New Mechanism for Resonant $dt\mu$ Formation and Epithermal Effects in Muon-Catalyzed Fusion

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A recent muon-catalysis experiment with a low-density D-T target saw a pronounced transient in the appearance of fusion neutrons. We propose that this transient is due to resonant molecular formation at epithermal energies. This effect is calculated, with inclusion of the contribution of direct (as opposed to dipole) mesomolecule formation. The direct process is the dominant one, but has not previously been considered. The results agree well with the experiment.

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The remarkable ability of a single negative muon to catalyze many d-t fusions is now firmly established, and systematic research is under way to disentangle the intricacies of the catalysis cycle.[1-4] An essential contributor to the rapidity of the cycle is the *resonant molecular-formation* mechanism, in which the colliding $t\mu+d$ form the loosely bound $J=1, v=1$ $dt\mu$ mesomolecular ion, with the energy released going into vibration and rotation of the resulting compound molecule $\{[(dt\mu)d2e]^*$, etc.$\}$. The kinetic energies (and therefore the target temperatures for thermalized $t\mu$ atoms) for which the collisions are resonant are readily determined once the $dt\mu$ binding energy, the hyperfine splittings, and the compound-molecule rovibrational energies are known.

Recently Breunlich *et al.*[2] reported a catalysis experiment at the Swiss Institute for Nuclear Research (SIN) using a low-density (1% liquid H_2) D-T target (with tritium fraction $C_t=0.88$) in which a rapidly decaying transient was seen in the appearance of the 14-MeV neutrons from d-t fusion. In analogy with earlier experimental results on $dd\mu$ fusion,[5] they interpret this transient in terms of a *triplet-quenching hypothesis,*[2] in which (i) the thermalization time of the $t\mu$ atoms is neglected, (ii) the molecular formation rate for triplet $t\mu$ is very large ($\sim 10^9$ s^{-1}) for rather low temperature (30 to 300 K), and (iii) the quenching rate for the triplet $t\mu$ depends strongly on temperature. However, in regard to (i), by using the cross sections calculated by Melezhik, Ponomarev, and Faifman[6] for the scattering of 1-eV ground-state $t\mu$ atoms from d and t, one finds for $C_t=0.88$ an elastic-scattering rate of only $\sim 2\times10^9\phi$ s^{-1} where ϕ is the density in units of liquid H_2 density (4.25×10^{22} atoms cm^{-3}), so that the thermalization time is clearly *not* negligible for $\phi=1\%$. An objection to (ii) comes from considering the energy balance: If we ignore the rotational contributions, the resonance energy for triplet $t\mu$ collisions with D_2 is about 219 meV; for DT, 107 meV. Hence, neither channel can contribute for a $t\mu$ atom thermalized at 30 K. One need not have confidence in the theory of resonant molecular formation[7,8] to subscribe to this conclusion, since it follows directly from the energy-balance equation [see Eq. (7) below]. An objection to (iii) is that so large a temperature dependence is not expected for the triplet-quenching rate, the process being dominated by triton exchange $[t\mu(\uparrow\uparrow)+t(\downarrow)\rightarrow t\mu(\downarrow\uparrow)+t(\uparrow)]$.[9] Breunlich *et al.*[2] point out this disagreement, but an even more serious discrepancy is suggested by data reported earlier for 10% tritium,[10] which has the transient falling off much more rapidly. The triplet-quenching model then implies that the quenching cross section is greater for $t\mu+d$ collisions than for $t\mu+t$ collisions, a situation that clearly contradicts the well-founded idea that t exchange dominates the $t\mu$-t quenching. For these reasons, therefore, we cannot accept the triplet-quenching hypothesis. (Breunlich *et al.*[2] included the caveat that different theoretical explanations of their data should be considered too.)

