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THEORETICAL SURVEY OF μCF

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INTRODUCTION

Since not everyone attending this Workshop is a μCF expert. I will try to give a helpful introduction to the theory of this fascinating subject. concentrating on the d-t fusion cycle. As many of you know, the largest part of this body of theory has been developed by Leonid Ponomarev and his colleagues and collaborators over the last two decades. We are fortunate in having Professor Ponomarev with us at this Workshop, and look forward to hearing from him later this morning.

The main steps in the muon-catalyzed d-t fusion cycle are shown in Fig. 1. Most of the stages are very fast, and therefore do not contribute significantly to the cycling time. Thus at liquid H_2 densities (ϕ=1 in the standard convention) the time for stopping the negative muon, its subsequent capture and deexcitation to the ground state is estimated to be $\sim 10^{-11}$ sec.[1] The muon spends essentially all of its time in either the (dμ) ground state, waiting for transfer to a (tμ) ground state to occur, or in the (tμ) ground state, writing for molecular formation to occur. Following the formation of this "mesomolecule" (actually a muonic molecular ion), deexcitation and fusion are again fast. Then the muon is (usually) liberated to go around again. We will now discuss these steps in some detail.

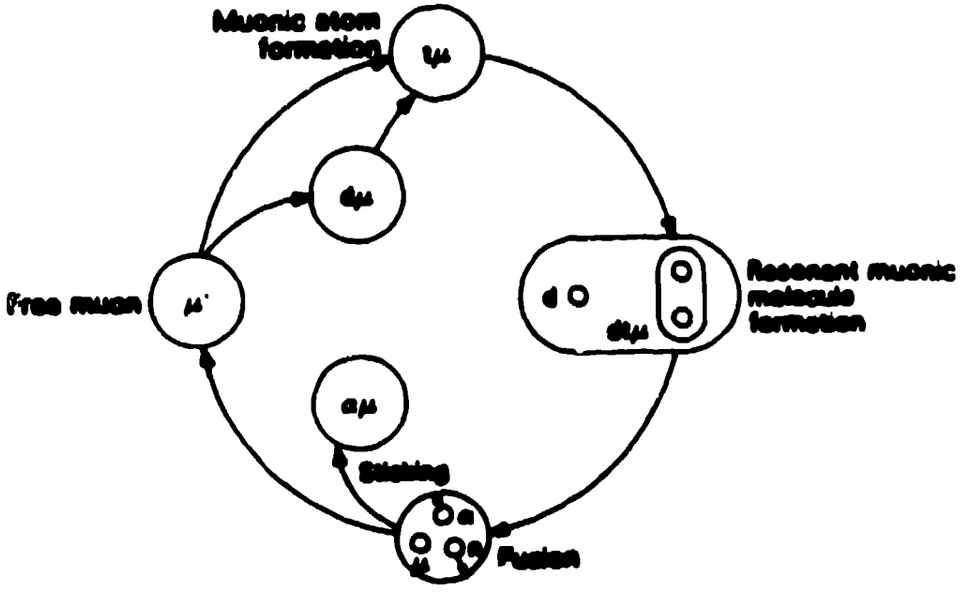

Fig. 1. The muon-catalyzed d-t fusion cycle.

MOLECULAR FORMATION

The *Auger* mechanism, where the energy released in forming the mesomolecule is carried off by an ejected electron, has been calculated by Ponomarev and Faifman,[2] using a two-level (or Born-Oppenheimer) approximation. The resulting molecular formation rate is quite slow for $dt\mu$: $\lambda^e_{dt\mu} \sim 3\text{x}10^{-4}s^{-1}$ at room temperature. (These rates are conventionally normalized to liquid H_2 target density, 4.25×10^{22} atoms/cm^3.)

