3. The phase shift,  $\delta$ , for the scattering of  $(d\mu)$  atoms by protons, divided by k versus k.

there is no such anomalous behavior, and those with protons which result in the formation of the  $(p\mu d)^+$ molecular ion. The latter is small, as will be seen, while the former will be small for the deuterium concentrations in natural liquid hydrogen. Increasing the deuterium concentration by using hydrogen enriched with deuterium should quench these gaps, as is indeed observed. The cross section for scattering  $(d\mu)$  atoms by deuterons at low energies is found to be  $\sim 40 \times 10^{-20}$ cm<sup>2</sup>. The density of the liquid hydrogen is  $\sim 3.5 \times 10^{22}$ atoms/cm<sup>3</sup>, so that for a deuterium concentration of  $2 \times 10^{-4}$  (natural hydrogen), we obtain a mean free path  $\sim 3$  mm, while for a concentration of 1%, the mean free path is 0.07 mm. These results are in reasonably good agreement with the experiments on the quenching of the gaps with increasing deuterium concentration.14

Although the cross sections for the various processes are of general interest, the exchange cross section,  $\sigma_{pd}$ , for  $(p\mu)$  atoms incident on deuterons is of great importance in the phenomenological analysis of the dependence of the rate of catalysis on the deuterium concentration. This exchange cross section is inversely



<sup>14</sup> Cresti, Gottstein, Rosenfeld, and Ticho, University of California (private communications).

<sup>&</sup>lt;sup>18</sup> See for instance: Leonard I. Schiff, Quantum Mechanics (McGraw-Hill Book Company, New York, 1955), p. 108.

proportional to the incident velocity for small energies. It is to be expected that the  $(p\mu)$  atoms will rapidly slow down and will then exchange at low energy with deuterons to form  $(d\mu)$  atoms. Thus the rate of such exchange is of great significance. From our calculations, we find

$$\lim_{W\to W_p} (\sigma_{pd} v_H) \approx 3.3 \times 10^{-13} \text{ cm}^3/\text{sec},$$

which leads to an exchange rate in pure deuterium of

$$R_{\rm ex} = 1.14 \times 10^{10} \, {\rm sec^{-1}}.$$

It will be seen in the next section that when this rate is taken in conjunction with the molecular-ion formation rate, one obtains reasonable agreement with the experimental dependence on deuterium concentration.

#### **III. MOLECULAR-ION FORMATION**

A. 
$$(pu) + p^+ \rightarrow (pup)^+$$

As has been shown in the phenomenological analysis, it is to be expected that the competition between molecular-ion formation and the exchange process discussed above will determine the dependence of the nuclear reaction rate on the deuterium concentration  $(c_d)$ . We have computed the rates for those mechanisms that seem most significant. These include radiative formation of the molecular ion and the ejection of an electron from its orbit in the hydrogen molecule in which the "free" proton resides. In addition, we have considered in a rough way such processes as three-body collisions, and have found their effects to be small compared to the most significant of the mechanisms treated here.

We will first treat the molecular-ion formation in which an electron is ejected into a p state of the continuum as the nuclear-mesonic system makes a transition from an incident s state to a bound p state. This will turn out to be the dominant formation mode. The perturbation Hamiltonian will be chosen to be

$$3C' = -e^2 \left[ \frac{1}{r_{e1}} + \frac{1}{r_{e2}} - \frac{1}{r_{e\mu}} - \frac{1}{r_{ec}} \right],$$

where the  $r_{ei}$  for i=1, 2, and  $\mu$  are the distances from the electron to the two nuclei, and to the meson, respectively. We will assume that the electron is in the Coulomb field of a fictitious charge at the center of mass of the three "i" particles (in addition to the field of the other proton in the hydrogen molecule), so that  $e^2/r_{ec}$  must be subtracted from the perturbation Hamiltonian, where  $r_{ec}$  is the distance from the electron to the center of mass of the three particles. Because the three particles must be very close to each other as compared to the size of the electron orbit in order that the molecular ion be formed, the precise choice of the perturbation is not important. As an approximate solution to the unperturbed electronic motion in the bound state, we have  $\mathrm{chosen^{15}}$ 

$$\psi_{e}{}^{I} = \left(\frac{(Z')^{3}}{2\pi a_{e}^{3}}\right)^{\frac{1}{2}} \left[e^{-(Z'/a_{e})r_{ee}} + e^{-(Z'/a_{e})r_{ep}}\right](1+\Delta)^{-\frac{1}{2}},$$

where  $r_{ep}$  is the distance from the electron to the other charge center in the molecule, and

$$\Delta = (1 + \rho + \frac{1}{3}\rho^2) \exp(-\rho)$$

where  $\rho = Z' r_{pc} / a_e$ , is introduced to normalize  $\psi_e^I$ ,  $r_{pc}$  is the spacing between the nuclei of the molecule,  $a_e$  is the electronic Bohr radius, and Z' is the effective charge. The best value for Z' is about 1.19, and for  $r_{pc}/a_e$  about 1.40. This leads to  $\Delta = 0.677$ . This wave function gives reasonably good agreement with the binding energy of the hydrogen molecule and seems to provide the principal features of that system. The introduction of a Z'takes some account of the binding of the electron provided by the other proton in the molecule. We will choose the outgoing-electron wave function to be a Coulomb wave function. In this case, presumably Z'will lie somewhere between 1 and 1.19, approaching 1 as the energy is increased. (It would be expected that Z' approaches 1 as the DeBroglie wavelength becomes short compared to the distance between the protons. Thus, Z' approaches 1 as the kinetic energy becomes large compared to the binding energy.)

Now, after expanding  $1/r_{ei}$  in the usual power series in  $(r_e/r_i)^n$ , we obtain for the dipole contribution to  $\mathcal{K}_{int}$ :

$$(I|\Im C_{int}|F) = -e^{2} \left[ \int d^{3}\tau \Psi_{I}^{*} \Psi_{F} \right]$$

$$\times \sum_{i} \epsilon_{i} \int_{0}^{\infty} r_{e}^{2} dr_{e} d\Omega_{e} \psi_{e}^{I*} \frac{r_{i} \cos\theta_{ie}}{r_{e}^{2}} \psi_{e}^{F} \right]$$

$$-e^{2} \left[ \int d^{3}\tau \Psi_{I}^{*} \Psi_{F} \sum_{i} \epsilon_{i} \int_{0}^{r_{i}} r_{e}^{2} dr_{e} d\Omega_{e} \right]$$

$$\times \psi_{e}^{I*} (r_{e}/r_{i}^{2} - r_{i}/r_{e}^{2}) \cos\theta_{ie} \psi_{e}^{F}$$

where  $\psi_e$  is the electronic wave function for the initial, *I*, and final, *F*, states,  $\Psi$  is the corresponding wave function for the three-body system,  $\epsilon_i$  is +1 for the two protons and -1 for the  $\mu$  meson,  $\theta_{ie}$  is the angle between  $\mathbf{r}_e$  and  $\mathbf{r}_i$ , and  $d^3\tau$  represents the volume element of the three-body system. The second term gives a contribution of order  $kr_i$  when compared to the rate obtained when the final state is an *s* state (which will be treated later) and is therefore negligible. The integration over the part of the wave function centered on  $\mathbf{r}_e$ can be carried out exactly, as is shown in Appendix C. The other term has been obtained by an approximate

<sup>&</sup>lt;sup>15</sup> See, for example: L. Pauling and E. B. Wilson, *Introduction to Quantum Mechanics* (McGraw-Hill Book Company, New York, 1935), p. 349.

numerical integration as is also indicated there. Then we may write

$$(I|\mathcal{K}_{\rm int}|F) = -4\pi e^2 i$$

$$\times \left(\frac{(Z')^3}{2\pi a_e^3(1+\Delta)}\right)^{\frac{1}{2}} \int \Psi_I^* \Psi_F d^3\tau \sum_i \epsilon_i r_i \cos\theta_{ik} I(k_e),$$

where  $\theta_{ik}$  is the angle between  $\mathbf{r}_i$  and  $\mathbf{k}_e$  the momentum of the ejected electrons, and  $I(k_e)$  is the value of the integral over the electronic wave function, i.e.,

$$I(k_e) = \int r^{-2} \left[ e^{-\alpha r} + e^{-\alpha |\mathbf{r}-\mathbf{r}_p|} \right] \psi_e^f(\mathbf{r}) d^3 r.$$