Instead we believe that the transient in the neutron appearance observed by Breunlich *et al.*[2] is due to a new phenomenon with significant implications: *epithermal molecular formation.*[11] Specifically, we propose that the observed time dependence actually represents the population of $t\mu$ atoms passing through the energy region of rapid (resonant) mesomolecule formation *during thermalization.* Several elements contribute to the argument: (1) The population of energetic ($\sim$1-eV ground-state $t\mu$ atoms grows as the target density ϕ is reduced; (2) thermalization slows as the tritium fraction or the target temperature is increased; (3) the molecular formation rates fall rapidly as the effective temperature of the $t\mu$ kinetic-energy distribution falls below $\sim10^3$ K. Furthermore, (4) the *dipole* formation mechanism, in which the $t\mu+d$ system makes a dipole transition to the loosely bound $J=1, v=1\,dt\mu$ state, is actually less important than *direct* formation, in which $t\mu$ and d approach in a relative p wave and form the bound state with no dipole transition involved.

We now deal with these four points in detail:

(1) The influence of the target density on the population of energetic $t\mu$ atoms comes from the role that

TABLE I. External Auger (λ_e) and radiative (λ_γ) deexcitation rates and the product $\lambda_{el}\tau$ for excited $t\mu$ atoms. λ_e is taken from Ref. 12, λ_{el} from the $d\mu$-d rates of Ref. 13. Units are 10^{11} s^{-1}.

	$n=4$			$n=3$		
ϕ	λ_e	λ_γ	$\lambda_{el}\tau$	λ_e	λ_γ	$\lambda_{el}\tau$
1.0	9.5	0.06	1.9	1.2	0.19	6.7
0.1	0.95	0.06	1.8	0.12	0.19	3.1
0.01	0.10	0.06	1.1	0.01	0.19	0.5

radiation plays in $t\mu$ deexcitation. The relevant rates for $n=4$ and 3 are shown in Table I. (Stark mixing is strong enough for $\phi \geqslant 1\%$ that we do not have to consider the different l levels separately.[14]) Ponomarev[1] has pointed out the importance of elastic scattering of *excited* $t\mu$ atoms to the thermalization process. Adopting the $d\mu+d$ excited-state elastic-scattering rates $\lambda_{el}(n)$ of Menshikov and Ponomarev[12] for $t\mu+t$ (for 1-eV kinetic energy) gives the products of rate by lifetime $\lambda_{el}\tau$ shown in Table I. Since about half the kinetic energy is lost at each collision, a rough measure of the surviving fraction of kinetic energy is given by $(\frac{1}{2})^{\lambda_{el}\tau}$. Thus we see from Table I that the fractional energy surviving the $n=3$ state increases drastically as ϕ is decreased. Although the kinetic energy upon arrival at $n=3$ is not known with any degree of confidence, it has long been thought[14] to be ~ 1 eV (the exact value is not critical to our arguments as long as it is $>> kT$). For definiteness we assume that the population of epithermal ground-state $t\mu$ atoms is Maxwell-distributed, with average energy of 1 eV.

(2) Elastic scattering of (ground-state) $t\mu$ atoms by d and t has been calculated by Melezhik, Ponomarev, and Faifman.[6] A striking result is that the cross section for scattering from t is an order of magnitude smaller than that from d. This implies much faster thermalization for lower tritium fraction, and thus, according to the epithermal hypothesis, a much faster decay of the transient—as observed by Breunlich *et al.*[10] Furthermore, thermalization though the region of $\sim 10^3$ K clearly must take longer as the target temperature is increased, explaining the temperature dependence of the transients.[2,10]

The time evolution of the nonequilibrium $t\mu$ energy-distribution function, $f(E,t)$, is determined (for arbitrary target temperature and composition) by a Monte Carlo simulation of the time-dependent Boltzmann equation.[15] We use the cross sections of Ref. 6 and some less accurate values for $t\mu+t$ scattering above the 0.24-eV inelastic threshold[16] (adjusting the latter to be consistent with the former). As an approximation we separate the calculation of the slowing from that of molecular formation, and ignore the effect of molecular formation on $f(E,t)$. Time $t=0$ corresponds to the arrival of the $t\mu$ atom at its ground $1S$ state. According to the calculations of Markushin[17] the cascade from a free μ^- with 2-keV kinetic energy to the $1S$ state takes only ~ 0.1 ns for $\phi=1\%$. Thus the unexplained *rise* seen in the experiment,[2] which takes ~ 50 ns, is presumably due to the $t=0$ $t\mu$ kinetic-energy distribution falling significantly *above* the energy at which the (epithermal) molecular formation rate is maximum. Hence the low-density muon-catalyzed d-t fusion experiments present an opportunity to gain unprecedented knowledge of the $t\mu$ cascade.

