Ref.TH.2679-CERN

HYDROGENIC MESOMOLECULES AND MUON CATALYZED FUSION

J. Rafelski
CERN -- Geneva

A B S T R A C T

Hydrogenic mesomolecules are discussed with particular emphasis on their rôle in the μ catalyzed fusion process. Recent theoretical and experimental evidence for weakly bound mesomolecules $dd\mu$ and $dt\mu$ derived from resonant mesomolecular formation and its dependence on the temperature is described. The fate of the muon stopped in dense hydrogenic target is followed as well as that of a muon sticking to the fusion product.

Lectures at the International School of Physics of
Exotic Atoms, Erice

25 March -- 5 April 1979

Ref.TH.2679-CERN

8 June 1979

1. - Introduction

With respect to many atomic and molecular processes the lifetime of the free muon, $\tau_{\mu} = 2.20 \times 10^{-6}$ sec, is sufficiently long to consider it as a stable object. So the possibility arises that mesomolecules of heavy hydrogen isotopes are formed. Such a mesomolecule is, up to recoil corrections, a scaled down model of the usual one-electron hydrogen molecule, H_2^+ , with the scaling factor being the ratio of muon to electron mass, 206.8. In particular, the hydrogenic isotopes are only about $1\text{\AA}/206.8 = 500$ fm apart. Consequently, the probability for spontaneous fusion is appreciable especially for fusion reaction channels governed by strong interactions. This is in particular the case for the dd and dt mesomolecules.

In view of these circumstances we must examine in more detail the processes leading to formation of mesomolecules - if they were sufficiently fast, one could think of a chain of reactions in which muons play the rôle of a catalyser : as soon as a mesomolecule is formed, a fusion process occurs, subsequently the freed muon forms a new mesomolecule, etc.

This reaction chain is interrupted either by the decay of the muon, or by its capture by an element other than hydrogen - also by its sticking to the fusion product, He.

This fascinating mechanism to catalyze "cold" fusion demands the understanding of many fundamental aspects of atomic and molecular physics and for that reason it has attracted much interest during the past decades. The first fusion catalyzed by muons has been discovered in 1956 in Berkeley ¹⁾ but theoretical work ²⁾⁻⁶⁾ goes back to the original suggestion by Frank in 1947. Interest persisted through the early sixties, when most of the relevant rates seemed to have been established qualitatively ^{5),6)} indicating that this process could not be of practical importance in the immediate future.

During the past years we have seen again growing interest in the μ catalyzed fusion mainly due to the discovery of the temperature dependence of the reaction rate ⁷⁾ for μ catalyzed fusion in deuterium, its theoretical explanation in terms of weakly bound mesomolecular states ⁸⁾⁻¹⁰⁾ and the suggestion to use the cold fusion as the heating mechanism to start the fusion reaction in compressed pellets ¹¹⁾. In these lectures, we shall discuss in particular these new developments that depend on new insights into mesoatomic and mesomolecular physics. To readers looking for a longer introductory paper to the subject of mesomolecular physics we recommend the excellent review by Gershtein and Ponomarev ¹²⁾.

Upon a short introduction to the present status of the μ catalyzed fusion in the next section, where we shall also present the relevant data, we turn to the discussion of special questions particularly important for the process of the catalyzed fusion in the subsequent sections. In Section 3 we will describe the general approach to the three-body Coulomb problem when one of the bodies (muon) is (not much) lighter than the others. Then

the process of resonant mesomolecule formation is discussed in Section 4. This is the new mechanism predicted by Vesman⁸⁾ that has reopened the field of μ catalyzed fusion. Then we turn to the discussion of the possible fate of the muon in liquid hydrogen. In particular in Section 5 we consider the probability that the muon will stick to the fusion product, helium, which would remove it from the catalytic chain of reactions. In the following section, we consider, assuming our present theoretical knowledge of the different rates discussed in Section 2, the necessary conditions to catalyze 100-200 fusions - a number that seems feasible today. In the last section we present an outlook devoted in the past to the discussion of fundamental physical problems involved.

In order to distinguish the different isotopes of hydrogen we will refer to ^1H as p or protium, to ^2H as d or deuterium and to ^3H as t or tritium. The word "(liquid) hydrogen" implies a mixture of these three isotopes.

2. - Overview

When a muon enters a hydrogen target, a chain of reactions begins - here we list those that dominate the history of the muon in a liquid ($\rho = 4.25 \times 10^{23}/\text{cm}^3$) hydrogen.

- 1 μ is stopped,
- 2 μ is captured by the dominant isotopic fraction,
- 3 μ cascades to the mesohydrogenic 1s state,
- 4 μ can be transferred to the heavy isotopes,
- 5 μ mesomolecule formation,
- 6 de-excitation to the mesomolecular ground state,
- 7 muon decay or nuclear fusion,
- 8 muon sticking to He, or at, point 1 or 2.

When fusion occurs, muon can be shaken off and is then available to repeat the chain of reactions 1 to 7. Naturally muons can decay at any stage between 1 and 7 but it is usually the mesomolecular state that involves a p that lives longer than

the muon lifetime. It can be assumed that about, or less than, 10^{-9} sec are required for fast muons to slow down and reach the 1s state in liquid hydrogen ¹²⁾. The properties of the bound mesohydrogenic 1s states are given in Table 1.