Thus, to evaluate  $(I|\mathfrak{K}_{int}|F)$ , we need the matrix element of the dipole moment of the three-body system, **d**, with respect to the center of mass in the direction of electron ejection  $\mathbf{k}_{e}$ . In Appendix C, it is shown that

$$(I | d_k | F) = \left(\frac{4\pi}{3}\right)^{\frac{1}{2}} \int_0^\infty \chi_{-I^*}(r_n) \chi_{+F}(r_n) \frac{1}{2} (1 + \rho_{\mu}) \\ \times \int \psi_{+} \xi \eta \psi_{-} d\tau_{\mu} r_n dr_n,$$

where we have chosen the final bound three-body state to be quantized with angular momentum  $(\mathbf{L})_k=0$ . The final bound state of the system is a p state and is of the (+) type [the (-) type having an almost completely repulsive potential and consequently no bound states] so that the spins of the nuclei must be in a triplet configuration. The initial state is an *s* state of the (-) type, which thus makes the initial state also antisymmetric in the nuclear spatial variables and again the spin state must be triplet. As discussed in I, the incident wave function must then be chosen asymptotically as:

$$\Psi_{I} \sim e^{ikz} \psi_{1} - e^{-ikz} \psi_{2} + (f_{11} - f_{12}) (e^{ikr}/r) (\psi_{1} - \psi_{2}).$$

Thus, because we have  $(f_{11}-f_{12})=k^{-1}e^{i\delta}-\sin\delta_{-}$ , we find that asymptotically,

$$\psi_I \sim (kr)^{-1} e^{i\delta_-} \sin(kr + \delta_-) (\psi_1 - \psi_2)$$
  
 
$$\sim \sqrt{2} (kr^{-1}) e^{i\delta_-} \sin(kr + \delta_-) \cdot \psi_-.$$

The initial  $\chi$  wave function must therefore be normalized asymptotically to  $\sqrt{2} \sin(kr+\delta_{-})$ . Finally, from the matrix element for  $d_k$  we obtain the total rate of molecular-ion formation in pure hydrogen as

$$R_{(p\mu p)} = 12v_e N (a_{\mu}/a_e)^5 (Z')^3 |I(k_e)d_k|^2, \qquad (3)$$

where  $v_e$  is the velocity of the electron and N is the number density of protons in liquid hydrogen. Here we have integrated the isotropic distribution in the electron direction over all angles  $d\Omega_k$  and have taken into account the fact that the triplet state occurs only in three-fourths of the collisions.

In order to evaluate  $d_k$ , the integration over the meson variables was carried out analytically, and the final integration over  $r_n$  was done numerically for  $r_n \leq 20$ . The contribution to the matrix element for  $r_n > 20$  was calculated analytically by the use of the asymptotic form of the  $\chi$ 's obtained from their values and derivatives at  $r_n=20$ . For an incident kinetic energy near zero, we find  $d_k=25.0$ . From the binding energy of the  $(p\mu p)^+$  system in the p state, we find an energy release of 93 ev in the reaction. Because the electron is bound in the molecule with 15.6 ev, the electron will escape with the energy of 77 ev. This gives a value for  $k_e$  of 2.38 atomic units. The value obtained for  $I(k_e)$  has some dependence on the choice of Z', the effective charge for the outgoing electronic wave function. Using N $= 3.5 \times 10^{22}$  cm<sup>3</sup>, and Z'=1.19, we obtain

$$R_{(p\mu p)} = 6.5 \times 10^6 \text{ sec}^{-1}$$

while for Z' = 1.00, we obtain a slightly smaller value.

In addition to the dipole ejection, molecular ions might be formed by a monopole ejection of the electron. This mechanism has been discussed previously by Jackson.<sup>16</sup> The perturbation Hamiltonian is identical to that used previously, but in this case the ejected electron will be in an *s* state. Now, we expand  $r_{ei}^{-1}$  and keep only the lowest order; *viz.*,  $r_{ec}^{-1}$  for  $r_e \ge r_i$  and  $r_i^{-1}$  for  $r_e \ge r_i$ . Then we obtain

$$(I|\mathfrak{W}_{int}|F) = -e^2 \int d^3\tau \\ \times \Psi_I^* \Psi_F \sum_i \epsilon_i \int_0^{r_i} \left(\frac{1}{r_i} - \frac{1}{r_e}\right) \psi_e^{I^*} \psi_e^F d\tau_e,$$

by extending the integral involving  $r_e^{-1}$  over the entire region  $0 \leq r_e < \infty$ , and then subtracting this part in the integral from 0 to  $r_e$ . For  $r_e$  larger than all the  $r_i$ 's, it is evident that  $\Im C_{int} = 0$ . Now, because the molecular ion is small compared to the electronic Bohr radius, we may replace  $\psi_e^I$  and  $\psi_e^F$  by their values at r=0 to obtain  $-(2\pi/3)r_i^2\psi_e^I(0)\psi_e^F(0)$  for the integral. Using the previous choice for the electronic wave function for  $\psi_e^I$  and a free particle  $\psi_e^F$ , we then obtain

$$v_{\rm inc}\sigma = \frac{4}{9} \frac{e^2}{\hbar c} \left(\frac{a_{\mu}}{a_e}\right)^4 \left(\frac{p_e c}{\hbar}\right) a_{\mu^3} |M'|^2,$$

where we have defined

$$M' = \frac{1 + e^{-\alpha r_p}}{(1 + \Delta)^{\frac{1}{2}}} \int \left(\frac{r_1^2 + r_2^2 - r_\mu^2}{a_\mu^2}\right) \Psi_I^* \Psi_F d^3\tau.$$

 $\psi_I$  is normalized to the incident part of  $e^{ikz}$  as  $r \to \infty$ ,  $v_{\text{inc}}$  is the relative velocity of the proton and  $(p\mu)$ , and  $p_e$  is the momentum of the outgoing electron. Using a density of liquid hydrogen of  $3.5 \times 10^{22}$  cm<sup>3</sup>, we find a

<sup>&</sup>lt;sup>16</sup> As will be seen, we disagree with Jackson's result (reference 5) by a factor of order  $10^{-6}$ . The discrepancy can be either a result of the fact that he did not take into account the effects of the orthogonality of the unperturbed wave functions, or that the perturbing Hamiltonian did not include the term  $e^2/r_{ec}$ .

rate,  $R_{ej}$ , for the molecular-ion formation in pure hydrogen:

$$R_{ej}^{(S)} \approx 2.2 \times 10^{-2} |M'|^2 \text{ sec}^{-1}.$$

Thus, since one would anticipate that  $\langle (r/a_{\mu})^2 \rangle_{\text{AV}} \leq 10^2$ , the monopole ejection cannot compete with the dipole case.

Finally, we might consider the radiative formation of the molecule ion. The total transition probability for dipole radiation is given by

$$R_{\gamma} = \frac{4}{3} \frac{e^2}{\hbar c} \frac{\omega^3}{c^2} |\mathbf{d}|^2,$$

where  $\omega = E/\hbar$ , *E* is the energy of the photon, and **d** is the dipole moment of the system.<sup>17</sup> Inserting the value of  $\langle d_k \rangle$  as computed for the electron-ejection process, we find a rate of radiative molecular-ion formation :

$$R_{\gamma} = 5.7 \times 10^2 \text{ sec}^{-1}$$

in pure liquid hydrogen. Thus the radiative formation is also quite negligible.

