Effects of Intramolecular Dynamics on Nuclear Fusion Rates and Sticking from Resonant States of the Molecular Ion $dt\mu$

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Several resonances of s symmetry in the continuum of the muonic molecule $dt\mu$ that cluster below the $(t\mu)_{2s}+d$ threshold have been analyzed. It has been found that the fusion rates for some of these states are comparable with their Coulomb decay rates, and that the associated sticking probability is approximately 1 order of magnitude lower than that of the ground state. This feature indicates that muon-catalyzed fusion might proceed more efficiently if it were occurring from the resonant states formed from the excited ($n=2$) states of muonic atoms.

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Negative muons may be used as a catalyst to fuse hydrogen nuclei into helium. The necessary confinement of nuclei is obtained on a microscopic scale by chemical bonding within "exotic" muonic molecules such as $dt\mu$. The fusion energy released by muon catalysis exceeds the rest-mass energy of participating muons, which has provoked a great deal of attention and extensive theoretical and experimental studies; for reviews, see, e.g., [1,2].

A single muon can catalyze many fusion reactions via spontaneous and recurrent formation of muonic molecules in D_2+T_2+DT mixture. The number of fusions Y catalyzed by a single muon is inversely proportional to the probability of muon loss, mostly by natural decay and by α capture after fusion reaction: $Y\simeq[\tau_c/\tau_0+(1-R)\omega_s^0]^{-1}$, where τ_c is the cycling time between consecutive fusions, $\tau_0=2.2$ μs is the lifetime of the muon, ω_s^0 is the branching ratio between reactions $dt\mu\rightarrow\alpha\mu+n$ and $dt\mu\rightarrow\alpha+\mu+n$ (known as the sticking fraction), and R is the probability of collisional stripping of a muon lost by α capture (R varies from 0.28 to 0.34 in the range of typical mixture densities between 0.1 and 1.2 of liquid hydrogen density [3]). For high cycling rates ($\tau_c\ll\tau_0$) the cumulative effectiveness of muon-catalyzed fusion (μCF) becomes $Y\simeq 1/(1-R)\omega_s^0$ and is hampered by the intrinsic value of sticking fraction ω_s^0.

The sticking fraction from a given molecular state is mostly dependent on the heat of the nuclear reaction, 17.6 MeV [4]. If fusion must proceed via bound states of the $dt\mu$ molecule, intrinsic sticking appears to have a fixed "God given" value $\omega_s^0=0.886\%$ (or $w_s^0=0.917\%$ including nuclear force effects) [5].

Recent calculations have established the existence of three-body resonances in $dt\mu$ [6–10]. In the present Letter we present an analysis of the fusion rates and $\alpha\mu$ sticking probabilities for a few of the resonant states that are below the $(t\mu)_{2s}$ threshold of the muonic molecule $td\mu$. We show that three-body resonances constitute a class of fast fusing states with low sticking fractions (as compared with the same properties of the ground state). We demonstrate that the sticking fraction is strongly dependent on the properties of the molecular state leading to fusion, and that the resonant states provide an example of a situation where the Coulombic forces in the low energy initial channel "modulate" the properties of the highly energetic final state with energy determined by the nuclear reaction.

The resonant states in question may be formed by the mechanisms considered in the bound state formation, the new features being that the initial state is in the $(t\mu)_{2s}+d$ molecular continuum, and that the formation entrance width is dictated by the width of the truly metastable state $dt\mu^*$ [which removes the formal difficulty of the molecular formation via the $dt\mu(1,1)$ state whose width is 0 [11]].

A resonant state can decay via Coulombic interactions or fusion according to the following reactions:

$$(dt\mu)^* \xrightarrow{\lambda_{t\mu+d}} t\mu+d, \text{ muon transfer}, \tag{1}$$

$$(dt\mu)^* \xrightarrow{\lambda_{d\mu+t}} d\mu+t, \text{ elastic decay}, \tag{2}$$

$$(dt\mu)^* \xrightarrow{\lambda_{\alpha+n+\mu}} \alpha+n+\mu, \text{ fusion without sticking}, \tag{3}$$

$$(dt\mu)^* \xrightarrow{\lambda_{\alpha\mu+n}} \alpha\mu+n, \text{ fusion with sticking}. \tag{4}$$

We focus on establishing the ratio of the rate for Coulombic decay to the rate of nuclear fusion, $\kappa=\lambda_f/\lambda_d$ (with $\lambda_f=\lambda_{\alpha+n+\mu}+\lambda_{\alpha\mu+n}$ and $\lambda_d=\lambda_{d\mu+t}+\lambda_{t\mu+d}$), and the sticking fraction, $\omega_s^0=\lambda_{\alpha\mu+n}/(\lambda_{\alpha\mu+n}+\lambda_{\alpha+n+\mu})$.