The last column gives the hyperfine splitting which plays an important rôle in the moderation of mesonic hydrogen. We further notice the strong isotope effects ΔE_{ij} on the binding energy of the 1s state. In particular, it leads to relatively high transfer rates from light to heavier isotopes. In Table 2 we give the results of the calculations by Ponomarev et al. ¹³⁾ for liquid hydrogen at room temperature for these transfer rates. Experimental figures for the $p\mu + d$ process agree well with theoretical calculations and among themselves up to a factor of two.

Table 1 : Main characteristics of mesonic atoms

M	M/m _e	-E _{1s} (eV)	ΔE (eV)	ΔE^{hfs} (eV)
μ	206.769			
p	1836.152	2528.437	$\Delta E_{pd} = 134.705$	0.183
d	3670.481	2663.142	$\Delta E_{pt} = 182.745$	0.049
t	5496.918	2711.182	$\Delta E_{dt} = 48.040$	0.241

Table 2 : Transfer rates of muons
between hydrogen isotopes

$p\mu + d \rightarrow p + d\mu$	$\Lambda_T \cdot \tau_\mu \sim 3.7 \times 10^4$
$p\mu + t \rightarrow p + t\mu$	$\Lambda_T \cdot \tau_\mu \sim 1.6 \times 10^4$
$d\mu + t \rightarrow d + t\mu$	$\Lambda_T \cdot \tau_\mu \sim 220$

From the point of view of other atoms, a mesic atom acts in many respects as if it were a "neutron" ; it is neutral and its size of ~ 500 fm is quite small when compared with the usual atomic sizes. So it can easily penetrate the electron cloud, without feeling the repulsive Coulomb potential of the atomic nucleus. However, when a mesic hydrogen approaches another hydrogen isotope relatively closely, a mesomolecule may be formed accompanied by conversion (Auger emission) of an electron. Such a mesomolecule is, from the viewpoint of other electronic atoms a very heavy isotope of hydrogen, being relatively small. Thus it can form an electronic molecule with another hydrogen atom. An observant reader will notice, at this point, that we could, in the first place, attach the mesic hydrogen to one of the centres of the electronic molecule (see Fig. 2.1). To do so without the conversion electron we must have an accidental degeneracy⁸⁾. The gain in energy from the mesomolecular formation must be absorbed by the internal degrees of freedom of the electronic molecule. Indeed, such a degeneracy has been observed⁷⁾

Figure 2.1 : Illustration of the resonant mesomolecule formation in the process $\mu t + (d_2ee) \rightarrow [(td\mu)dee]^*$.

and we shall speak of this process as the resonant mesomolecule formation. Naturally, the rates of the resonant process can be influenced by external parameters, such as temperature ⁸⁾. We will discuss this at length further below.

In order to achieve an accidental degeneracy one has to compare energies of the electronic molecular hydrogen of the order eV with those of mesonic atoms and molecules that are usually 200 times larger. However, the mesic molecules can have weakly bound excited states. In Table 3 we show the mesomolecular spectrum recently obtained by the Dubna group ^{10),14)} for the mesomolecular energies. J is the rotational quantum number and ν the vibrational quantum number. We notice that both in $dd\mu$ and $dt\mu$ weakly bound states of 2 and 0.9 eV have been predicted. This is a very important result, since it opens the possibility of the accidental degeneracy described above. The energies given in Table 3 are the binding energies

Table 3 : Binding energy $\epsilon_{J\nu}$ (eV) of μ molecules of the hydrogen isotopes

Angular momentum	$J = 0$		$J = 1$		$J = 2$	$J = 3$
Vibrational state	$\nu = 0$	$\nu = 1$	$\nu = 0$	$\nu = 1$	$\nu = 0$	$\nu = 0$
$pp\mu$	253.0	-	105.6	-	-	-
$pd\mu$	221.5	-	96.3	-	-	-
$pt\mu$	213.3	-	97.5	-	-	-
$dd\mu$	325.0	35.6	226.3	2.0	85.6	-
$dt\mu$	319.1	34.7	232.2	0.9	102.3	-
$tt\mu$	362.9	83.7	288.9	44.9	172.0	47.7

with respect to the mesic atom - that is, e.g., in the case of a $d\mu$ molecule, the total binding of the μ is obtained adding 2711.18 eV from Table 1. From this example, we see that in order to establish that the mesomolecular bound state actually exists, one has to solve the three-body Coulomb problem with a relative precision better than 10^{-4} !

We note further that the weakly bound states happen to have allowed E1 transitions to the ground states of the respective mesomolecules. Consequently, within 10^{-10} sec, we are actually in the mesomolecular ground state and the nuclear fusion can follow.

In Table 4 we give the different possible fusion reactions that can proceed in mesomolecular states. The reactions of dd and dt are here of particular interest. They can proceed via strong interactions and are accompanied by the emission of fast neutrons. For this reason there is a significant recoil of helium as indicated in the last column of Table 4. Due to this recoil, muons can be shaken off the fusion product. We shall consider the probability W_s of the muon to stick to the recoiling helium further below in Section 5.

A compilation of the essential rates concerning the catalyzed fusion process is shown in Table 5. We see that in the dd and dt reactions the non-resonant molecular formation rate is very small. This has been the bottle-neck in the catalytic chain until now. We notice that when the resonant mechanism is included, there is still the bottle-neck for the $d+d$ reaction (the reader is warned that all numbers involving t are theoretical predictions). Without the resonant effect the chances for two fusions generated by one muon are highest in the $d+d$ case and should occur in less than $\sim 10\%$ of the events (in liquid hydrogen). This changes when we satisfy the resonance conditions in the dt reaction. Here we expect up to 100-200 fusions per muon under optimal conditions.