B. 
$$(d\mathfrak{y}) + p^+ \rightarrow (d\mathfrak{y}p)^+$$

In the case of  $(d\mu) + p^+ \rightarrow (d\mu p)^+$ , the major mechanism is, as before, the interaction of the dipole moment of the three-body system with the electron to eject it. There are, of course, numerical differences between the  $(d\mu p)^+$  and  $(p\mu p)^+$  cases. The energy released is 90 ev rather than 93 ev. The matrix element is also different. In addition, because the nuclei in  $(d\mu p)^+$  are not identical, the incident wave function is normalized asumptotically to  $k^{-1}\sin(kr+\delta)$  rather than  $\sqrt{2}$  times this value, while the various spin states of the system are of no importance. With this normalization, we find for the matrix element  $\langle d_k \rangle = 29.4$ . Also, we find  $I(k_e)$  $= 0.575k_e^{-1}$ . Thus, from an equation similar to Eq. (3), we obtain for the rate of molecular-ion formation in pure hydrogen:

# $R_{(p\mu d)} = 2.5 \times 10^6 \text{ sec}^{-1}$ .

Once the  $(p\mu p)^+$  molecular ion is formed, the most likely possibility is that the  $\mu$  meson will decay with its natural half-life. Since the system has no dipole moment between two (+) states, it cannot readily decay to the ground state. On the other hand, the  $(d\mu p)^+$ molecular ion can readily decay from the p state in which it is formed to the ground s state, because it will have a dipole moment as a result of the asymmetry in the two nuclei. In this process, 124 ev is released. Just as in the molecular-ion formation, the dominant mode of decay is electron ejection by a dipole interaction. Because the initial and final states are both bound, the wave functions are normalized differently than for free particles. The expression for  $(I|d_k|F)$  is given in Appendix C. We find  $(I|d_k|F)=0.271a_{\mu}$ , and hence a transition rate of

$$R_{PS} = 2.5 \times 10^{10} \text{ sec}^{-1}$$
.

In this calculation, we have assumed that the transition takes place from a molecule with two electrons present. Because one electron has been ejected in the molecular-ion formation, it is possible that only one electron remains to be ejected in this decay process. It would seem that the exchange of an electron from another molecule in the liquid to the ion would take place quite rapidly. Whether or not this is the case, the computed rate is so large that the molecular ion will proceed in a negligible time to the ground state.

C. 
$$(d\mu) + d^+ \rightarrow (d\mu d)^+$$

Finally, we will briefly consider the formation of the  $(d\mu d)^+$  molecular ion. This case is similar to the  $(p\mu p)^+$  case, but there are two differences. In the first place, the deuterons satisfy Bose statistics, so in the final state their spins must be in an antisymmetric configuration. This gives a statistical factor of  $\frac{1}{3}$  rather than  $\frac{3}{4}$  in the rate of formation. Secondly, there are two bound s states, so that the free s-state wave function has an extra node compared to  $(p\mu p)^+$ . As a result, there is much cancellation in the evaluation of the matrix element of **d**, and it is very small. Thus, the rate of molecular-ion formation in this case in pure deuterium is very small compared to the corresponding  $(p\mu p)^+$  rate. Specifically, we find

$$R_{(d\mu d)} = 5.9 \times 10^4 \text{ sec}^{-1}$$

Because this rate is so small, we would expect that an experiment carried out in deuterium would lead to rather few (dd) nuclear reactions. In addition the likelihood for nuclear reactions is further decreased because the final states of the molecular ion will be predominantly p states for which the probability of finding the nuclei at small distances will be small. This latter effect will be compensated to some extent by a much higher intrinsic nuclear rate, since no electromagnetic interaction is involved. For molecular ions formed at higher energy, we would expect that some of them will form by a transition from incident p to final s states. These, of course, will react very quickly.

# D. Comparison with Experiments

The phenomenological analyses lead to a dependence of the yield of rejuvenated muons on deuterium concentration given by  $^{5,6}$ 

$$1/Y = A + B/c_d$$

where Y is the number of energetic muons per incident muon,  $c_d$  is the concentration of deuterium,  $R_0$  is the rate of natural decay, and we have

$$B = (A+1)(R_0 + R_{(p\mu p)})/R_{ex}.$$

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<sup>&</sup>lt;sup>17</sup> See, for instance, Walter Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1954), third edition.

Evidently, A gives the rate at saturation  $(c_d \rightarrow 1)$ . Among other things, the value of A will depend on the nuclear reaction rate. The experimental results are in reasonably good agreement with this dependence on  $c_d$ , and they lead to a value for (A+1)/B of about  $1.3 \times 10^3$ . Our calculations give

$$(A+1)/B = R_{\rm ex}/(R_0 + R_{(p\mu p)}) = 1.6 \times 10^3.$$

Considering the fact that the experiments do not give a very precise value for this ratio, as the error seems to be something like  $\pm 0.3 \times 10^3$  and the correction terms in the calculation of the three-body system are of order  $M_{\mu}/M_n \sim 0.2$ , this agreement seems quite satisfactory.

#### **IV. NUCLEAR REACTIONS**

#### A. Estimates of Matrix Elements

Once the  $(p\mu d)^+$  molecular ion is formed, there is a sizable probability that the two nuclei come sufficiently close together to undergo a nuclear reaction, with the formation of He<sup>3</sup>. Two possible processes have been observed:

and

(b) 
$$p+d+\mu \rightarrow \text{He}^3+\mu$$
.

(a)  $p+d \rightarrow \text{He}^3 + \gamma$ ,

The excess energy of 5.5 Mev is taken away in the former by the  $\gamma$  ray and in the latter by the  $\mu$  meson. As one sees on comparing the results of Ashmore *et al.*<sup>4</sup> with those of Alvarez *et al.*<sup>3</sup> process (a) is much more likely than process (b). We have carried out a somewhat phenomenological treatment of the nuclear reactions, in which we have assumed that the proton and neutron in the deuteron are distinct particles and maintain their respective identities throughout the interactions.

In the initial molecular-ion state, at small separations, the two nuclei are largely in an s state with respect to each other, corresponding to the (+) part of  $\Psi$ , with a small admixture of p state corresponding to the (-) part. Since, in this state the probability is small that the nuclei are close enough together for nuclear forces to be significant, the wave function will be only slightly perturbed by the nuclear effects. Because of the exclusion principle for the two protons in this system, we must treat the singlet and triplet proton spin states separately. In the incident singlet case, we can write symbolically for a state of  $J_z = +\frac{1}{2}$ ,

$$\Psi_{I}^{(s)} \sim S_{pp}^{0,0} \sigma_{n}^{+\frac{1}{2}} \{ L_{pp}(0) L_{pp,n}(0) + L_{pp}(2) L_{pp,n}(2) \\ + \cdots + \alpha [L_{pp}(0) L_{pp,n}(1) + \cdots ] \},$$

where  $S_{pp}^{s,s_z}$  represents the two-proton spin function of total spin s and z component of spin  $s_z$ , and  $\sigma_n^{+\frac{1}{2}}$  is the neutron spin function of z component  $+\frac{1}{2}$ . The  $L_{pp}(l)$  represent the orbital angular-momentum functions for the two protons about their center of mass, while  $L_{pp,n}(l)$  represents the orbital angular-momentum function for the neutron moving about the pair of protons as a unit. The terms multiplied by  $\alpha$  represent the small admixture of (-) functions in  $\Psi_{I}^{(s)}$ . The total orbital angular momentum of the initial system is zero, and the terms such as  $L_{pp}(2)L_{pp,n}(2)$  must be combined to give a total L of zero. Thus the total J for the singlet state is one-half. For the terms multiplied by  $\alpha$ , the  $\mu$  meson (which has not been represented here) must be considered in treating the total angular momentum. In the (-) case, for small values of  $r_n$ , the meson is essentially in a p state, so that the nuclear functions must be combined to form a total L of one. The functions  $L_{pp, n}(l)$  are symmetric in the two protons, and so any l may appear in them; because of the exclusion principle, however, only the even terms in  $L_{pp}(l)$  may occur in  $\Psi_{I}(s)$ . The final nuclear state is that of  $He^3$ , with a total J of one-half. For it, the two protons are in a singlet state. Thus, the final state,  $\Psi_F$ , will be of a form similar to  $\Psi_{I}^{(s)}$ , except that the  $\alpha$ terms will not be present. Now let us consider the electromagnetic transitions for such states.