The resonances reported here were obtained by means of a variational calculation within the framework of the complex coordinate method (CCM). The details of the CCM can be found elsewhere [12], here we will only mention that one studies the spectral properties of the dilated Hamiltonian $H(\eta)=U(\eta)HU^{-1}(\eta)$ where the transformation $U(\eta)$ is defined as $Uf(\mathbf{r})=\eta^{3/2}f(\eta\mathbf{r})$ and η is a complex dilation parameter: $\eta=\alpha e^{i\beta}$. The complete three-body Coulombic problem for the $dt\mu$ molecule is described by the Hamiltonian [6]

$$H(\eta)=-\frac{\eta^{-2}}{2}\nabla_1^2-\frac{\eta^{-2}}{2}\frac{m_{\mu t}}{m_{\mu d}}\nabla_2^2-\eta^{-2}\frac{m_{\mu t}}{m_\mu}\nabla_1\cdot\nabla_2+\frac{1}{\eta r_1}+\frac{1}{\eta r_2}-\frac{1}{\eta r_{12}}, \tag{5}$$

where r_1 and r_2 are the muon-triton and muon-deuteron distances, respectively, r_{12} denotes the internuclear distance, and $m_{\mu t}$ and $m_{\mu d}$ refer to reduced masses of the muon-triton and muon-deuteron systems, respectively, while the muon mass is denoted by m_μ. The values of m_t, m_d, and m_μ are 5496.899, 3670.481, and 206.7686, respectively. The continuous spectrum of the dilated Hamiltonian is rotated out in the complex plane by an angle β around the consecutive thresholds. For the $dt\mu$ system, such rotation separates the branches belonging to $(t\mu)_{nl}+d$ and $(d\mu)_{nl}+t$ thresholds. The bound states remain uninfluenced by the transformation, whereas the discrete complex spectrum corresponds to resonances.

The solution of the dilated eigenvalue problem has been obtained in a discretized form by solving the complex non-Hermitian matrix eigenvalue problem using a basis of $N=356$ generalized Hylleraas functions, i.e., as

$$\Psi(r_1,r_2,r_{12})=\sum_{i=1}^{N} h_i r_1^{k(i)} r_2^{l(i)} r_{12}^{m(i)} \exp(-\alpha r_1-\beta r_2-\gamma r_{12}) . \tag{6}$$

The expansion coefficients h_i and parameters α, β, and γ are determined variationally for a given state. Coulombic cusp conditions were imposed on the eigenfunctions. The energies and widths of the resonant states ($E_r=E-i\epsilon$) have been obtained as the real and imaginary parts of those complex eigenvalues which are stable with respect to variations in the dilation parameter η. The computational technique is described in Ref. [12].

The resonances seem to cluster just below the $(t\mu)_{2s}$ threshold, and their exact number is unknown. The results for three resonant states that are believed to originate from the 3σ adiabatic potential are presented in Table I. The typical lifetimes are of the order of 10^{-12}–10^{-13} s and are comparable at the fusion time, meaning that a certain fraction of resonant states may fuse.

The fusion rates reported here are calculated according to Jackson's expression [4],

$$\lambda_f=\tfrac{3}{2}A_s|F(0)|^2 = A_s\frac{\int|\Psi(r_{d\mu}=r_{t\mu}=r,r_{dt}=0)|^2 d^3r}{\int|\Psi(r_{d\mu},r_{t\mu},r_{dt})|^2 d\tau} , \tag{7}$$

where A_s is the nuclear reaction constant for d-t reactions, $A_s=1.36\times10^{-14}$ cm^{3}s^{-1}, and $\frac{3}{2}$ is the nuclear spin statistical factor. The integrand in the above equation represents the conditional probability for fusion as a function of muon position, obtained from the three-body wave function at the point of d-t coalescence. The rate of fusion from resonant states appears to be surprisingly large and is comparable to the rate of fusion from the ground state (see Table I).

TABLE I. Present results for the ground state and three resonances of $dt\mu$. First column, energy levels; second column, binding energy with respect to closest threshold. ϵ and τ_d are Coulombic width and lifetime, respectively; λ_f is fusion rate. The notation D^{-12}, for example, means $\times10^{-12}$.