Table 4 : Fusion reactions, their Q values and recoil

Fusion reaction	Q value	Recoil/Nuc.
$p + p \rightarrow d + e^+ + \nu$	2.2 MeV	-
$p + d \rightarrow {}^3\text{He} + \gamma$	5.4 MeV	-
$t + p \quad (50\%)$	4 MeV	-
$d + d \rightarrow {}^3\text{He} + n \quad (50\%)$	3.3 MeV	0.27 MeV ($\frac{1}{12} E$)
${}^4\text{He} + \gamma \quad (\sim 0\%)$	24 MeV	-
$d + t \rightarrow {}^4\text{He} + n$	17.6 MeV	0.88 MeV ($\frac{1}{20} E$)
$p + t \rightarrow {}^4\text{He} + \gamma$	20 MeV	-
$t + t \rightarrow {}^4\text{He} + n + n$	10 MeV	-

Table 5 : Essential rates for catalyzed fusion (liquid hydrogen density)

Process	Non-resonant molecular rate $\Lambda_m \cdot \tau_\mu$	Total molecular rate $\Lambda_{mT} \cdot \tau_\mu$	Nuclear fusion rate $\Lambda_f \cdot \tau_\mu$	Fusion before sticking W_s^{-1}
$p\mu + p$	5	5	~ 0	-
$d\mu + p$	5 ~ 10	5 ~ 10	0.7	1
$d\mu + d$	0.1	1.8	$\sim 10^5$	~ 8
$t\mu + d$	0.07	207	$\sim 10^6$	~ 100
$t\mu + p$	1	1	10	1
$t\mu + t$	1.5	1.5	?	1

We shall now explain in the next sections how the critical numbers Λ_{mT} and W_s can be computed. We start with a brief discussion of the three-body Coulomb problem in the adiabatic basis.

3. - Mesomolecular spectrum

In this section we shall only outline the main aspects of the theoretical approach and describe the difficulties in computation of weakly bound mesomolecular states. As indicated in Section 2 we want to establish the binding energy of a muon in a mesomolecule up to 0.1 eV out of c. 3 keV. Thus we must reach a relative precision of better than 10^{-4} . On the other side we notice that since the muon is relatively heavy we must take account of the nuclear recoil due to the muon motion. Both these facts make the numerical work extremely difficult, while the formulation of the problem is relatively straightforward.

In the Born-Oppenheimer adiabatic approach the fast motion of the muon is treated as a separated two-centre problem for a fixed separation $|\vec{R}|$ between the nuclei that are assumed to oscillate slowly. Thus we solve the Schrödinger equation to obtain the adiabatic basis :

$$\left(-\frac{1}{2m_\mu^r} \Delta_{\vec{r}} - \frac{e_1 e}{r_1} - \frac{e_2 e}{r_2}\right) \phi_n(\vec{r}; R) = \epsilon_n(R) \phi_n(\vec{r}; R) \quad (3.1)$$

where m_μ^r is the reduced mass of the muon with respect to the two centres $M_1 + M_2$: $1/m_\mu^r = 1/m_\mu + 1/(M_1 + M_2)$. The quantities r_1 and r_2 are the distances of the muon from the centres 1 and 2, as shown in Fig. 3.1. In the adiabatic approximation the total wave function $\psi(\vec{r}, \vec{R})$ of the three-body problem is expanded in terms of this two-centre basis

$$\psi_i(\vec{r}, \vec{R}) = \sum_n \phi_n(\vec{r}; R) \chi_n^i(\vec{R}) \quad (3.2)$$

Figure 3.1 : The co-ordinates of the three-body Coulomb problem.

ψ_1 satisfies the three-body Coulomb problem Hamiltonian. In order to obtain an equation for $\chi(R)$ we can consider the projection of the total three-body problem with the basis ϕ_n . We then find

$$\begin{aligned} & \left(-\frac{1}{2\bar{M}} \Delta_{\vec{R}} + \frac{e_1 e_2}{R} + E_n(R) - E_i \right) \chi_n^i(\vec{R}) = \\ & = \frac{1}{2\bar{M}} \sum_k \int d^3r (\phi_n^* \Delta_{\vec{r}} \phi_k) \chi_k^i + \frac{1}{\bar{M}} \sum_k \int d^3r (\phi_n^* \vec{\nabla}_{\vec{r}} \phi_k) \cdot \vec{\nabla}_{\vec{R}} \chi_k^i \end{aligned} \quad (3.3)$$

where $1/\bar{M} = 1/M_1 + 1/M_2$ and $\bar{M} = M m_\mu / m_\mu^r$.

Usually the dynamic coupling of the molecular motion of the nuclei to the muon wave function, the right-hand side of Eq. (3.3), is neglected and we have in the left-hand side of Eq. (3.3) the usual molecular problem. Another way of seeing this is to assume that m_n/M is very small : this is the case for electronic atoms.