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For the  $\frac{1}{2} \rightarrow \frac{1}{2}$  transitions, either electric dipole or magnetic dipole reactions are possible. The perturbation Hamiltonian has the matrix elements<sup>18</sup>

$$M_{1,0}' = (e\hbar/2Mc)(3/4\pi)^{\frac{1}{2}} \sum_{i} \mu_{i} \int \psi_{I}^{*}(\sigma_{i})_{z} \psi_{F} d\tau_{i}$$

 $Q_{1,0} = e(3/4\pi)^{\frac{1}{2}} \sum_{i} \int \psi_{I}^{*} z_{i} \psi_{F} d\tau,$ 

respectively, where  $z_i$  is the z component of the displacement of the *i*th particle from the center of mass of the system,  $\mu_i$  is its magnetic moment in nuclear magnetons, and  $\sigma_i$  is its Pauli spin matrix. The part of  $\psi_I$  independent of  $\alpha$  will have only a nonvanishing  $M_{1,0}'$ , while the part proportional to  $\alpha$  will only contribute to  $Q_{1,0}$ .

We might roughly estimate these two rates by considering that the nuclear states are simply of constant density to some distance, R, and then zero outside. From the normalized eigenstate of the molecular ion, we find that the (-) solution is given by

# $\Psi_{-}\approx 0.0317 (r/a_{\mu})$

for small *r*. This function increases linearly with *r*, as is appropriate to a *p* state. Putting this wave function into the expression for  $Q_{1,0}$ , we obtain as a very rough estimate,  $Q_{1,0} \sim (0.0317/5) (r/a_{\mu})^{\frac{1}{2}} eR$ . If we set R=4 $\times 10^{-13}$  cm, we find for  $R_{Ed}$ , the rate of electric-dipole transitions,

$$R_{E_d} \sim 7 \times 10^4 \text{ sec}^{-1}$$
.

If we make a similar rough estimate of  $M_{1,0}$ , we find first

$$\Psi_{+} \approx 0.0146.$$

<sup>&</sup>lt;sup>18</sup> See, for instance, J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, New York, 1952), p. 599.

Again using  $R=4\times 10^{-13}$  cm, we have  $M_{1,0}/Q_{1,0} \sim 12$ , so that this rate becomes:

$$R_{M_d} \sim 10^7$$
 sec.

Thus, it would seem that for the singlet initial states magnetic-dipole transitions are the most likely. In addition, one might consider the element  $Q_{1,0}'$ , but because it involves both the magnetic moment and the  $\alpha$ terms, its contribution will be extremely small.

Let us now consider the initial triplet states. Again we write symbolically

$$\Psi_{I}^{(t)} \sim S_{pp}^{1,0} \sigma_{n}^{12} \{ L_{pp}(1) L_{pp,n}(1) + \cdots + \alpha [L_{pp}(1) L_{pp,n}(0) + \cdots ] \}.$$

The orbital angular momentum will again be zero, but now J is either  $\frac{1}{2}$  or  $\frac{3}{2}$  depending on the spin orientations. In this case, in order to produce a transition to the He<sup>3</sup> ground state, it is necessary to flip a spin, thus changing the protons from a triplet to a singlet configuration, and in addition to change the orbital wave function from an odd  $L_{pp}$  to an even one. Now, various transitions are possible, for example,

and  

$$Q_{2,0}' = -\frac{i\kappa c\hbar}{4Mc} \left(\frac{5}{16\pi}\right)^{\frac{1}{2}} \sum_{i} \mu_{i} \int (3z_{i}^{2} - r_{i}^{2}) \nabla \cdot \{\Psi_{I}^{*}(\mathbf{r}_{i} + \boldsymbol{\sigma}_{i})\Psi_{F}\} d\tau,$$

 $Q_{1,0}' = \frac{i\kappa\epsilon\hbar}{4M_c} \left(\frac{3}{4\pi}\right)^{\frac{1}{2}} \sum_i \mu_i \int \Psi_I^*(\mathbf{r}_i \times \boldsymbol{\sigma}_i)_z \Psi_F d\tau,$ 

where  $\kappa = E/\hbar c$ . The matrix element of  $Q_{1,0}$  in this triplet case may be compared to that of  $Q_{1,0}$  in the singlet case. The former is smaller by a factor  $E_{\gamma}/4Mc^2$  $\sim 1/200$ , as are all of the triplet transitions, because the spin must be flipped. However, it does not involve the small components of the molecular-ion wave function, and one might therefore estimate  $Q_{1,0}'/Q_{1,0} \sim \frac{1}{6}$ . Thus, because the electric-dipole rate from singlet states is small, the rate of nuclear reaction in this process will be negligible compared to the natural decay rate of the  $\mu$  meson. Similarly, we have roughly estimated the  $Q_{2,0}$ element. In this case, the element involves only the small components and so is quite negligible. Finally, we have estimated  $M_{2,0}$ '. Again we find that the matrix element is smaller than the singlet  $Q_{1,0}$  (by a factor  $\sim \frac{1}{2}$ ), and the transition rate is also negligible compared to the natural decay, having a value of  $\sim 10^3$  sec<sup>-1</sup>. Thus, from these considerations, we would conclude that essentially only the singlet initial states will undergo nuclear reactions to produce He<sup>3</sup>, with  $\gamma$ -ray emission.19

In addition to the  $\gamma$  emission, it has also been suggested that a monopole transition may occur, with ejection of the  $\mu$  meson.<sup>5,6</sup> This process is similar to that for the monopole ejection of an electron in the molecular-ion formation discussed in Sec. III A. For this process, we find a rate of ejection,  $R_{ej}$ ,

$$R_{ej} = \frac{32}{9} \left( \frac{e^4 m_{\mu} \dot{p}_{\mu}}{\hbar^4 a_{\mu}^3} \right) \left| \int (r_1^2 + r_2^2) \Psi_I^* \Psi_F d\tau \right|^2,$$

where  $r_1$  and  $r_2$  are the positions of the two protons. In this case, transitions from the triplet initial state are forbidden. On making the same rough estimate as previously, we find

$$R_{ej} \sim 5 \times 10^5 \text{ sec}^{-1}$$
.

## **B. Magnetic-Moment Effects**

We will now consider the nuclear reaction rates from the  $(p\mu d)^+$  molecular ion, under the assumption that transitions from the triplet state are negligible, as is indicated by the above considerations. Because of the magnetic-moment interactions between the three particles in the molecular ion, some of the spin degeneracy in the eigenvalues will be removed, with the formation of definite spin eigenstates. As the energies involved are very small ( $\sim 0.1$  to 1.0 ev) compared to the Coulomb binding energies of the system, the orbital functions will not be affected appreciably by these interactions. However, the energy splitting is large enough that after a molecular ion is formed and until a nuclear reaction occurs, one may consider that the system is in a definite spin eigenstate, because interference effects between states will only persist for times of the order of  $10^{-14}$  sec. Thus the J of the nuclear system alone will not be a good quantum number. In computing the nuclear decay rate from a particular eigenstate, we will assume that the rate is proportional to the probability of finding the system in the singlet state, and we will introduce a phenomenological constant,  $R_n$ , which will give the nuclear rate if the system is a pure singlet configuration. Thus, we set

$$R_t = P_t^{(s)} R_n,$$

where  $R_t$  is the nuclear rate from the *t*th eigenstate, and  $P_t^{(s)}$  is the probability of a singlet configuration in that *t* state.