(3) The *dipole* molecular formation rates are calculated following Vinitsky *et al.*[7] and Leon,[8] but including the electron-shielding correction pointed out by Cohen and Martin.[18] As long as the resonances involved (for a given vibrational excitation) are not close to threshold, rather than including all the many different individual resonances, we can to a good approximation sum over all the final molecular rotational states, ignoring the rotational energy differences. Then instead of the many partial-wave terms of Ref. 8, this sum rule gives

$$\sum_{K_f} F(K_i,K_f) \simeq \int d\rho\, y_0(\rho) y_\nu(\rho) \mathscr{E}(\rho) \int d\rho'\, y_0(\rho') y_\nu(\rho') \mathscr{E}(\rho') j_0(\eta k_2(\rho-\rho')), \tag{1}$$

where the y_i are the vibrational wave functions, j_0 is the zero-order spherical Bessel function, k_2 is the $t\mu+D_2$ (DT) relative momentum, $\eta=\frac{1}{2}$ ($\frac{3}{5}$) for D_2 (DT) collisions, and $\mathscr{E}(\rho)$ is the electric field from the spectator nucleus plus electrons.[18] For D_2 collisions we include contributions from $\nu=2$ through 5, for DT $\nu=3$ through 6, using the sum rule for all but the lowest ν value. The results for singlet $t\mu$ atoms are shown in Fig. 1. In general, $\nu=3$ gives the dominant contribution, but for D_2 collisions at low temperature the $\nu=2$ contribution is largest. The weighted sum of these contributions appropriate to $C_t=0.88$ does indeed fall for temperature $< 10^3$ K.

The temperature in Fig. 1 is not that of the target, but rather characterizes the kinetic-energy distribution in the $t\mu+D_2$ (or DT) c.m. system. As one can readily show, when the projectile ($t\mu$) Maxwell distribution (with temperature T_a) is combined with the target molecule (D_2 or DT) Maxwell distribution (T_b), the result in the c.m. system is also a Maxwell distribution, with temperature $T_{c.m.}=(m_a T_b+m_b T_a)/(m_a+m_b)$ (m_a is the projectile mass and m_b the target mass).

(4) The *direct* molecular formation process has not previously been considered. We approximate it by making use of some results of Melezhik, Ponomarev,

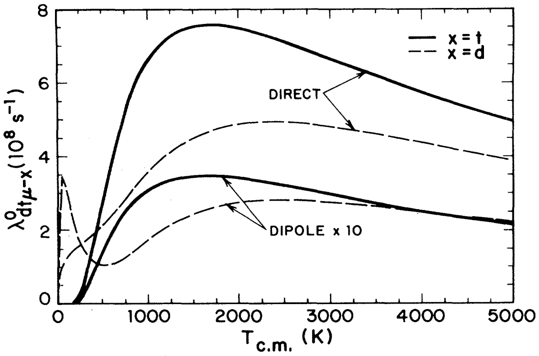

FIG. 1. Epithermal molecular formation rates as functions of c.m. temperature. The upper curves are the direct process, the lower the dipole (× 10), for D_2 (broken curves) and DT (solid curves) collisions (normalized to $C_d = \phi = 1$).