In particular when weakly bound molecular states are considered we have to include the effects of the right-hand side of Eq. (3.3). The diagonal ($n=k$) terms can be easily included in the molecular potential on the left-hand side of

Eq. (3.3). The off diagonal terms demand a simultaneous solution of coupled channel equations, a task recently performed by the Dubna group ^{10),14)}. With the high accuracy required here, in particular, the coupling to the continuum is essential to obtain the proper energy shown in Table 3 for the $J=1, v=1$ states in dd and dt . The multichannel calculations increase the binding in both cases by ~ 1.3 eV (a 10^{-4} effect on the total energy). In the simple molecular picture the $dd\mu$ state could be weakly bound, while the $dt\mu$ state would be weakly unbound. The dynamic coupling makes also the $(dt\mu)$ state weakly bound and opens the way for new and exciting process of μ catalyzed dt fusion

In Table 3 we showed the difference between the molecular energy E_i and the atomic energy of the heaviest isotope given in Table 1. Thus Table 3 gives the additional binding due to the presence of a second centre ; the molecular quantum number in Table 3, angular momentum J and vibration v , are summarized by the index i in Eq. (3.3) while n describes the muon configuration of the molecule.

As we have already emphasized, the absolute precision of both atomic and molecular energies must exceed 0.1 eV in order to obtain to within 10% the energies of weakly bound mesomolecular states. In order to appreciate the level of precision needed, let us consider as an example the effect of vacuum polarization. The vacuum polarization shifts for the $1s$ states in $\mu p, \mu d$ and μt are ¹⁵⁾ -1.896, -2.126, -2.212 eV, respectively (the minus sign indicates increased binding). The molecular energies are affected also, since the form of the wave functions at the nuclear centre is different from the atomic case and because there is additional repulsion between the nuclei due to the internuclear vacuum polarization. Fortunately, these effects nearly cancel for the weakly bound molecular states : Melezhik and Ponomarev ¹⁵⁾ find 0.008 eV for

the $(dd\mu)_{J=1}^{v=1}$ state and -0.003 eV for the $(dt\mu)_{J=1}^{v=1}$ state. We will discuss further below that even these small contributions will ultimately be measurable and are relevant for the μ catalyzed process. Naturally we must gain control of all other contributions to the molecular energy levels at this high relative precision of 10^{-6} - 10^{-7} beforehand. It is worth mentioning that this precision has an "equivalent" of perhaps 10^{-10} - 10^{-11} in electronic molecules, due to the increased importance of the recoil effect in mesic molecules.

4. - Resonant formation of mesomolecules

From the point of view of an electron a mesic hydrogen looks much like a quasi-neutron. Thus it can penetrate the electron cloud of the molecule and stick to one of the molecular centres. This process can proceed without the conversion (Auger emission) of an electron only when the mesomolecular energy gain - the energy given in Table 3 - is degenerate with vibrational or rotational states of the electronic molecule. We must also take under consideration the thermal energy ϵ_T at temperature T

$$\begin{aligned}\epsilon_T &= \frac{3}{2} kT \\ &= .054 \text{ eV for } T = 400^\circ \text{K}\end{aligned}\tag{4.1}$$

We recall that the thermal distribution

$$\gamma(\epsilon, \epsilon_T) = \left(\frac{27}{2\pi} \frac{\epsilon}{\epsilon_T} \right)^{1/2} \frac{1}{\epsilon_T} \exp(-\frac{3}{2} \epsilon / \epsilon_T) \tag{4.2}$$

is a weakly peaked distribution, thus the resonance condition on the energy of the electron molecule ϵ_r

$$\epsilon_r = \epsilon_T + \epsilon_{mol}^{\mu} \tag{4.3}$$

is not very pronounced - in the distribution (4.2) we have in the range of temperatures $30^{\circ}\text{K} < T < 1000^{\circ}\text{K}$ always an appreciable probability of finding a particular value of ϵ in the domain $0.005 < \epsilon < 0.1 \text{ eV}$.

The resonance condition is shown in Fig. 4.1 with the special example of the $[(dt\mu)d2e]$ molecule illustrated in part (b) of the Figure. As indicated there, the amount of $\sim 1 \text{ eV}$ mesomolecular binding can be absorbed by exciting the fourth vibrational state. In the case of the $[(dd\mu)d2e]$

(a)

(b)

Figure 4.1 : a) Schematic level scheme of mesomolecules in the effective potential and the thermic energy.
b) Resonant molecular formation and the excitation of electromolecule.

molecule it is the eighth vibrational state that is excited by ~ 2 eV mesomolecular energy. Another component in the energy balance indicated in Fig. 4.1 is the rotational energy of the electronic molecule, before and after the attachment of the mesonic t or d to one of the molecular centres. This energy is

$$E^J = J(J+1)/2\Theta$$

$$\Theta = R^2\mu \quad (4.4)$$

Here μ is the reduced mass of the two molecular centres of the electromolecules and $R = 1.4\text{\AA}$. Up to isotopic effects of the order of 10% this energy is $0.004 \frac{1}{2}J(J+1)$ eV. We have to include a suitable amount of energy for the change in the rotational quantum number by one. We note that at $T \sim 400\text{--}500^\circ\text{K}$ several rotational states ($J \leq 3$) of the molecule can be excited thus making the set of rotational-vibrational states suitable for accidental degeneracy dense.

We expect that the molecular rates for the formation of $(dt\mu)$ and $(dd\mu)$ molecules will depend on temperature as illustrated by the resonance condition (4.3) and in Fig. 4.1. Indeed in a recent experiment of Bystritsky, Dzhelepov et al.⁷⁾ this effect has been investigated systematically. The collection of all available results for the rate of formation of the $(dd\mu)$ molecule is shown in Fig. 4.2. We see a systematic behaviour and the rate rises by factor 10 when T reaches 400°K . The solid line is the result of the theoretical study of Vinitzky, Ponomarev et al.¹⁰⁾ - a satisfactory agreement between theory and experiment can be achieved.