The interaction Hamiltonian for the spins is

$$\mathcal{K}_{\rm spin} = \boldsymbol{\mathfrak{y}}_p \cdot \boldsymbol{\mathfrak{y}}_d \langle 1/r_{pd^3} \rangle + \boldsymbol{\mathfrak{y}}_p \cdot \boldsymbol{\mathfrak{y}}_\mu \langle 1/r_{p\mu^3} \rangle + \boldsymbol{\mathfrak{y}}_d \cdot \boldsymbol{\mathfrak{y}}_\mu \langle 1/r_{d\mu^3} \rangle,$$

where  $\mathbf{y}_m$  is the magnetic moment of the *m*th particle, and the expectation values of  $r^{-3}$  are taken with the molecular-ion wave function. Here  $\mu_{\mu}$  is much larger than either  $\mu_p$  or  $\mu_d$ , and  $\langle r_{pd}^{-3} \rangle$  is much smaller than  $\langle r_{p\mu}^{-3} \rangle$  and  $\langle r_{d\mu}^{-3} \rangle$ , so that in the following we will neglect the term in  $\boldsymbol{y}_p \cdot \boldsymbol{y}_d$ . In addition, we will approximate  $\langle r_{p\mu}^{-3} \rangle \approx \langle r_{d\mu}^{-3} \rangle$ , which is valid because the (-) part of the eigenstate is always small. We may separate

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<sup>&</sup>lt;sup>19</sup> Jackson (reference 5) has attempted to correlate the (pd) nuclear reaction rate treated here with that of the "mirror" (nd) rate at low energies. However, because the process involves the magnetic moments, it is not a true mirror process, and a correction would be necessary. We feel another significant correction to the calculation is the effect of the nuclear forces in mixing in the "unallowed" Coulomb wave functions. This leads to a large correction in the calculated rate for the (pd) system.

the various spin states according to their z components of **J**, as this is a good quantum number in the presence of the interaction. Let us now designate by  $X_t(s)$ , the spin state of the *t*th particle, corresponding to a z component of spin equal to *s*. Then the unperturbed states,  $\Sigma_s$ , may be chosen as:

$$\begin{split} J_z &= +2: \quad \Sigma_2 = \chi_\mu(\frac{1}{2})\chi_p(\frac{1}{2})\chi_d(1), \\ J_z &= +1: \quad \Sigma_1{}^A = \chi_\mu(\frac{1}{2})\chi_p(\frac{1}{2})\chi_d(0), \\ & \Sigma_1{}^B = \chi_\mu(-\frac{1}{2})\chi_p(\frac{1}{2})\chi_d(1), \\ J_z &= 0: \quad \Sigma_0{}^A = \chi_\mu(-\frac{1}{2})\chi_p(-\frac{1}{2})\chi_d(1), \\ & \Sigma_0{}^B = \chi_\mu(-\frac{1}{2})\chi_p(\frac{1}{2})\chi_d(0), \\ & \Sigma_0{}^C = \chi_\mu(\frac{1}{2})\chi_p(-\frac{1}{2})\chi_d(0), \\ & \Sigma_0{}^D = \chi_\mu(\frac{1}{2})\chi_p(\frac{1}{2})\chi_d(-1), \end{split}$$

and similarly for  $J_z < 0$ . The states  $J_z = \pm 2$  are evidently good eigenstates, but since they are pure triplets, we have  $P_t^{(s)} = 0$ . In order to determine the eigenstates for  $J_z = \pm 1$ , it is necessary to solve a cubic equation, while for  $J_z = 0$  it would seem that one has a quartic equation to solve. However, in this case the matrix may readily be split into two second-rank matrices, and one must solve two quadratic equations. The eigenvalue,  $\lambda_t$ , and coefficients  $\alpha_t^{(i)}$ , for each  $\Sigma_s^{(i)}$  are given in Table II. The eigenvalues are given in units of  $(e\hbar/m_{\mu}c)(e\hbar/Mc)\langle r_{p\mu}^{-3}\rangle$ . The eigenstate is given as

$$\Sigma_{J_z}^{t} = \sum_{t} (\alpha_t^{(i)} \Sigma_{J_z}^{(i)}).$$

For  $J_z = +1$ , it is clear that  $\Sigma_1^B$  is pure triplet (since all the nucleons are aligned in the same direction). One readily finds that the contribution from other states to the singlet probability,  $P_t^{(s)}$ , is

$$P_t^{(s)}(1) = \frac{1}{2} \left[ \alpha_t^C - (1/\sqrt{2}) \alpha_t^A \right]^2.$$

In addition, for  $J_z = 0$ , one finds

$$P_t^{(s)}(0) = [\alpha_t^A - (1/\sqrt{2})\alpha_t^B]^2.$$

These values are also tabulated in Table II.

# C. Analysis of the Time Dependence of the $\gamma$ -Ray Experiment

Our analysis leads to a somewhat different analysis of the time dependence for the  $\gamma$ -ray production that was observed by Ashmore *et al.* than the previous treatment.<sup>4</sup> Instead of having a single rate of nuclear

TABLE II. Eigenvalues and coefficients for  $\Sigma_s^{i}$ .

$J_z$	$\lambda_t$	$\alpha \iota^A$	$\alpha t^B$	$\alpha t^C$	$\alpha \iota^D$	$P_t^{(s)}$
$\pm 1$	0.978	0.143	0.771	0.621		0.135
$\pm 1$	-0.407	0.733	0.504	0.456		0.002
$\pm 1$	-0.258	-0.665	0.390	0.638		0.613
0	-0.416	0.435	0.557	0.557	0.435	0.002
0	-0.144	0.557	-0.435	-0.435	0.557	0.748
0	0.990	0.141	0.693	-0.693	-0.141	0.122
0	-0.298	0.693	-0.141	0.141	-0.693	0.629

FIG. 5. Time distribution of  $\gamma$  rays from the (pd) nuclear fusion. The experimental points are those of Ashmore *et al.*<sup>4</sup> Theoretical curves for three values of the parameter  $R_n$  are given.



reaction to produce  $\gamma$  rays, we must now consider 7 different rates  $(\pm J_z \text{ give equal rates, and } J_z = \pm 2 \text{ gives}$ a zero rate). We have made a new analysis of their time distribution in which we have used the calculated value of the  $(p\mu d)^+$  molecular-ion formation rate and have then fitted  $R_n$  and the background rate to the experimental data. Our expression for the number of  $\gamma$ rays is similar to Eq. (2) of Ashmore *et al.*,

$$\frac{dn^{\gamma}}{dt} = \frac{1}{12} \sum_{m=1}^{12} \left[ \lambda_1^{(t)} \lambda_m / (\lambda_y^{(t)} - \lambda_x^{(t)}) \right] \\ \times \left[ \exp(-\lambda_x^{(t)} t) - \exp(-\lambda_y^{(t)} t) \right]$$

where the notation is the same as in their paper with an added index t to distinguish the different eigenstates. The factor  $\frac{1}{12}$  is simply the statistical weight of each of the spin eigenstates, t. The total number of  $\gamma$  rays emitted per incident  $\mu$  meson is

$$n_T^{\gamma} = \frac{1}{12} \sum_{m=1}^{12} \lambda_1^{(t)} \lambda_m / (\lambda_b \lambda_c^{(t)} - \lambda_2^{(t)} \lambda_m).$$

The ratio of rejuvenated mesons to  $\gamma$  rays which is needed for the recycling of the process was fitted by an iterative procedure by the use of both the experimental data on  $\gamma$  rays and on rejuvenated muons. Because the internal conversion ratio was rather small, the method was rapidly convergent. Representative curves that were obtained are given in Fig. 5. From these curves we believe that the best fit to  $R_n$  gives

$$R_n = (1.25 \pm 0.10) \times 10^6$$
 sec

From this value, we then can calculate the number of  $\gamma$  rays per incident muon as

$$n_T^{\gamma} = 0.20 \gamma/\mu$$
.

This is considerably lower than that of Ashmore *et al.*  $(0.34\gamma/\mu)$  but is in fact in good agreement with an earlier value obtained by them by a simple integration

of their data, together with an assumption that the background was simply given by the average of one point obtained at  $\tau < 0$  and two points at the largest values of  $\tau$ .<sup>20</sup> It would appear from the curves that the analysis presented here represents the data reasonably well.