E (a.u.)	E (eV)	ϵ (eV)	τ_d (s)	λ_f (s^{-1})	$\lambda_f\tau_d$
−0.558854	−319.138	···	···	1.36D12	
−0.165182	−217.889	0.001	1.06D−12	0.11D12	0.11
−0.150764	−139.705	0.014	0.46D−13	0.83D12	0.04
−0.139557	−78.937	0.033	0.20D−13	1.88D12	0.04

The total sticking fraction is a sum of transition probabilities between the initial state of the $td\mu$ molecular ion and the bound states of the hydrogenic ion $\alpha\mu$,

$$w_s^0=\sum_{nl} w_{nl} , \tag{8}$$

where w_{nl} refer to contributions of different hydrogenic functions φ_i that describe the final state. $M=15$ hydrogenic functions were used, although the contribution from $n\geq5$ was within 1% of the total. In the sudden impulse approximation the partial sticking fractions are proportional to the overlap between the initial and final wave functions taken at the point of coalescence of d, t, α, and n (neutron),

$$w_{nl}=\sum_m\left|\int d^3r\, R_{nl}^*(r)Y_{lm}^*(\theta,\phi)e^{-iqr}\psi(r)\right|^2 = (2l+1)\left|4\pi\int dr\, r^2 R_{nl}(r)j_l(qr)\psi(r)\right|^2 , \tag{9}$$

where $q=6.0626$ muonic units is the effective recoil momentum obtained by using energy and momentum conservation in the fusion reaction, derived through relativistic considerations (see details in Ref. [13]). $R_{nl}(r)$ and $Y_{lm}(\theta,\phi)$ refer to radial hydrogenic wave functions of $\alpha\mu$ and spherical harmonics, respectively, labeled by principal and orbital quantum numbers n,l. $j_l(qr)$ is the spherical Bessel function. $\psi(r)\equiv\Psi(r_{t\mu}=r_{d\mu}=r,r_{dt}=0)$ is the three-body wave function evaluated at zero internuclear distance and then normalized, which will be called the collapsed wave function.

Values for w_s^0 and contributions w_{nl} are presented in Table II. In order to assess the quality of our solutions we include also the results for the ground state, and compare them with values reported in the literature. We first notice that partial sticking in different excited states has roughly the same distribution for the resonant states and for the ground state. However, the sticking probability for the three resonant states analyzed here is consistently *lower* as compared to sticking from the ground state. In order to understand this feature we have performed an adiabatic analysis of the collapsed wave function $\psi(r)$. The three-body wave function can be written in terms of the general adiabatic expansion as

$$\Psi(r_{dt},r_{d\mu},r_{t\mu})=\sum_n \phi_n(r_{t\mu},r_{d\mu})\chi_n(r_{dt}) \tag{10}$$

and the collapsed wave function takes the form

$$\psi(r)=\sum_n \phi_n(r)\chi_n(0) , \tag{11}$$

TABLE II. Final state distribution of partial contributions to $a\mu$ sticking probability, w_{nl}, and total initial sticking fraction w_s^0 for the ground state and three resonances of $dt\mu$.

w_{nl}	$E\simeq -0.559$	$E_r\simeq -0.165$	$E_r\simeq -0.151$	$E_r\simeq -0.138$
w_{1s}	0.679D−2	0.733D−3	0.905D−3	0.130D−2
w_{1s} [a]	0.683D−2	···	···	···
w_{2s}	0.973D−3	0.114D−3	0.140D−3	0.200D−3
w_{2s} [a]	0.979D−3	···	···	···
w_{2p}	0.236D−3	0.247D−4	0.312D−4	0.463D−4
w_{2p} [a]	0.238D−3	···	···	···
w_{3s}	0.296D−3	0.353D−4	0.432D−4	0.617D−4
w_{3s} [a]	0.297D−3	···	···	···
w_{3p}	0.851D−4	0.911D−5	0.114D−4	0.170D−4
w_{3p} [a]	0.860D−4	···	···	···
w_{4s}	0.126D−3	0.151D−4	0.185D−4	0.264D−4
w_{4s} [a]	0.127D−3	···	···	···
w_s^0	0.879D−2	0.964D−3	0.119D−2	0.171D−2
w_s^0 [a]	0.886D−2	···	···	···

[a]Reference [13].