In view of these experimental and theoretical results we are led to the conclusion that the similar effect predicted for the $(dt\mu)$ molecule should also be present, leading to very

high mesomolecular formation rates. The theoretical prediction here is based on the lower excitation of the electromolecule in order to absorb the lower mesomolecular binding - ~ 1 eV - of this mesomolecule. Gerstein and Ponomarev ⁹⁾ base their prediction of $\Lambda_m \cdot \tau_\mu \approx 200$ on the excitation of the fourth vibrational state in the electromolecule. Indeed the success of the theory for the $(dd\mu)$ molecule as documented by Fig. 4.2 lends credibility to this prediction. On the other hand we should remember that a relative error of 10^{-5} in the determination of the total energy (c. 2710 eV) of the mesomolecule $(dt\mu)$ for $J=v=1$ would significantly change the resonance condition and the predicted mesomolecule formation rate. In the detailed numerical work the difference between the $(dd\mu)$ and $(dt\mu)$ molecules is large - the latter being asymmetric, with the weak binding provided by the non-trivial terms in the right-hand side of Eq. (3.3) Furthermore the adiabatic

Fig. 4.2 : Collection of all data on the $dd\mu$ formation rate as function of temperature

approach is not quite correct, as will be described in the next section. This also influences the energies of the weakly bound states. We see that we must establish the total mesomolecular energy with a precision of 10^{-5} - 10^{-6} to obtain a reliable prediction for A_m . As we have already discussed, even the effect of the vacuum polarization has to be considered - and indeed the numbers given at the end of the previous section indicate that this effect changes the resonant temperature by ~ 5 - 10% . Incidentally, the measurement of the resonance temperature can be viewed as the measurement of the molecular vacuum polarization effect ¹⁵⁾.

5. - Muon sticking probability

The motion of the muon is faster than that of the oscillating nuclei - that is the basis for the adiabatic approach outlined in Section 3. Thus the muon cloud can adjust its wave function to each distance R between the oscillating nuclei. When we follow the mesomolecular energy in the ground state of (dt) as a function of R , we see that it evolves into the $1s$ state of helium in the united atom limit. Thus when fusion occurs in the vicinity of $R=0$, muon wave function resembles much the $1s$ wave function of helium. The process of fusion itself is sudden from the point of view of the muon. Let us now start the calculations assuming with Zel'dovich and Gershtein ⁶⁾ that the initial state of the muon when fusion occurs is the $1s$ state in helium.

We can apply the sudden approximation to compute the probable fate of the muon after fusion. That is we consider probability amplitude

$$W_{fi} = \int d^3x \phi_f^{*He} \psi_i \quad (5.1)$$

where φ_i is the initial muon wave function and ϕ_f^{He} is the final state of the muon in the presence of the fusion product, here indicated by the upper index He in a particular channel of the dd fusion and for all dt fusions.

We now concentrate on the fusion in the dtμ molecule, this being the only case where many fusions seem feasible per muon considering the numbers given in Table 5. In the process of the dt fusion 17.6 MeV energy is liberated of which a large part is carried away by the neutron, the remainder of c. 3.5 MeV being the recoil energy of the α particle (see Table 4). Thus with respect to the muon "frozen" in the orbital in the sense of the sudden approximation, the new nucleus moves with substantial momentum P_{rec} . This situation is illustrated in Fig. 5.1. Therefore, if we want to find the probability of sticking W_s , of the muon to the moving helium we must find its overlap with the moving bound muonic state of the fusion product. Thus :

$$W_s = \sum_n W_{ni} = \sum_n \int d^3x \Phi_n^{\text{He}}(\vec{x}, P_{\text{rec}}) \varphi_i(\vec{x}) \quad (5.2)$$

where

$$\Phi_n^{\text{He}}(\vec{x}, P_{\text{rec}}) = \varphi_n^{\text{He}}(\vec{x}) e^{i \vec{P}_{\text{rec}} \cdot \vec{x}} \quad (5.3)$$

and φ_n^{He} are the usual Schrödinger Coulomb bound states of He. As already discussed φ_i is taken to be the 1s state of He.

The largest contribution in Eq. (5.2) arises from the $n=1s$ state. This has been computed ⁴⁾ to be

$$\begin{aligned} W_s &\approx (1+G)^{-2} \approx .01 \\ R &= E_{\text{rec}}^M / 4E_0 \end{aligned} \quad (5.4)$$

Figure 5.1 : Muon sticking in fusion.

where E_{rec}^{μ} is the muon kinetic energy when sticking and ϵ_0 its binding energy in the $1s$ state of helium : 11.1 keV. In the case of the dt fusion $E_{\text{rec}}^{\mu} \sim 0.1$ MeV and we find the value indicated in Eq. (5.4). All other matrix elements in Eq. (5.2) are negligible compared to (5.4) (see below).

So far this calculation has neglected the non-adiabasy of the nuclear motion and the slight difference in the initial wave function as compared to $1s-\mu\text{He}$ and further states in Eq. (5.2). Clearly, a more thorough study is needed here ; in particular the non-adiabasy effect will lead to a reduced population of the $1s\text{-He}$ state. This is due to the fact that the nuclear oscillations are only several times slower than muonic velocities. Thus during the nuclear motion leading to

the united atom limit muons from the (μdt) molecule can populate the 2s or even 2p state in helium. For the symmetrical ($dd\mu$) molecule only the 2s state needs to be considered, but it could be populated relatively more strongly. Incidentally the above-described non-adiabasy affects also slightly the energies of mesomolecules, as mentioned before.