This Auger rate is completely dwarfed by the rate for *resonant molecular formation*, the mechanism first suggested by Vesman[3] in 1967 to explain the striking observed temperature dependence for $dd\mu$ formation. This mechanism requires the existence of a loosely bound state, with binding energy $\lesssim 4$ eV, so that the energy released upon mesomolecular formation can go into the vibration and rotation of the resulting compound molecule:

$$t\mu + (D_2)_{0,0} \rightarrow [(dt\mu)^* dee]^*_{\nu,J} \qquad (1)$$

This one-body final state means that the cross section for this process contains δ-functions at the resonance energies; temperature dependence of $\lambda_{dt\mu}$ from this mechanism comes from the overlap of these δ-functions with the Maxwell distribution of $t\mu + D_2$ kinetic energy.

The first requirement in proving the validity of this resonance idea is an accurate calculation of the binding energies of the postulated bound states. This task was embarked upon by Ponomarev and his collaborators following Vesman's suggestion, and required their developing the adiabatic representation (method of perturbed stationary states) into a powerful tool for calculating mesomolecular wave functions (solving the non-relativistic Coulomb three-body problem). By 1977 Vinitsky and Ponomarev[4] established that for the (J,v)=(1,1) (J $\equiv$ angular momentum, v $\equiv$ vibration quantum number) state of $dd\mu$ and $dt\mu$, $\epsilon_{11} \simeq -2.2$eV and -1.1eV respectively. This enabled Gershtein and Ponomarev[5] to point out that the resonant molecular formation rate $\lambda_{dt\mu} \simeq 10^8 s^{-1}$, so that $\simeq 100$ fusions per muon in a dense D-T target is possible. This prediction of a hundred fusions per muon did much to trigger the present blossoming of interest in μCF.

The adiabatic representation has continued to develop,[6] but was overtaken (at least as far as claimed precision in the binding energy is concerned) about two years ago by variational calculations. The recent values $\epsilon_{11} = -1.975$ eV (-0.660 eV) for $dd\mu(dt\mu)$ should be accurate to a fraction of an meV, so that it is the "relativistic" corrections to these energies that are of greatest current interest. For $dd\mu$, the vacuum polarization term is completely dominant, providing + 10 meV out of a total correction to ϵ_{11} of +11 meV.[7] In contrast, for $dt\mu$ the nuclear charge distribution is dominant, contributing 15 meV of a total + 23 meV correction. Higher precision calculations are now under way, and we can hope to hear about them soon.

There are two channels of molecular formation that are especially sensitive to these energies, because the resonance energies turn out to be small. First, in the $dd\mu$ reaction

$$(d\mu)^F + (D_2)_{0,0} \rightarrow [(dd\mu)^S_{11} dee]_{7,1} \qquad (2)$$

with $d\mu$ hyperfine state $F=\frac{3}{2}$ and $dd\mu$ total spin $S=\frac{1}{2}$, the resonance energy according to Zmeskal et al.[8] is only about 4 meV; this gives rise to the spectacular (and unexpected) hyperfine effect observed at SIN.[9] Since the temperature dependence of $\lambda_{dd\mu}^{\frac{3}{2}\rightarrow\frac{1}{2}}$ should eventually allow the determination of this resonance energy to within about 0.1 meV, this case will provide an extremely stringent test of the calculated $dd\mu$ J=1, v=1 binding energy, including all the corrections.

The other sensitive reaction is in $dt\mu$ formation:

$$(t\mu)^F + (D_2)_{0,J} \rightarrow [(dt\mu)^S_{11} dee]_{2,J+L} \qquad (1')$$

with F=0, S=1. Here the strongest resonances (those with orbital angular momentum L=0,1) actually lie *below* threshold! This circumstance is believed to lead to the three-body contribution to $\lambda_{dt\mu}$ discovered at LAMPF[10] (see below).

The pioneering calculation of the rates for resonant molecular formation (the Vesman mechanism) was made by Vinitsky et al.[11] in 1977; the importance of L,J and hyperfine effects,[12] and electron screening[13] was pointed out somewhat later. This left the computed molecular formation rates significantly smaller than the experimental value.[14,15,8] The situation was remedied when Menshikov and Faifman[16] pointed out the importance of using *undistorted* $d\mu$ + d and $t\mu$ + d wave functions in calculating the transition matrix element. Finally Menshikov et al.[17] for the $dd\mu$ case pulled all the many strands together – including "back decay" (the reverse of reaction 2)[18] and transitions of the $[(dd\mu)dee]^*$ complex – in a beautiful *ab initio* calculation, which fits the experimental results[8,14,15] very well indeed.