TABLE III. Analysis of the collapsed wave function in terms of its adiabatic and nonadiabatic components.

	Probability $\lvert\langle\phi_{nl}(r)\rvert\psi(r)\rangle\rvert^2$			
n,l	Bound state	Res. state 1	Res. state 2	Res. state 3
1,0	0.987D+0	0.416D+0	0.419D+0	0.395D+0
2,0	0.777D−2	0.424D+0	0.455D+0	0.516D+0
3,0	0.137D−2	0.555D−1	0.361D−1	0.638D−2
4,0	0.452D−3	0.714D−2	0.569D−2	0.497D−2
5,0	0.205D−3	0.323D−2	0.260D−2	0.246D−2
6,0	0.111D−3	0.177D−2	0.144D−2	0.138D−2
7,0	0.674D−4	0.108D−2	0.881D−3	0.858D−3
8,0	0.440D−4	0.710D−3	0.581D−3	0.568D−3
9,0	0.304D−4	0.493D−3	0.403D−3	0.396D−3
10,0	0.219D−4	0.356D−3	0.292D−3	0.287D−3
11,0	0.163D−4	0.266D−3	0.218D−3	0.215D−3
12,0	0.125D−4	0.203D−3	0.167D−3	0.165D−3
13,0	0.975D−5	0.159D−3	0.131D−3	0.129D−3
14,0	0.777D−5	0.127D−3	0.104D−3	0.103D−3
15,0	0.630D−5	0.103D−3	0.844D−4	0.830D−4
	Bound state contribution			
	0.997D+0	0.912D+0	0.922D+0	0.929D+0
	Continuum			
	0.287D−2	0.881D−1	0.776D−1	0.709D−1

where $\chi_n(0)$ can be interpreted as the expansion coefficients of the collapsed wave function $\psi(r)$ into hydrogeniclike functions of the united ^{5}He atom: $\chi_n\equiv c_n=\langle\phi_n(r)|\psi(r)\rangle$, $\phi_n=R_{n0}(r)Y_{00}(\theta,\varphi)$. The coefficients of such expansion are displayed in Table III.

The manifold $\{\phi_n\}_1^M$ is quantitatively and qualitatively different from the original Hylleraas basis ($M<N$ and the nonlinear exponents are different). To make the expansion complete we introduce a remainder $\phi_c=\psi-\sum c_n\times\phi_n$, which in view of the bound character of functions ϕ_n is interpreted as the L^2 approximation to the continuous part of the collapsed wave function.

We call the collapsed wave function $\psi(r)$ adiabatic if all $c_n=\chi_n(0)$ are 0 except one. The collapsed wave function for the bound state is almost adiabatic ($\psi\simeq\phi_1$ in 99%, see the first column of Table III). On the basis of the *global* properties of resonances (such as energy) one could expect their wave functions to be adiabatic of 3σ character, and we therefore refer to ϕ_3 as the adiabatic component of their collapsed wave functions. Inspecting Table III one immediately notices the presence of large nonadiabatic components in the resonant states. Their collapsed wave functions appear to be mixtures of the $1s$, $2s$, and $3s$ orbitals of ^{5}Heμ, with the third largest component coming from the continuum (see columns 2–4 of Table III).

The presence of large nonadiabatic components in the collapsed wave function leads to *cancellations* in the sticking probability. In order to visualize this, we express the sticking probability as

$$w_s^0=\sum_i w_i^0=\sum_i\left|\langle\varphi_i|e^{-i\mathbf{q}\cdot\mathbf{r}}\left|\sum_n c_n\phi_n\right\rangle\right|^2=\sum_{n,n'}\left\{\sum_i\langle\varphi_i|e^{-i\mathbf{q}\cdot\mathbf{r}}|c_n\phi_n\rangle\langle\varphi_i|e^{-i\mathbf{q}\cdot\mathbf{r}}|c_{n'}\phi_{n'}\rangle^*\right\}\equiv\sum_{n,n'}Q(n,n')\,, \tag{12}$$

where the summation over n includes also the normalized component ϕ_c (c_c being its norm). The matrix $Q(n,n')$ is presented in Table IV. For the ground state, there is only one dominant component $Q(1,1)$ representing sticking from the adiabatic component $1s$ [this is sticking in Born-Oppenheimer (BO) approximation]. The joint effect of other components is reduction of sticking from its BO value to the final sticking of 0.879% (see Table II). We notice that a 1% admixture of nonadiabaticity results in a change of sticking by 24%.