and Faifman[6] and assuming that the p-wave $t\mu + d$ elastic scattering is dominated by the $J=1$, $v=1$ state with binding energy $\epsilon_b \simeq 0.64$ eV.[1] In the present formulation we need to deal with collisions of $t\mu$ both with a deuteron and with the D_2 (or DT) molecule; we follow the convention of using M_1, k_1, and E_1 for the reduced mass, relative wave number, and c.m.-system collision energy of the former, and M_2, k_2, and E_2 for the latter. We write the p-wave elastic-scattering cross section as

$$\sigma_p^{\mathrm{el}} = \frac{12\pi}{k_1^2}\frac{\Gamma_{\mathrm{el}}^2(k_1)/4}{(E_1+\epsilon_b)^2+\Gamma_{\mathrm{el}}^2(k_1)/4}, \tag{2}$$

with the *elastic width* $\Gamma_{\mathrm{el}}(k_1)$ (in electronvolts) being given by

$$\Gamma_{\mathrm{el}}(k_1) = a_3k_1^3 + a_2k_1^2 = 0.65E_1^{3/2} + 0.049E_1 \tag{3}$$

(E_1 expressed in electronvolts). The $a_3k_1^3$ term is due to the bound state at $-\epsilon_b$, and a_3 is chosen to fit the calculated values of Melezhik, Ponomarev, and Faifman,[6] while the $a_2k_1^2$ term comes from the known long-range polarization potential.[19] Equations (2) and (3) describe the results of Ref. 6 extremely well.

We make the jump from elastic $t\mu + d$ scattering to resonant mesomolecule formation in $t\mu + D_2$ (or DT) collisions using the impulse approximation. This method has previously been applied to rovibrational excitation in collisions of atoms with diatomic molecules,[20] a process which is closely related to direct mesomolecule formation. The basic assumption is appropriate for $t\mu + D_2$ collisions even at relatively low energy since the small neutral $t\mu$ atom interacts significantly with a deuteron only at distances small compared to typical internuclear distances of D_2. In the impulse approximation (with approximate treatment of the internal target momentum—see below), the molecular formation cross section can be written as the product

$$\sigma_{\mathrm{direct}}^{\mathrm{mf}} \simeq (M_2/M_1)^2\tilde{\sigma}_p|\mathcal{F}(\nu,K_f;0,K_i)|^2. \tag{4}$$

In this expression

$$\mathcal{F}(\nu,K_f;0,K_i) \equiv \langle \nu,K_f|\exp(i\eta\mathbf{k}_2\cdot\boldsymbol{\rho})|0,K_i\rangle$$

is the molecular bound-state form factor. We define the quantity $\tilde{\sigma}_p$ to be the result of taking the p-wave amplitude off the energy shell; $\tilde{\sigma}_p \rightarrow \sigma_p^{\mathrm{el}}$ as the molecular binding is turned off.

In approximating $\tilde{\sigma}_p$ in Eq. (4) we are guided by the fact that the scattering is strongly dominated by the bound state. Expressing the energy denominator in Eq. (2) in terms of E_2 to describe the molecular collision, we write the final result

$$\sigma_{\mathrm{direct}}^{\mathrm{mf}} \simeq \left(\frac{M_2}{M_1}\right)^4\left(\frac{12\pi}{k_1^2}\right)\frac{\Gamma_{\mathrm{en}}(k_1)\Gamma_{\mathrm{deex}}/4}{(E_2-E_{\mathrm{res}})^2+\Gamma_{\mathrm{tot}}^2/4}. \tag{5}$$

In this expression

$$\Gamma_{\mathrm{en}}(k_1) \equiv F(\nu,K_f;0,K_i)|^2\Gamma_{\mathrm{el}}(k_1)$$

is the "entrance width," Γ_{deex} is the deexcitation rate for all processes from the compound molecule state formed in the collision to states with too little energy for the system to "back decay",[21] and Γ_{tot} is the total width of the compound molecule state. Presumably Γ_{deex} is dominated by Auger deexcitation,[22] but collisional deexcitation may contribute at large ϕ.