The extension of this calculation to $\lambda_{dt\mu}$ is straight forward for all channels except the crucially important F=0, D_2 reaction, with its strong, below-threshold resonances (presumably $J \rightarrow J'$: $0 \rightarrow 1$, $0 \rightarrow 2$, $1 \rightarrow 2$, etc.). The precise location of these resonances will soon be known, when the precise calculations of the relativistic corrections are completed. At low enough density, treatment of the F=0, D_2 channel is also straightforward: the Auger deexcitation of the $[(dt\mu_{11}dee]^*$ complex broadens the resonance δ-functions into Lorentzians,[19] with width $\Gamma_e \simeq 0.8$ meV.[20] As a result the contributions of the below-threshold resonances are dominant, the higher-L resonances being suppressed by a centrifugal barrier factor.[12] (This assumes that the $t\mu$ atoms are thermalized at the target temperature.)

At higher densities ($\phi >$ few %), three-body contributions to molecular formation become signification.[21] The *impact approximation*, so important for the theory of collisional line broadening in spectroscopy, has been applied extensively to this problem.[19,22–25] This entails including in the total width of the Lorentzians a *collisional contribution*, expressing the effect of collisions with neighboring molecules:

$$\Gamma = \Gamma_e + \phi\Gamma_c \qquad (3)$$

with

$$\Gamma_c = n_o < v\sigma > . \qquad (4)$$

But what collision cross section σ should be used here? Petrov[19] and others[22,23,25] have used only the inelastic cross section on the final state, while in optics the initial state inelastic scattering plus the integral of the square of the difference of the elastic scattering amplitudes also enters:[27]

$$\sigma = \sigma^i_{inel} + \sigma^f_{inel} + \int d\Omega \mid f^i_{el}(\Omega) - f^f_{el}(\Omega) \mid^2 . \tag{5}$$

In contrast, Menshikov[24] claims that, because of the momentum carried by the $t\mu$, the appropriate cross section is the final state *total* cross section:

$$\sigma = \sigma^f_{el} + \sigma^f_{inel}. \tag{6}$$

However, in spite of its seductiveness, it is evident that the basic conditions for the impact approximation are *not* met for this problem of molecular formation.[26] There are two conditions: First, the "detuning" (displacement from the imperturbed energy) ΔE must be related to the collision duration τ_c by[27]

$$\mid \Delta E \mid << \frac{\hbar}{\tau_c}. \tag{7}$$

For a realistic inter-molecular interaction and the temperature range of interest, the RHS of this relation is $\sim$1 meV; thus the impact approximation is confined to a completely uninteresting energy range. This limitation arises because, for $\Delta E \gtrsim \hbar/\tau_c$, information about the scattering wave function is needed for times $t < \tau_c$, *not* just the asymptotic properties of this wave function (phase shifts, etc.). A second condition is the requirement of purely binary collisions; this leads to[26]

$$\phi < \begin{cases} 0.14 & @300K \\ 0.02 & @\ 30K \end{cases} , \tag{8}$$

which by itself removes most of the experimental data points!

It appears that what is needed is the generalization (to the massive incoming $t\mu$ as opposed to the photon) of the (many-body) *quasistatic approximation* of collisional line-broadening theory[28] (which may be related to the "Quasi-resonant molecular formation" of Menshikov and Ponomarev[21]). In my opinion, the theory of three-body molecular formation barely exists at present.