Sticking from resonant states is composed of few dominant diagonal components [notably $Q(1,1)$, $Q(2,2)$, and $Q(c,c)$] and several negative off-diagonal components, which leads to *cancellations*. The collapsed wave function is a *linear combination* of physically realizable states representing the system in its united atom limit, with $|c_n|^2$ giving the probability of finding the system in these states at the instant of fusion. The physical predictions concerning this state will therefore depend not only on the moduli of c_n but also on their relative phases. Hence the presence of large off-diagonal elements can be interpreted as a *quantal interference effect* occurring because of heavy admixture of nonadiabatic components in the collapsed wave function. This results in drastic reduction of sticking, which is transparently seen to depend not only on the nuclear reaction heat, but also on the intramolecu-

TABLE IV. Analysis of interference effects in sticking in terms of the adiabatic and nonadiabatic components of the collapsed wave function. The entries represent contributions to sticking given by matrix $Q(n,n')$ of Eq. (12). Only dominant contributions (i.e., for $n \leq 5$ and $n=c$) are displayed. The off-diagonal elements contain $\{Q(n,n')+Q(n',n)\}/2$. Other resonances show the same interference pattern.

	$E=-0.5588$ (bound state)					
	$1s$	$2s$	$3s$	$4s$	$5s$	ϕ_c
$1s$	0.113D−1	−0.764D−3	−0.177D−3	−0.663D−4	−0.320D−4	−0.184D−2
$2s$	···	0.129D−4	0.596D−5	0.223D−5	0.108D−5	0.621D−4
$3s$	···	···	0.690D−6	0.517D−6	0.250D−6	0.144D−4
$4s$	···	···	···	0.968D−7	0.935D−7	0.539D−5
$5s$	···	···	···	···	0.226D−7	0.260D−5
ϕ_c	···	···	···	···	···	0.749D−4
	$E_r=-0.1652$ (resonant state)					
	$1s$	$2s$	$3s$	$4s$	$5s$	ϕ_c
$1s$	0.478D−2	−0.493D−3	−0.429D−3	−0.153D−3	−0.754D−4	−0.113D−1
$2s$	···	0.702D−3	−0.225D−3	−0.286D−4	−0.124D−4	0.258D−3
$3s$	···	···	0.279D−4	0.116D−4	0.545D−5	0.474D−3
$4s$	···	···	···	0.153D−5	0.147D−5	0.176D−3
$5s$	···	···	···	···	0.355D−6	0.872D−4
ϕ_c	···	···	···	···	···	0.668D−2

lar dynamics of the state leading to fusion.

The features might lead to an overall decrease of sticking in the cycle of dt μCF if the conditions favorable for fusion via resonant states could be found. In this respect we note that the formation rate of resonant states is faster than the corresponding rate for the bound state by a factor $(d_{\text{res}}/d_{\text{bound}})^2 \simeq 25$, where d is a characteristic size of the molecular dipole moment. The formation rates might be as high as $\lambda_{dt\mu^*} \simeq 10^{12}\ \text{s}^{-1}$ for resonances in the range of Vesman formation mechanism, formed in collisions of *excited* $(t\mu)_{2s}$ atoms. The fastest deexcitation mechanism of the $(t\mu)_{2s}$ states is radiative deexcitation after admitting Stark mixing ($\lambda_{\text{deex}} \leq 8\times 10^{10}\ \text{s}^{-1}$) but it does not exceed the above estimation of the formation rate.

We suggest that fusion via resonant states might constitute a side path in the main dt μCF cycle. Traces of such a side path being active might perhaps be seen indirectly in the form of an effective sticking that is lower than expected on the basis of the intrinsic ground state sticking alone. Such an effect is qualitatively in line with experimental findings originated at LAMPF [14] and refined at SIN [15], and accommodates the agreement between experimental and theoretical sticking in the $dd\mu$ case. Indeed, we have found that the resonances of $dd\mu$ have lifetimes of the same order of magnitude as in $dt\mu$, but since the fusion rates are 3 orders of magnitude smaller the $dd\mu$ cycle is not expected to be influenced by a resonance side path.

Direct signals of the side path might be found in the form of energetic products of the resonance decay to the dissociative $t\mu+d+2$ keV and $d\mu+t+2$ keV continuum, but it may very well be that sticking measurements based on the cycling rate theory including the resonance formation might be the most accessible way of probing the incidence of the side path.

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