We take the internal target motion into account in an approximate way by averaging over the dueteron momentum in the initial molecular state; this gives the relation between E_1 and E_2,

$$E_1(M_1/M_2)E_2+\eta(M_1/m_d)(\tfrac{1}{2}E_0^i + E_{K_i}^i), \tag{6}$$

where m_d is the deuteron mass and $\frac{1}{2}E_0^i + E_{K_i}^i$ is the initial rovibrational kinetic energy. The resonance energy E_{res} is given by the energy balance equation:

$$E_{\mathrm{res}} = E_\nu^f - E_0^i + E_{K_f}^f - E_{K_i}^i - \Delta\epsilon_{\mathrm{hf}} - \epsilon_b. \tag{7}$$

Here E_ν^i (E_ν^f) and $E_{K_i}^i$ ($E_{K_f}^f$) are the initial (final) vibrational and rotational energies, and $\Delta\epsilon_{\mathrm{hf}}$ gives the difference in hyperfine energies in the $t\mu$ and $dt\mu$ states.

Since we expect that $\Gamma_{\mathrm{deex}} \simeq \Gamma_{\mathrm{tot}}$ and $\Gamma_{\mathrm{tot}} << E_2$, the integral over energy, $\int dE_2 f(E_2,t)\sigma_{\mathrm{direct}}^{\mathrm{mf}}(E_2)$, becomes a sum over resonances and the dependence on Γ_{tot} disappears. Furthermore, we can with sufficient accuracy use the sum-rule result, analogous to Eq. (1) but with the electric field factors $\mathcal{E}(\rho)$ missing, for all but the lowest vibrational contribution to the direct process. The same ν values are included as for the dipole mechanism; the dominant contribution is from $\nu = 3$. Considerations of the initial D_2 rotational states are also the same as for the dipole mechanism.[23]

The resultant direct contributions to the singlet D_2

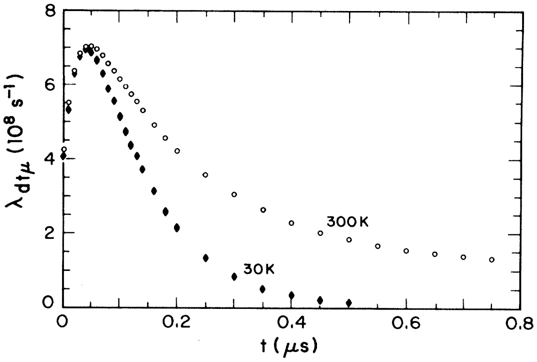

FIG. 2. Time dependence of the molecular formation rate, for $C_t = 0.88$ and $\phi = 1\%$ ($\lambda_{dt\mu}$ normalized to $C_d = \phi = 1$; muon decay not included).

and DT molecular formation rates as functions of $T_{\text{c.m.}}$ are shown in Fig. 1. The direct process is more than 20 times stronger than the dipole, and the weighted sum falls for $T_{\text{c.m.}} < 10^3$ K. (The triplet rates are similar to the singlet ones, allowing us to avoid explicit treatment of triplet contributions and quenching.)

With the above four elements in hand, we combine the energy-distribution function $f(E_2,t)$ with the total molecular-formation cross section $\sigma_{\text{mf}}(E_2)$ to give the time-dependent molecular formation rate $\lambda_{dt\mu}(t)$ shown in Fig. 2, for target temperatures 30 and 300 K and $C_t = 0.88$. As expected, more rapid $t\mu$ energy loss for the 30-K target results in faster decay of the transient, in agreement with the experiment.[2] Furthermore, the time scale of both the rise and decay of the molecular formation rate is in quite good agreement with the experiment. The magnitude of the peak in $\lambda_{dt\mu}(t)$ also agrees fairly well with the values, $(8-9) \times 10^8$ s^{-1}, quoted by Breunlich *et al.*[2] (which they call "triplet").

In addition, on the basis of the present epithermal molecular formation hypothesis, we predict that the *amplitude* of the transient signal will decrease as the target density is increased (see Table I). The triplet-quenching hypothesis implies no such dependence.