A possibility mentioned several times[29,30] for solid and liquid targets is that one or more *phonons* carry off enough energy to make the strongest resonance ($0 \rightarrow 1$, at $\sim$ - 12 meV) accessible to the physical region. So far, only a calculation for a *metallic*, rather than molecular, hydrogen target has appeared.[31]

DEEXCITATION AND FUSION

Once the $[(dt\mu)^*_{11} dee]^*$ complex is formed, Auger transitions will carry it quickly to a J=0 state of $dt\mu$ where fusion is very rapid. The chain of transitions has been studied by Bogdanova et al.[20]

The proper method of calculating the fusion rate λ_f (and the sticking, discussed below) has generated some controversy. What we shall refer to as the *orthodox* view, expounded by Bogdanova,[32] Markushin[33] and others, holds that a good approximation is provided by the simple formulae

$$\lambda_f = \kappa \cdot p \tag{9}$$

$$p \equiv \int d^3\mathbf{r} \mid \Psi^{o,v}(\mathbf{r},\mathrm{o}) \mid^2, \tag{10}$$

$$\kappa \equiv \lim_{v \to 0} [v\sigma C^{-2}], \tag{11}$$

where C is the Gamow factor for the d-t system, σ is the reaction cross section (d + t $\rightarrow$ ^{4}He + n), and $\Psi^{J,v}$ (**r**,**R**) is the three-body wave function for the $(\mathrm{dt}\mu)_{Jv}$ mesomolecule (**R** being the d-t seperation and **r** the muon position relative to the d-t CM). This approximation could fail (in the orthodox view) only if significant *rearrangement* of the dtμ spectrum by the strong interaction occurs. This would happen if the $^5\mathrm{He}^*(\frac{3}{2}^+)$ resonance, which dominates the fusion reaction, were to be closely degenerate with a dtμ bound state. However, both the resonance energy and its width serve to prevent this rearrangement.

This point of view is supported., e.g., by the recent R-matrix calculation of Struensee et al.,[34] which finds λ_f values close to the orthodox results. Obviously, the *dissenters* don't agree, and I am sure we will be told why during this Workshop.

STICKING AND REACTIVATION

Sticking of the negative muon to the daughter He nucleus limits the number of fusions per muon that can be attained. Two factors are involved: *initial sticking* $\omega_s^0(n,\ell)$ in the (n,ℓ) state of the ($\mu\alpha$) system, and the probability $[R(n,\ell;\phi)]$ for *reactivation* of the muon during the slowing-down of the $(\mu\alpha)^+$:

$$\omega_s(\phi) = \sum_{n,\ell} [1 - R(n,\ell;\phi)]\omega_s^0(n,\ell). \tag{12}$$

The simplest approximation for the initial sticking uses the *adiabatic* (Born-Oppenheimer) approximation for the muon wave function as the d and t approach one another, and takes the overlap (*sudden* approximation) with the final state wavefunction of a muon traveling in a bound state around the retreating α-particle:

$$\omega_s^0(n,\ell) = |< \Psi_{1s}(\mu - {}^5He) \mid e^{iM\mathbf{V}\cdot\mathbf{r}}\Psi_{n\ell 0}(\mu - \alpha) >|^2 . \tag{13}$$

Calculated in this way the total initial sticking comes out to be[35]

$$\sum_{n,\ell} \omega_s^0(n,\ell) = 1.16\%. \tag{14}$$

The largest correction to this approximation comes from the fact that the true wave function does not adiabatically adjust as the d-t separation goes to zero, but rather "lags-behind". Thus instead of $\Psi_{1s}(\mu - {}^5He)$ in Eq.

13, the true three-body wave function, with $R \to o$, is needed. The results with the wave function calculated by several different methods (quantum Monte Carlo,[36] adiabatic representation,[35] variational[37]) agree quite well and reduce the sticking by about 25%:[38]

$$\Sigma\omega_s^0 = 0.85\%. \tag{15}$$

Next, we consider strong interaction effects on ω_s°, in particular the effect of the energy dependence of the $\frac{3}{2}^+$ ^{5}He* t-matrix element. According to the orthodox view, this affects only the non-adiabatic corrections to ω_s°, and merely increases $\Sigma\ \omega_s^\circ$ by $< 3\%$.[32,33] The dissenting view is quite different; e.g., Rafelski et al.[39] claim the strong interaction effects reduce $\Sigma\omega_s^\circ$ by a factor of two! We will certainly hear from both sides during the Workshop.