It is less likely that more weakly bound muon in the 2s state will stick - it has larger chances to be shaken off - we find the sticking probability in the 2s state to be ~ 5 times smaller than in the 1s state. The probability that the muon will then continue in the 1s state dominates this contribution. Of this order of magnitude is also the sticking to the 2s state if the initial state is 1s. Thus we conclude that in most cases the sticking muon will be in the 1s state of the recoiling helium, although some population of the 2s state can be expected.

As we continue to follow the fate of the muon that sticks to the fusion product, we realize that the muon is not lost yet from the catalytic chain of reactions. The muonic helium will have appreciable kinetic energy of the order of 3.6 MeV - the muon's part being some 100 KeV. Thus there is enough energy to overcome the binding energy of muons in Helium of 11 keV in the 1s state, or less if transfer to a bound state of d or t occurs. Thus muons can be, in principle, reactivated.

First we observe that those muons that are initially in the (metastable) 2s state will either be directly transferred to the t or d 1s state or decay to the 1s state in consequence of the collisional quenching. As regards the transfer process we only recall that the 2s state of He and the 1s state of t are almost degenerate. However, it can be expected that the collision induced radiative decay to the 1s state of

He will dominate the transfer process in the collision of $(\mu\text{He})^+$ with d^+ or t^+ . Again this point deserves further attention.

Let us now consider what is known on the fate of the muon in the $1s$ state of He. In particular the process of direct ionization into the continuum has been considered by Jackson⁵⁾. Using the conventional Bohr model he finds an activation probability of $\sim 40\%$ for the muon. He then corrects his estimate comparing with the (electronic) experimental He^+ ionization cross-sections in H_2 and finds 22% . He neglects the fact that from the point of view of a muon many more channels are open - there is only one muon at a time. Thus direct transfer processes from $\mu\text{He}^+ + d$ or t , forbidden in the He^+ experiment can now proceed. It is very likely that the transfer processes are as important as the direct ionization contribution. To see this we recall that we are in the sudden limit of the Coulomb collision process. The velocity of the recoiling helium is initially c. 5 times higher than that of the muon in orbit. Therefore direct ionization can proceed only in relatively deep $\mu\text{He}^+ + d$ (or t) collisions, where the high frequencies are available. But in such collisions the muon may choose to go to the bound states of d (or t) and this at comparable strength. In liquid hydrogen many such collisions will occur, since the slowing down by electro-excitation of electrons is $\sim 10^{-9} - 10^{-10}$ sec while the collision time for Coulomb collisions is $\sim 10^{-11} - 10^{-12}$ sec. Therefore even a small probability for the transfer process can add up to a substantial contribution. We note that since in direct ionization more energy is required than to transfer the muon from the $1s$ state in He to the $1s$ state in t or d the direct transfer process could be more important than the ionization into continuum. Finally, there is the possibility that multistep processes will occur, that is that the muon will be excited to an intermediate state of

helium in one collision and in a second collision it will be either transferred or ionized. However, since the radiation rate for dipole transitions is 10^{13} /sec, most of the intermediate states will have decayed before the second collision takes place. All these processes can be computed and the degree of difficulty does not exceed the one of similar calculations in atomic physics. Since the need for a more precise study of muon regeneration has arisen only recently, this problem has not been settled yet.

For completeness we would like to mention that in the dd fusion reaction an estimate of the sticking probability gives $W_{dd} \sim 0.1-0.2$, in view of the now much smaller recoil (see Table 4). For the same reason the reactivation probability is negligible. Nonetheless, as can be seen from Table 5, it is the mesomolecule formation rate that limits here the number of possible fusions.

6. - How to catalyze 100-200 fusions in dense hydrogen

We consider the fate of a muon stopped in hydrogen isotope mixture. Our assumption will be that the ratios of rates that follow from Tables 2, 5 are correct. In Fig. 6.1 we see a flow chart with the essential reaction channels concerning muon transfer and mesomolecule formation. The numbers show us rate ratios at equal relative isotope densities. They indicate up to which relative isotope density a particular reaction is dominant.

Let us begin at the point $pp\mu$ in Fig. 6.1. We have here the competition between the $pp\mu$ molecule formation and the transfer to d or t.

Since once $pp\mu$ molecule is formed, the muon is definitively lost from the reaction chain, we conclude that the common relative heavy isotope concentration must be larger than c. 1%

Figure 6.1 : Muon flow chart in hydrogen with the relative strength of reaction channels indicated at equal relative isotope density.

to allow that the $pp\mu$ molecule is avoided in 100-200 reactions. Since we wish a dt reaction we must have both isotopes present. Therefore let us turn now to the point $d\mu$ in Fig. 6.1. Now we must avoid both the $dp\mu$ and $dd\mu$ molecules. In order that the transfer of μ from d to t can proceed 200 times the relative concentration of t and p must be of the same order of magnitude ; otherwise the $pd\mu$ molecule is formed. Only now are we sure that all muons find their way to tritium. We can now turn our attention to the $t\mu$ point in Fig. 6.1. In order to avoid the $pt\mu$ molecule we must have as many d 's present as p 's. Consequently we arrive at the result that all isotopes must be present in about equal (order of magnitude) relative concentrations in order to sustain 100-200 ($dt\mu$)

mesomolecule formations per lifetime of the muon. In this optic the presence of p 's seems not to be desirable. Naturally a completely different conclusion is reached when one wants to measure relative rates and uses competitive and known p processes as reference. Then a fraction of the $(dt\mu)$ formation per muon is sufficient, thus p can dominate the isotope mixture.