# **V. CONCLUSION**

As can be seen from the preceding analysis, the original qualitative considerations concerning the enhancement of nuclear reactions through the formation of  $\mu$ -mesonic molecular ions have been well verified by more quantitative calculations. The saturation phenomenon on increasing deuterium concentration seems to agree reasonably well with the experiments. The phenomenon of the gaps also seems to be well understood. In this case one might argue that the calculations are not sufficiently accurate to provide the precise energy at which the Ramsauer effect takes place, but that such an effect occurs for a small energy seems very likely.

Finally, the nuclear reaction rates as obtained on a phenomenological basis are in qualitative agreement with rough estimates made for them. Here, however, our analysis leads to a ratio of 0.20 for  $\gamma$  rays per stopped muon, which is smaller than the ratio obtained previously by Ashmore *et al.* If this number is used in conjunction with the experiments, we then obtain for the internal conversion coefficient,

# $\alpha \approx 12\%$

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# APPENDIX A

In this Appendix we will develop the test problem which was used to check the accuracy of the coding of the solution of the numerical integration. Because the equal- and unequal-mass cases were programmed separately, it was possible to use a test problem to convert the equal-mass equations

and

$$\frac{d^{2}\phi_{+}}{dr^{2}} + \frac{1}{\epsilon}(W - V_{+})\phi_{+} = 0$$
$$\frac{d^{2}\phi_{-}}{dr^{2}} + \frac{1}{\epsilon}(W - V_{-})\phi_{-} = 0$$

into the form of the equations in the unequal mass case,

 $^{20}$  A. Ashmore, University of Liverpool, England (private communication).

Eqs. (2) of I. This can be done by making a unitary transformation on the two-component  $\phi$ . Thus we set

$$\binom{\phi_+}{\phi_-} = \binom{(1-\rho^2)^{\frac{1}{2}}}{-\rho} \binom{\rho}{(1-\rho^2)^{\frac{1}{2}}} \binom{\phi_1}{\phi_2},$$

where  $\rho$  is to be chosen as a function of r. If this transformation is introduced, we obtain the equation for  $\phi_1$ ,

$$\begin{aligned} \frac{d^2\phi_1}{dr^2} + \frac{1}{\epsilon} \left[ W - V_+ + \rho^2 (V_+ - V_-) - \epsilon \frac{(d\rho/dr)^2}{1 - \rho^2} \right] \phi_1 \\ + \frac{2d\rho/dr}{1 - \rho^2} \frac{d\phi_2}{dr} + \left[ \frac{(d^2\rho/dr^2) + (1 - \rho^2)^{-1}\rho(d\rho/dr)^2}{(1 - \rho^2)^{\frac{1}{2}}} - \frac{1}{\epsilon}\rho(1 - \rho^2)^{\frac{1}{2}}(V_+ - V_-) \right] \phi_2 = 0 \end{aligned}$$

and a similar equation for  $\phi_2$ . These equations are of the proper form if we make the interpretations:

$$(V_{+})_{\text{test}} = \rho^{2} V_{+} + (1 - \rho^{2}) V_{-} + \epsilon \frac{(d\rho/dr)^{2}}{1 - \rho^{2}},$$
  

$$(V_{-})_{\text{test}} = \rho^{2} V_{-} + (1 - \rho^{2}) V_{+} + \epsilon \frac{(d\rho/dr)^{2}}{1 - \rho^{2}},$$
  

$$(f)_{\text{test}} = \frac{(d\rho/dr)}{1 - \rho^{2}},$$
  

$$(g)_{\text{test}} = -\frac{1}{\epsilon} \rho (1 - \rho^{2})^{\frac{1}{2}} (V_{+} - V_{-}).$$

Further, if we have  $\rho \to 1 + \vartheta(r^2)$  as  $r \to 0$ , then  $(f)_{\text{test}}$  approaches h = const and  $(g)_{\text{test}}$  approaches -2h/r. These are of course the boundary conditions found for the true f and g, and one would like to have them satisfied in a test problem as well. The true  $V_+$  and  $V_-$  become equal as r goes to  $\infty$ , and this can be accomplished by requiring that  $\rho$  approaches  $1/\sqrt{2}$  in this limit. Finally, the true g approaches a nonzero constant as r goes to  $\infty$ , and to satisfy this requirement, the difference between  $V_+$  and  $V_-$  must approach a nonzero constant in this limit. With these restrictions, the test problem will duplicate all of the features of the unequal-mass equations.

To produce all the test functions, we have used the equal-mass  $V_+$  and  $V_-$ , with the latter displaced by a constant so that  $(g)_{\text{test}}$  does not go to 0 as r approaches  $\infty$ , and we have chosen for  $\rho$ 

$$\rho = \frac{1}{\sqrt{2}} + \left(\frac{\sqrt{2} - 1}{\sqrt{2}}\right) (\cosh r)^{-1}.$$

The solutions of the test problem then have been compared to those of the uncoupled case by making a unitary transformation, and agreement was found to within the rounding errors involved. It might be noted that because the test problem is just a different form of two uncoupled equations, no exchange scattering should occur. This was also found to be true within the errors of the numerical calculations.

#### APPENDIX B

Because the energies of interest in this system may be very small (thermal energy  $\sim 1/400$  ev), one might question whether the electrons in the molecule play a significant role in such processes as the scattering of  $(d\mu)$  atoms from protons, because the latter will generally have electrons around them. The direct interaction between the two systems seems to be the largest perturbation where we have

$$\Delta V = e^2 \int |\psi_{\mu}|^2 |\psi_{e}|^2 \left[\frac{1}{r_{\mu e}} - \frac{1}{r_{d e}}\right] d^3 r_{e} d^3 r_{\mu}.$$

In this expression,  $\psi_{\mu}$  is the wave function of the  $\mu$  meson in the  $(d\mu)$  atom, and  $\psi_e$  is the electronic wave function in the (pe) atom. The separation of the nuclei,  $r_n$ , will be considered as a fixed parameter. These are the only interactions that have not been included previously. On inserting hydrogenic wave functions for the  $\psi$ 's which will be valid for the  $\mu$  for  $r_n \gg a_{\mu}$  (the only region for which this perturbation could be significant), we find:

$$\Delta V = \frac{2e^2}{a_e} \left(\frac{a_\mu}{a_e}\right)^2 e^{-r_n/a_e}$$

where  $a_e$  is the electronic Bohr radius. Because we have  $a_{\mu}/a_e \sim 1/200$ ,  $\Delta V$  has a maximum value of  $\sim 1/800$  ev, but since it has a long range, we will consider its effect a little further.

For  $r_n \gg a_{\mu}$ ,  $\Delta V$  will be the only potential present. If this potential is put into the Schrödinger equation, we find as a solution

$$\psi = A J_{i\eta}(i\xi e^{-r/a_{\theta}}) + B J_{-i\eta}(i\xi e^{-r/a_{\theta}}),$$

where  $\eta = (2MEa_0/m_ee^2)^{\frac{1}{2}}$ , and  $\xi = (4Mm_e/m^2)^{\frac{1}{2}}$ . Here M is the nuclear reduced mass,  $m_e$  is the electronic mass, and m is the mesonic mass. Given a particular phase shift resulting from the potential at small  $r_n$ , by appropriate matching of  $\psi$  and  $\psi'$  one can determine the effect on the phase shift of this potential. However, we are here interested in determining the limits of the energy, E, for which  $\Delta V$  can be neglected, and for this purpose we can expand  $J_{\pm i\eta}$  in a series in its argument, keeping only the first two terms.