The reactivation of muons in stripping or transfer collisions is very important, especially for $dt\mu$ because here the recoil velocity of the α is so large ($v \simeq 6$ a.u.). The reactivation depends on the competition among all the excitation, deexcitation, Stark mixing and transfer processes, and the slowing of the $(\alpha\mu)^+$. Since the $R(n,\ell;\phi)$ obviously depends drastically on the (n,ℓ) values, there is an important dependence on the initial populations $\omega_s^\circ(n,\ell)$. The most complete calculations are those of Cohen[40] and Markushin,[41] which in fact agree very well for $R(\phi)$ and the x-ray intensities $K_\alpha(\phi)$, $K_\beta(\phi)$, ... The x-ray intensities ($dd\mu$ as well as $dt\mu$) are in fair agreement with SIN results.[42] The theoretical $R(\phi)$ variation with density ϕ is too small to account for the variation in $\omega_s(\phi)$ reported from the LAMPF neutron data.[10]

SCAVENGING BY HELIUM

^{3}He from tritium decay appears in any d-t experiment, so He scavenging will always be present. There are obviously six possible reactions:

$$x\mu + {}^A\mathrm{He} \to {}^A\mathrm{He}\mu + x \tag{16}$$

(x = p,d,t; A = 3,4).
The reactions are believed to proceed via the (Auger) formation of the $(xHe\mu)$ mesomolecular state $2p\sigma$, followed by radiative dissociation:[43]

$$\begin{aligned} x\mu + He &\to (xHe\mu)_{2p\sigma} + e^- \\ &\hookrightarrow (He\mu)_{1s} + x + \gamma \end{aligned} \tag{17}$$

The rates $\lambda_{x^A_{He}}$ as functions of temperature have been calculated by Fomichev et al.[44] The predictions that (1) $\lambda_{x^A_{He}}(T)$ increases as T decreases, and (2) $\lambda_{t^3_{He}}(T)$ is an order of magnitude larger than $\lambda_{d^3_{He}}(T)$, appear to be experimentally verified.[43,45]

In addition to this ground-state transfer, for d-t targets (with their large cycling rates) the initial capture by, and excited-state transfer to, He is significant; the total scavenging rate will have both contributions:

$$\lambda_{He} = \lambda_{He}(g.s.) + \lambda_c\omega_{He}. \tag{18}$$

The quantity ω_{He} is analogous to $(1-q_{1s})$ (see below). So far, there is no calculation of this quantity.

ELASTIC SCATTERING

Elastic scattering of muonic atoms from target molecules plays an extremely important role in the catalysis cycle, since it determines the rate of thermalization of the $d\mu$ and $t\mu$ atoms. The early calculations of Matveenko and Ponomarev[46] used a simple two-level (Born-Oppenheimer) approximation. More recent work (Melezhik and collaborators[47,48]) uses the adiabatic representation (PSS method) for calculating the nuclear scattering ($d\mu$ + d, etc.). To this is added the effect of electron screening (in Born approximation), and of molecular structure (using the Fermi pseudo-potential method). An example of the resulting cross sections, for $d\mu$ ($F=\frac{1}{2}$) scattering from d,D, and D_2 is shown in Fig. 2.