Now we continue our quest for 100-200 fusions and consider, in view of the above conclusions, the d - t isotope mixture as shown in Fig. 6.2. We wish to determine the best concentrations c_d and c_t of d and t , respectively, with the property :

$$c_d + c_t = 1 \quad (6.1)$$

Thus we must minimize the fraction of losses due to the formation of $dd\mu$ and $tt\mu$ molecules. We shall denote by R_d the reaction branching ratio at the point $d\mu$ in Fig. 6.2 :

Figure 6.2 : Muon flow chart in deuterium-tritium.

$$R_d = \frac{\Lambda_{dd\mu}^m \cdot C_d}{\Lambda_{dt}^T \cdot C_t} = r_d \frac{C_d}{C_t} \quad (6.2)$$

where r_d is $\sim 1/200$ and the upper indices m and T refer to mesomolecules and transfer reactions, respectively. Similarly at the point $t\mu$ we introduce the ratio

$$R_t = \frac{\Lambda_{tt\mu}^m \cdot C_t}{\Lambda_{dt\mu}^m \cdot C_d} = r_t \frac{C_t}{C_d} \quad (6.3)$$

The actual prediction for r_t at the resonance of the $dt\mu$ ratio is $\sim 1/150$. We assume that the capture of the muon in d or t is directly proportional to their respective relative densities. Thus the probability of loss of the muon from the chain of catalyzed fusion reactions is given by

$$P_{\text{loss}} = C_d \frac{R_d}{1+R_d} K_d + \left(C_t + \frac{C_d}{1+R_d} \right) \frac{R_t}{1+R_t} K_t \quad (6.4)$$

The factors K_i express the fact that the loss of the muon may not be necessarily permanent - in the $dd\mu$ case it corresponds to the sticking probability W_{dd} (see Section 5). It is perhaps of the same order of magnitude in the case of the $tt\mu$. Both K_d and K_t should be largely independent of c_d and c_t . Thus we can minimize Eq. (6.4) with respect to c_t upon elimination of c_d with the help of Eq. (6.1). We have, since for $c_d \sim c_t$, $R_d \sim R_t \ll 1$

$$P_{\text{loss}} \approx \frac{(1-C_t)^2}{C_t} (r_d K_d) + \frac{C_t^2}{1-C_t} (r_t K_t) \quad (6.5)$$

and from $\partial P_{\text{loss}} / \partial c_t = 0$ we find (the inverse) relation

$$\chi = \frac{(1-C_t)^3}{C_t^3} \frac{C_t+1}{2-C_t} \quad , \quad 0 < C_t < 1 \quad (6.6)$$

Figure 6.3 : Left scale, full line - optimum density for tritium as a function of X , the ratio of rates, Eq. (6.7). Right scale, dotted and dashed lines - reduced loss rates of muons in deuterium-tritium mixture.

where

$$X = \frac{r_t K_t}{r_d K_d} = \frac{\Lambda_{tt\mu}^m}{\Lambda_{dd\mu}^m} \cdot \frac{\Lambda_{dt}^T}{\Lambda_{dt\mu}^m} \cdot \frac{K_t}{K_d} \quad (6.7)$$

In Fig. 6.3 (solid line, left scale) we show c_t as a function of X as defined by Eq. (6.6). As can easily be expected when the losses to $tt\mu$ and $dd\mu$ are equal and $X=1$ we find $c_t = \frac{1}{2}$. As we can see in Fig. 6.3, a large change in X is reflected by a small change of optimum c_t . Since we expect $X \sim 1$, c_t will be ~ 0.5 . This is, as one could expect in the first place, but we have also now a quantitative feeling about the dependence of c_t on the different rates. For $X=1$, i.e., $c_t = \frac{1}{2}$, we can easily compute $P_{\text{loss}} = r_d K_d \sim 1/2000$. Thus we see that if the estimates of the different rates are correct, we could have more fusions (2000) than can be sustained at the present theoretical molecular formation rate $\Lambda_{dt\mu}^m$ during the

lifetime of the muon. This situation is illustrated by the dotted line in Fig. 6.3, right scale, which describes P_{loss} at the extremum of Eq. (6.5). The inverse of the dotted values times $(r_d K_d)^{-1}$ is the number of fusions expected (up to the restriction imposed by lifetime of the muon). Thus we see that as X decreases below 1 the number of possible fusions increases (at constant $r_d K_d$). The opposite is true at constant $r_t K_t$ as shown in Fig. 6.3 by the dash-dotted curve (right scale).

Thus we see that competitive processes do not limit the number of fusions in a d-t mixture and that the actual limit is imposed at present by the mesomolecular formation rate as compared to the lifetime of the muon. As we have alluded to before, when this limit is overcome by some kind of activation of chemical rates by external parameters, it will be ultimately the slow-down time of the muon that will limit the number of fusions per muon to at most 10.000.

So far we have excluded from our discussion the possibility that μd reacts directly with t to form a μdt molecule (and similarly with other systems). Here the energetics of the reaction changes, since we are gaining in addition to the energies shown in Table 3 the isotopic energy shown in Table 1, in the dt case, 48.04 eV. This is the reason why the transfer process is a much more likely event, except if there were a molecular resonant state for $J=2$ or 3 at this energy - but there is no indication at present that this is the case.