We introduce

$$\left(\frac{\psi}{a_{e}\psi'}\right)_{r_{1}}=\frac{1}{\eta}\tan\left(\frac{\eta r_{1}}{a_{e}}+\delta_{0}\right),$$

where  $r_1$  is the point at which the wave function is to be matched, and  $\delta_0$  is the phase shift without  $\Delta V$ . Then, upon expansion of  $J_{\pm i\eta}$ , we find

$$\tan\left(\frac{\eta r_1}{a_e} + \delta_1\right) = \tan\left(\frac{\eta r_1}{a_e} + \delta_0\right) - \frac{\xi^2 \eta}{4(1+\eta^2)} e^{-2\tau_1/a_e},$$

where  $\delta_1$  is the phase shift including the effect of the potential  $\Delta V$ . For  $r_1 \sim 20a_{\mu}$ , at low energy, we find that the electronic effects will be negligible if we have  $E \gtrsim 1/200$  ev.

#### APPENDIX C

In computing the rate of molecular-ion formation in Sec. III A, it is necessary to carry out an integral, over the electronic coordinates, of the form :

$$J = \int d^3 \mathbf{r}_e \, \mathbf{r}_e^{-2} [\exp(-Z' \mathbf{r}_{ec}/a_e) + \exp(-Z' |\mathbf{r}_e - \mathbf{r}_p|/a_e)] \mathbf{r}_i \cos\theta_{ie} \psi_e^F.$$

In this integral, we choose for  $\psi_e^F$  the *p*-state Coulomb solutions,  $u_c(kr_e)$ , about  $\mathbf{r}_e$  which tend asymptotically to approximate plane waves. Then we have

$$\psi_{e}F=3i\cos\theta_{ek}u_{c}(kr_{e}),$$

where  $\theta_{ek}$  is the angle between  $\mathbf{r}_e$  and  $\mathbf{k}$ , the direction of ejection of the electron. The integration of the term containing  $\exp(-Z'r_{ec}/a_e)$  can be carried out exactly. Because the other term decreases exponentially with the distance from  $\mathbf{r}_p$  rather than from  $\mathbf{r}_c$ , we expect that it will be small compared to the contribution of the former. This expectation is borne out by the calculations. Thus, we will only keep those terms that interfere with the first term and will neglect the contribution of the quadratically dependent (noninterfering) ones.<sup>21</sup> This approximation leads to a retention of only the part of  $\exp(-Z'r_{ep}/a_e)$  which is spherically symmetric about  $\mathbf{r}_c$ .

Let us consider first the integration of this second term. We must evaluate

$$J_2 \equiv 3i \int_0^\infty d\mathbf{r}_e \int d\Omega_e \exp(-Z' |\mathbf{r}_e - \mathbf{r}_p| / a_e) \\ \times \cos\theta_{ie} \cos\theta_{ek} u_c(kr_e),$$

where  $\theta_{ie}$  is the angle between  $\mathbf{r}_e$  and  $\mathbf{r}_i$ , and  $\theta_{ek}$  is the angle between  $\mathbf{r}_e$  and  $\mathbf{k}$ , the direction of electron ejection. To carry out this integral, we expand

$$\exp(-Z'|\mathbf{r}_e-\mathbf{r}_p|/a_e) = \frac{1}{\sqrt{2}}f_s(r_e) + \frac{\sqrt{6}}{2}\mu f_p(r_e) + \cdots,$$

<sup>&</sup>lt;sup>21</sup> In the final determination of the rate of molecular-ion formation, it is necessary to average over all orientations of  $\mathbf{r}_p$ . Thus, by interfering terms we mean here those cross terms between the first and second parts which are obtained on squaring the matrix element and which are nonzero after such an averaging on  $\mathbf{r}_p$ .

where  $\mu$  is the angle between  $\mathbf{r}_e$  and  $\mathbf{r}_p$ , and we have

$$f_s(\mathbf{r}_e) = \frac{1}{\sqrt{2}} \int_{-1}^{1} \exp(-Z' |\mathbf{r}_e - \mathbf{r}_p| / a_e) d\mu,$$
  
$$f_p(\mathbf{r}_e) = \frac{\sqrt{6}}{2} \int_{-1}^{1} \mu \exp(-Z' |\mathbf{r}_e - \mathbf{r}_p| / a_e) d\mu,$$

and so on.

On integrating over  $d\Omega_e$ , we find that the "s" part gives

$$J_{2}(s) = \frac{2\pi}{3} \cos\theta_{ki} \int_{0}^{\infty} dr_{e} u_{c}(kr_{e}) \\ \times \int_{-1}^{1} \exp(-Z' |\mathbf{r}_{e} - \mathbf{r}_{p}| / a_{e}) d\mu.$$

The "p" part vanishes, and the "d" part gives

$$J_2(d) = \frac{8\pi}{15} \{ \left[ \cos^2 \theta_{ip} - \frac{1}{3} \right] \cos \theta_{ik}$$

 $+\sin\theta_{ip}\cos\theta_{ik}\sin\theta_{ik}\cos\phi_{pik}$ 

$$\times \int_{0}^{\infty} dr_{e} u_{c}(kr_{e}) \int_{-1}^{1} (\mu^{2} - \frac{1}{3}) \\ \times \exp(-Z' |\mathbf{r}_{e} - \mathbf{r}_{p}|/a_{e}) d\mu,$$

where  $\theta_{ip}$  is the angle between  $\mathbf{r}_p$  and  $\mathbf{r}_i$ , and  $\phi_{pik}$  is the azimuthal angle between the  $(\mathbf{p}, \mathbf{i})$  and  $(\mathbf{i}, \mathbf{k})$  planes. The term  $J_2(d)$  will not interfere with the spherically symmetric term,  $J_2(s)$ , however, because on integrating over the direction of  $\mathbf{k}$ , the  $\cos\phi_{pdk}$  term will vanish, while on averaging on the direction of  $\mathbf{p}$ , the first term will vanish. All other terms also vanish, and so we obtain  $J_2=J_2(s)$  in this approximation. The integral over  $\mu$  can be carried out analytically, and we have performed the final integration numerically. In the latter integration, we have replaced  $u_c(kr_e)$  by  $j_1(kr_e)$ .<sup>22</sup> Inserting the appropriate values for k,  $r_p$ , and Z'(2.50, 1.40, and 1.19, respectively), we find

 $J_2 = 0.948i \cos \theta_{ik}$ .

Let us now proceed to the exact integration of the first term.

Here the angular integration is simple, and using  $\cos\theta_{ie}$ =  $\cos\theta_{ik} \cos\theta_{ek} + \sin\theta_{ik} \sin\theta_{ek} \cos\phi$ , we obtain  $(4\pi i) \cos\theta_{ik}$ for the angular integration. To carry out the radial integration, we need the radial wave function. This function is given in terms of the Whittaker function,<sup>23</sup>  $W_{km}$ , where  $k=i\alpha Z/k_e$ , and  $m=l+\frac{1}{2}$ . Here we have  $\alpha=e^2/\hbar c$ . A very useful form of the radial function for arbitrary l is given by the integral representation

$$R_{l}(r_{c}) = \frac{1}{2} \left(\frac{kr_{e}}{2}\right)^{l} \frac{\exp\left[(\pi/2)\eta\right]}{|\Gamma(l+1+i\eta)|} \int_{-1}^{1} du(1-u)^{l-i\eta} \times (1+u)^{l+i\eta} e^{ikr_{e}u}$$

where we have  $\eta = \alpha Z' \cdot (c/v_e) = Z'/(a_ek_e)$ , c is the velocity of light, and  $v_e$  is the velocity of the electron at infinity. This wave function has the desired asymptotic behavior for  $r_e \to \infty$ :

$$R_l(r_e) \sim (k_e r_e)^{-1} \cos \left[ k_e r_e + \eta \ln 2k_e r_e - \frac{\pi}{2} (l+1) - \theta_{\Gamma} \right],$$

where  $\theta_{\Gamma}$  is the argument of  $\Gamma(l+1+i\eta)$ . Using this representation, we may carry out the radial integration. We define