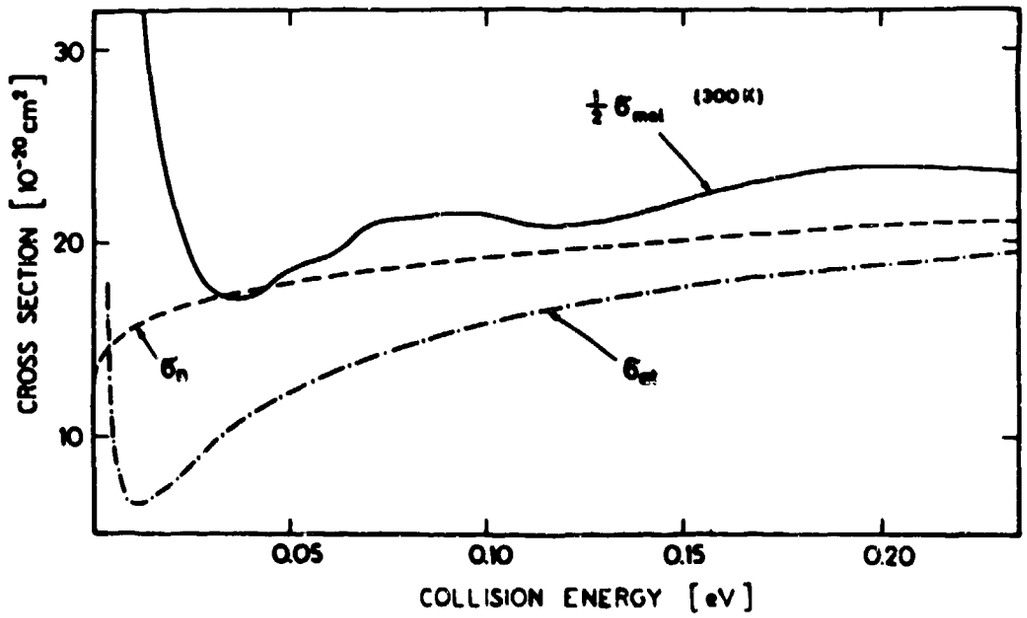

Fig. 2. Elastic scattering of $F=\frac{1}{2}$ $d\mu$ atoms from d (σ_n), D atoms (σ_{at}) and D_2 molecules (σ_{mol}) (from ref. 48).

HYPERFINE TRANSITIONS

The hyperfine transitions

$$(d\mu)^{F=\frac{3}{2}} + d \rightleftharpoons (d\mu)^{F=\frac{1}{2}} + d \tag{19}$$

$$(t\mu)^{F=1} + t \rightarrow (t\mu)^{F=0} + t \tag{20}$$

come about because of *exchange scattering*. Matveenko and Ponomarev[46] calculated these rates using the two level approximation. Recent calculations of Melezhik and collaborators[47] use the adiabatic representation. Results for the rates for $d\mu$ + d are shown in Fig. 3, along with the low temperature SIN point.[9] While this looks like good agreement, the theoretical value will be significantly raised when the contribution from *resonant hyperfine quenching*,[17,49], i.e., the sequence

$$(d\mu)^{\frac{3}{2}} + D_2 \rightarrow [(dd\mu)^* dee]^* \rightarrow (d\mu)^{\frac{1}{2}} + D_2, \tag{21}$$

is included, thus leaving a discrepancy.

For quenching of the triplet $t\mu$ state the older two-level calculations[46] giving $\lambda_t = 9 \times 10^8 s^{-1}$ agree quite well with adiabatic representation result $9.1 \times 10^8 s^{-1}$ of Melezhik.[47] Kammel et al.[50] at SIN looked for the 'build-up' in time of the neutron signal at very low C_t ($\equiv$ tritium fraction) but saw only a very much faster build-up; this *may* indicate an experimental λ_t significantly larger than the predicted value. More experimental information is sorely needed.

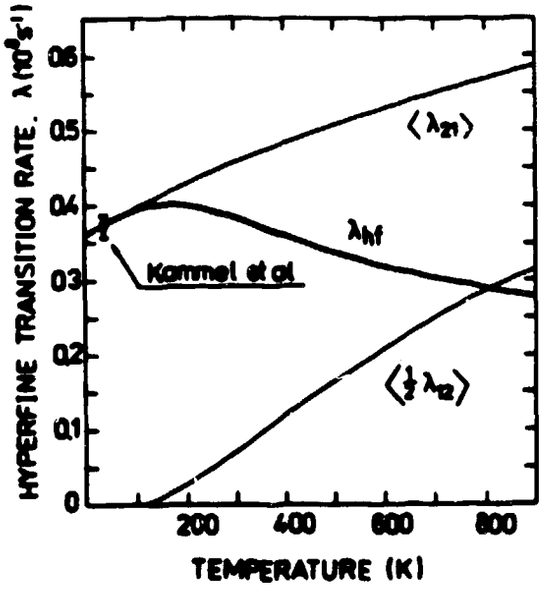

Fig. 3. Hyperfine transition rates for $d\mu$ atoms colliding with d's (from ref. 47).