In conclusion, (i) triplet-quenching hypothesis is probably not a viable explanation of the experimental results; (ii) the epithermal molecular formation hypothesis appears to be in quite good agreement with experiment and with theoretical expectations; (iii) observation of the density dependence of the amplitude of the transient in neutron production would provide additional evidence for choosing between the two hypotheses[24]; (iv) direct mesomolecule formation is more important than the dipole mechanism; and (v) the detailed time dependence of the transient offers us valuable and hitherto unavailable information on the $t\mu$ cascade and collision processes.

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[1] L. I. Ponomarev, Atomkernenerg. Kerntech. **43**, 175 (1983).

[2] W. H. Breunlich *et al.*, Phys. Rev. Lett. **53**, 1137 (1984).

[3] S. E. Jones *et al.*, Phys. Rev. Lett. **51**, 1757 (1983), and in *Proceedings of the Ninth International Conference on Atomic Physics, Seattle, Washington, 1984,* edited by R. S. Van Dyck, Jr., *et al.* (World Scientific, Singapore, 1985).

[4] V. M. Bystritsky *et al.*, Zh. Eskp. Teor. Fiz. **80**, 1700 (1981) [Sov. Phys. JETP **53**, 877 (1981)].

[5] P. Kammel *et al.*, Phys. Rev. A **28**, 2611 (1983).

[6] V. S. Melezhik, L. I. Ponomarev, and M. P. Faifman, Zh. Eksp. Teor. Fiz. **85**, 434 (1983) [Sov. Phys. JETP **58**, 254 (1983)].

[7] S. I. Vintisky *et al.*, Zh. Eksp. Teor. Fiz. **74**, 849 (1978) [Sov. Phys. JETP **47**, 444 (1978)].

[8] M. Leon, Phys. Rev. Lett. **52**, 605, 1655 (1984).

[9] A. V. Matveenko and L. I. Ponomarev, Zh. Eskp. Teor. Fiz. **59**, 1593 (1970) [Sov. Phys. JETP **32**, 871 (1971)].

[10] W. H. Breunlich *et al.*, in Proceedings of the Fourth Course of the International School of Physics of Exotic Atoms, Erice, Italy, 31 March–6 April 1984 (to be published).

[11] The possibility of epithermal molecular formation has been raised before, especially by J. Rafelski, in *Exotic Atoms '79,* edited by K. Crowe *et al.* (Plenum, N.Y., 1980), p. 203.

[12] L. I. Menshikov and L. I. Ponomarev, Institute of Atomic Energy, Report No. IAE-4041/12 (to be published).

[13] L. I. Menshikov and L. I. Ponomarev, Institute of Atomic Energy, Report No. IAE-4006/12 (to be published).

[14] M. Leon and H. A. Behte, Phys. Rev. **127**, 636 (1962); M. Leon, Phys. Lett. **35B**, 413 (1971).

[15] K. Koura, J. Chem. Phys. **65**, 3883 (1976).

[16] A. V. Matveenko and L. I. Ponomarev, Zh. Eksp. Teor. Fiz. **58**, 1640 (1970) [Sov. Phys. JETP **31**, 880 (1970)]; A. V. Matveenko *et al.*, Zh. Eksp. Teor. Fiz. **68**, 437 (1975) [Sov. Phys. JETP **41**, 212 (1975)].

[17] V. E. Markushin, Zh. Eksp. Teor. Fiz. **80**, 35 (1981) [Sov. Phys. JETP **53**, 16 (1981)].

[18] J. S. Cohen and R. L. Martin, Phys. Rev. Lett. **53**, 738 (1984).

[19] T. F. O'Malley, L. Spruch, and L. Rosenberg, J. Math. Phys. **2**, 491 (1961).

[20] A. Bogan, Phys. Rev. A **9**, 1230 (1974).

[21] A. M. Lane, Phys. Lett. **98A**, 337 (1983).

[22] L. N. Bogdanova *et al.*, Zh. Eksp. Teor. Fiz. **83**, 1165 (1982) [Sov. Phys. JETP **56**, 931 (1982)].

[23] M. Leon and J. S. Cohen, Phys. Rev. A **31**, 2680 (1985).

[24] Evidence for such density dependence has been seen in the LAMPF *d-t* catalysis data (A. N. Anderson, private communication).