 $J_1 \equiv 4\pi i \cos\theta_{ik} I_1(k_e),$ 

$$I_1(k_e) \equiv \int_0^\infty dr_e \exp(-Z'r_e/a_e) R_1(r_e)$$

Interchanging the u and  $r_e$  integrations, we can integrate over the variable  $r_e$  to obtain

$$I_{1} = -\frac{\exp[(\pi/2)\eta]}{4k_{e}|\Gamma(2+i\eta)|} \int_{-1}^{1} du \frac{(1-u)^{1-i\eta}(1+u)^{1+i\eta}}{(u+i\xi)^{2}},$$

where we define

$$\xi = Z'/(a_e k_e).$$

Then we have

$$I_{1}(k_{e}) = -\frac{1}{4k_{e}(1+\eta^{2})^{\frac{1}{2}}} \left(\frac{\exp(2\pi\eta)-1}{2\pi\eta}\right)^{\frac{1}{2}} \\ \times \int_{-1}^{1} \frac{(1-u)^{2}}{(u+i\xi)^{2}} \exp\{i\eta \ln[(1+u)/(1-u)]\} du.$$

This integral can be evaluated by the methods of contour integration. One introduces a cut in the u plane between -1 and +1, and then the integral can be replaced by a contour integral on a path C taken counterclockwise around the cut. One finds

$$\begin{split} \int_{-1}^{1} \frac{(1-u)^2}{(u+i\xi)^2} \exp\{i\eta \ln[(1+u)/(1-u)]\} du \\ = [1-\exp(2\pi\eta)]^{-1} \int_{c} \frac{(1-u)^2}{(u+i\xi)^2} \exp\{i\eta \ln\left[\frac{1+u}{1-u}\right]\} du, \end{split}$$

<sup>23</sup> See, for instance, E. T. Whittaker and G. N. Watson, *Modern Analysis* (Cambridge University Press, London, 1927), fourth edition, Chap. XVI.

<sup>&</sup>lt;sup>22</sup> One can expand the function  $\exp(-Z'|\mathbf{r}_e - \mathbf{r}_p|/a_e)$  in the complete set of hydrogen-like wave functions about  $r_e$ . Then the lowest term is of the form  $e^{-\alpha r}$ , and hence can also be calculated exactly with the Coulomb function,  $u_e$ . If this term is subtracted off, the integration of the remainder with  $j_1(kr)$  replacing  $u_e$  is small, and so the effect of the Coulomb correction on this term is expected to be very small. Thus, for simplicity, we have integrated this remaining part using  $j_1(kr)$  rather than  $u_e$ . The result quoted for  $J_2$  is that obtained before subtracting the lowest term and calculating this portion of the integral exactly with  $u_e$ .

where  $\ln[(1+u)/(1-u)]$  is defined to be real on the lower side of the cut. On expanding C to a circle of infinite radius, one obtains two contributions to the integral—one from the pole at  $u=-i\xi$  and one from the circle. Finally, one obtains

$$I_1(k_e) = \frac{\{(\eta + \xi) \exp(2\eta \tan^{-1}\xi) + (\eta - \xi) \exp(\pi\eta)\}}{k_e \{(1 + \eta^2)(2\eta/\pi) [\exp(2\pi\eta) - 1]\}^{\frac{1}{2}}}$$

For the total integral J, we then find

$$J = \left[4\pi i \cos\theta_{ik} I_1(k_e) + J_2(k_e)\right] r_i.$$

Thus in terms of the  $I(k_e)$  defined in Sec. II A, we find

$$I(k_e) = I_1(k_e) + (4\pi i \cos \theta_{ik})^{-1} J_2(k_e).$$

To obtain the rate of molecular-ion formation, we must compute the dipole moment  $\mathbf{d}$  of the three-body system:

$$\mathbf{d} = \mathbf{r}_1 + \mathbf{r}_2 - \mathbf{r}_\mu - \mathbf{r}_c.$$

On inversion of the relations between the  $\mathbf{r}_i$  terms and  $\mathbf{r}_c$ ,  $\mathbf{r}_n$ , and  $\mathbf{R}_{\mu}$ , given in I, we find

$$\mathbf{d} = (f_2 - f_1) \mathbf{r}_n - (1 + \rho_e) \mathbf{R}_{\mu}.$$

Here  $\mathbf{R}_{\mu}$  is the vector to the center of mass of the two nuclei, while the mesonic wave function is expressed in terms of the vector from the center of charge,  $\mathbf{R}_{\mu}'$ . Making this change of variable, we obtain

$$\mathbf{d} = \frac{1}{2} (f_2 - f_1) (1 - \rho_e) \mathbf{r}_n - (1 + \rho_e) \mathbf{R}_{\mu'}.$$

In the case of greatest interest, the initial state is an s state, and the final (bound) state, a p state. It is con-

venient to quantize the latter in the direction **k**. The only component of the matrix element of **d** different from zero will then be that in the direction parallel to **k**. The first term of  $d_k$  is independent of the mesonic variables, and we obtain for it

$$\left(\frac{4\pi}{3}\right)^{\frac{1}{2}}(f_2-f_1)(1-\rho_e)\int_0^\infty r_n dr_n [\chi_+^{I*}\chi_+^F + \chi_-^{I*}\chi_-^F],$$

where the  $\chi^{I}$  is chosen to give the proper asymptotic dependence for the incident wave, while  $\chi^{F}$  is a bound state, normalized so that we have

$$\int_0^\infty dr_n\{|X_+^F|^2+|X_-^F|^2\}=1.$$

The angular dependence of the bound state is  $(3/4\pi)^{\frac{1}{2}}$  $\times \cos\theta$ , where  $\theta$  is the angle between  $\mathbf{r}_n$  and  $\mathbf{k}$ . In the ellipsoidal coordinates, one readily finds  $\mathbf{R}_{\mu}' \cdot \mathbf{r}_n/r_n = -(r_n/2)\xi\eta$ . The other two components of  $\mathbf{R}_{\mu}'$  vanish on integration over  $d\tau_{\mu}$ , and so we find for the second term in  $d_k$ :

$$\left(\frac{4\pi}{3}\right)^{\frac{1}{2}}(1+\rho_{\mu})\int_{0}^{\infty}r_{n}dr_{n}[\chi_{+}^{I*}\chi_{-}^{F}+\chi_{-}^{I*}\chi_{+}^{F}]\langle\xi\eta\rangle,$$

where we define

$$\langle \xi\eta 
angle = \int d au_{\mu} \psi_{+} \xi\eta \psi_{-}.$$

On carrying out this integration employing the approximate  $\psi_+$  and  $\psi_-$  used in I, we obtain

$$\langle \xi \eta \rangle = \frac{E_3(\Sigma P/2) [C_1(\Sigma) - C_1(\Delta)] - E_1(\Sigma P/2) [C_3(\Sigma) - C_3(\Delta)]}{[E_2(P_+)C_0(Q_+) - E_0(P_+)C_2(Q_+)]^{\frac{1}{2}} [E_2(P_-)C_0'(Q_-) - E_0(P_-)C_2'(Q_-)]^{\frac{1}{2}}},$$

where the notation is the same as in I. This expression is correct for  $r > r_c$ , and for  $r < r_c$  the subscripts of some of the  $E_n$  terms must be modified in a manner similar to that carried out in I.

In the  $(p\mu p)$  system, we have  $f_1=f_2$ , and the first term in **d** vanishes. Thus, in this case, mesonic transitions are necessary to obtain a nonzero matrix element for  $d_k$ . In addition, the bound-state function,  $\chi_{+}^{F}$ , is purely  $\chi_{+}^{F}$ , so that we obtain

$$(I | d_k | F) = \left(\frac{4\pi}{3}\right)^{\frac{1}{2}} (1 + \rho_{\mu}) \int_0^\infty r_n dr_n \chi_{-I^*} \langle \xi \eta \rangle \chi_{+F}.$$

In the  $(p\mu d)$  system it is necessary to keep all the terms. In both cases, the final integrations were carried out numerically.

In computing the decay between two bound states, as is done in Sec. III B, the initial state is normalized differently. The radial function is normalized as in Eq. (4), while an s state has an angular function equal to  $(4\pi)^{-\frac{1}{2}}$ . Otherwise the calculation is as above.