d → t TRANSFER

The two-level calculation of Matveenko and Ponomarev[46] gave $1.9 \times 10^8 s^{-1}$ for the (ground state) d → t transfer, while the recent calculations of Melezhik[51] and Kobayashi et al.[52] give 2.7 and $2.6 \times 10^8 s^{-1}$ respectively. In discussing the measured values of λ_{dt}, we must keep in mind that these

depend on what is *assumed* about the triplet quenching rate λ_t. The experimental numbers from several groups, $\simeq 2.8 \times 10^8 s^{-1}$,[14,15,53,54] correspond to neglect of the $t\mu$ triplet state, i.e., $\lambda_t = \infty$; smaller values of λ_t give larger λ_{dt} values.

KINETICS OF THE d-t CYCLE

In general, in order to calculate the rate of production of fusion neutrons as a function of time in terms of the underlying physical rates, it is necessary to solve the kinetic equations describing the system.[55] However, for the steady-state cycling rate λ_c (i.e., after any transients have become negligible), it is sufficiently simply to add the times the muon spends in each state, to get the cycle time[12]:

$$\lambda_c^{-1} = T_d + T_t, \tag{22}$$

$$T_d = \frac{q_{1s} C_d}{\lambda_{dt} C_t} \qquad (d\mu \text{ ground state}) \tag{23}$$

$$T_t = T_t^1 + T_t^\circ \qquad \text{(triplet and singlet } t\mu \text{ ground state)}; \tag{24}$$

here,

$$T_t^1 = \frac{\frac{3}{4}}{\lambda_t C_t + \lambda_{dt\mu}^1 C_d}, \tag{25}$$

$$T_t^\circ = \frac{\frac{1}{4} + \frac{3}{4}\chi}{\lambda_{dt\mu}^\circ C_d}, \tag{26}$$

and the branching ratio χ is given by

$$\chi = \frac{\lambda_t C_t}{\lambda_t C_t + \lambda_{dt\mu}^1 C_d}. \tag{27}$$

These times and rates are normalized to liquid-hydrogen density, and the $dd\mu$ and $tt\mu$ channels are neglected. If the high temperature ratio of D_2 and DT molecules holds, we can write for the molecular formation rates

$$\lambda_{dt\mu}^F = C_d \lambda_{dt\mu-d}^F + C_t \lambda_{dt\mu-t}^F. \tag{28}$$

The factor $q_{1s}(\phi, C_t)$ is the probability of a muon, initially captured into a highly excited $d\mu$ atom, *not* transferring to a t during the (mainly collisional) deexcitation cascade. While some calculations of q_{1s} have been carried out,[56,57] the experiments seem to favor less drastic ϕ and C_t dependence than predicted. It seems likely that the deexcitation cascade is much more complex than the present models allow.

CONCLUSIONS

While much has been accomplished toward a complete quantitative description of the d-t catalysis cycle, the job is by no means finished. In particular, a really quantitative theory of three-body (many-body) molecular formation is sorely lacking. A more detailed treatment of the deexcitation cascade, from the initial atomic capture of the μ^- to the ground state $d\mu$ or $t\mu$, is needed for calculating ω_{He} and more realistic $q_{1s}(\phi,C_t)$ values. And according to the dissenting view mentioned above, more careful treatment of the strong-interaction effects on λ_f and ω_s° is called for.

More precise calculations of the relativistic corrections to the $dt\mu$ and $dd\mu$ binding energies will soon become available, so that the two-body molecular formation rates $\lambda_{dd\mu}$ and $\lambda_{dt\mu}$ can be computed more precisely. Presumably, more complete results for elastic scattering, hyperfine quenching and $d \rightarrow t$ transfer will also soon be completed, allowing more definitive conclusions about thermalization, kinetics, etc. Then comparison with experiment will show where problems remain.

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