7. - Outlook

We have seen that the prediction of the weakly bound mesomolecular ($dd\mu$) and ($dt\mu$) states has led to a breakthrough and has completely changed the relevance of muon catalyzed fusion among the mesomolecular reactions. In particular

recent experimental evidence for temperature dependence of the mesomolecular formation rate of the $dd\mu$ molecule lends strong credibility to the theoretical work.

Many relevant rates seem not to be known that determine the fate of muons stopped in a deuterium-tritium mixture. There is in particular strong interest in the muon transfer rates between heavy hydrogen isotopes and in molecular formation rates that involve tritium. Apparently, the problem of the muon sticking to the fusion products, as well as the subsequent muon reactivation probability in the process of slowing down of muonic helium through collisions with heavy hydrogen isotopes, seems not to have been definitively settled.

The important fundamental aspect of this field is the test of our ability to understand the three-body Coulomb problem, including such an effect as the vacuum polarization.

We have so far been assuming that the resonant reaction proceeds in thermal equilibrium. However, as is now beginning to be known, the chemical reaction rates can be activated by external means such as high intensity lasers ¹⁶⁾. Another reason to think off-equilibrium is the fact that any muonic hydrogen is formed at finite - order eV - kinetic energy. Given the dense set of vibrational and rotational states of the electronic (dd) and (dt) molecules - resonant formation of a mesonic (dt) molecule could proceed even before thermal equilibrium has been reached - this particular aspect is absent if the dominant isotope is p. The non-equilibrium reactions do not seem to depend on the density of the hydrogenic target but on the number of collisions until thermalization, thus on the initial kinetic energy of the μt and the relative isotopic concentrations.

It goes without saying that even though the catalized fusions may lead to more thermal energy than the mass of the muon, the energy lost in the conventional way of making muons will probably outweigh the eventual fusion gains - even perhaps in most optimistic circumstances. However, one can think of the "cold" catalyzed fusion as a possible ignition point of the hot fusion process. Anyway an effort to reduce the energy cost for a muon could be of great practical importance concerning the relevance of the μ catalyzed fusion process.

In the last paragraph of this overview, let me remind experimentalists reading this paper that the μ catalyzed fusion is still practically an open field. Clearly tritium is, in many respects, the name of the game - to the author's knowledge no experimental rates have been established involving t and μ up-to-day.

Acknowledgements

I would like to thank L.I. Ponomarev and H. Rafelski for many fruitful discussions that have stimulated many of the thoughts presented here as well as J. Duclos for his constant interest and encouragement. I also would like to thank G. Fiorentini for a careful reading of the manuscript and stimulating discussions that have clarified many points of this report and led to the discussion of the muon regeneration problem as presented in Section 5.

References

1. 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, M.L. Stevenson, H.K. Ticho and R.D. Tripp, Phys.Rev. 105:1127 (1957).
2. F.C. Frank, Nature 160:525 (1947).
3. A.D. Sakharov, Report of the Physics Institute, Academy of Sciences (1948).
4. Ya.B. Zel'dovich, Dokl.Akad.Nauk SSR 95:493 (1954).
5. J.D. Jackson, Phys.Rev. 106:330(1957).
6. Ya.B. Zel'dovich and S.S. Gershtein, [Soviet Uspekhi 3:593 (1961)], Usp.Fiz.Nauk 71:581 (1960).
7. V.M. Bystritsky, V.P. Dzhelepov, A.I. Rudenko, V.M. Suvorov, V.V. Filchenkov, N.N. Khovanskii and B.A. Khomenko, "Mesons in Matter", Proc.Intern.Symp. on Mesonic Chemistry and Mesic Molecular Process in Matter, Dubna (1977).
8. E.A. Vesman, Zh.Eksp.Theor.Fiz.Pisma 5:113 (1967) ; [Soviet Phys.JETP Letters 5:91 (1967)].
9. S.S. Gerstein and L.I. Ponomarev, Phys.Letters 72B:80 (1977) ; Erratum 76B:664 (1978).
10. S.I. Vinitzky, L.I. Ponomarev, I.V. Puzynin, T.P. Puzynina L.N. Somov and M.P. Faifman, "Resonance Formation of Hydrogen μ Mesomolecules", Dubna Preprint P4-10929 (1977) ; Zh.Eksp.Teor.Fiz. 74 (1978).
11. W.P.S. Tan, Nature 263:656 (1976).
12. S.S. Gershtein and L.I. Ponomarev, "Mesomolecular Processes Induced by μ^- and π^- Mesons", in Myon Physics III - Chemistry and Solids, V.W. Hughes and C.S. Wu Editors, Academic Press, New York (1975), p. 141.
13. L.I. Ponomarev, " μ Atomic and μ Molecular Processes in Hydrogen Isotope Mixtures", SIN Preprint PR-77-011 (1977).
14. S.I. Vinitzky, L.I. Ponomarev, I.V. Puzynin, T.P. Puzynina and L.N. Somov, "The Calculation of the Energy Levels of the Hydrogen Isotope μ Molecules in the Adiabatic Representation", Dubna Preprint P4-10336 (1976).
15. V.S. Melezhik and L.I. Ponomarev, Phys.Letters 77B:217 (1978).
16. A.M. Ronn, "Laser Chemistry", in the May 1979 issue of "Scientific American", p. 103. See also references p. 150.