

Time-of-Flight Spectroscopy of Muonic Hydrogen Atoms and Molecules

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Abstract. Studies of muonic hydrogen atoms and molecules have been performed traditionally in bulk targets of gas, liquid or solid. At TRIUMF, Canada's meson facility, we have developed a new type of target system using multilayer thin films of solid hydrogen, which provides a beam of muonic hydrogen atoms in vacuum. Using the time-of-flight of the muonic atoms, the energy-dependent information of muonic reactions are obtained in direct manner. We discuss some unique measurements enabled by the new technique, with emphasis on processes relevant to muon catalyzed fusion.

1 Introduction

Simple atoms such as hydrogen and helium provide effective testing ground for fundamental theories. Studies of simple *exotic atoms* could add useful information, despite considerable challenges, both experimentally and theoretically [1].

In this article, we shall focus on the reactions of muonic hydrogen atoms and molecules, and their energy dependent properties revealed by a newly developed time-of-flight spectroscopy technique [2,3,4,5], with particular emphasis on reactions related to muon catalyzed fusion phenomena.

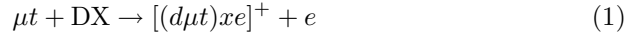
2 Muon Catalyzed Fusion

A muon is a lepton of the second generation with its mass about 207 times heavier than that of electrons, and has a finite lifetime of 2.2 μ s. It is created from the decay of a pion which is obtained, typically, from intermediate energy ($E \geq 500$ MeV) collisions of a proton beam on a target nucleus.

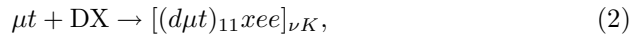
A negative muon can participate in a variety of atomic and molecular processes. A muonic atom is formed when a muon stops in matter replacing an electron. A muonic atom interacting with ordinary atoms or molecules can form a muonic molecule. The latter in turn can result in fusion reactions between the nuclei if the target consists of hydrogen isotopes, a phenomenon known as muon catalyzed fusion (μ CF) [6].

A single muon stopped in a target of deuterium-tritium mixture can catalyze more than 100 fusions, but this number is limited by two major bottle-necks. One is the rate at which a muon can go through the catalysis cycle before its decay (cycling rate), and another is a poisoning process called μ - α sticking in which, with a probability $\omega_s \leq 0.01$, the muon gets captured after the fusion reaction to atomic bound states of the fusion product ${}^4\text{He}$, and hence lost from the cycle (see Section 5).

The first bottle-neck, cycling rate is determined mainly by the rate of formation of muonic molecule $d\mu t$. In fact, a straightforward mechanism for molecular formation via Auger process:



is much too slow, yielding the fusion efficiency of the order of only one fusion per muon. Here, DX refers to either D_2 , DT or HD molecule and x the nucleus p , d or t . A resonant mechanism, however, can give much higher rates when certain conditions are satisfied. In the resonant formation:



the reaction product is a hydrogen-like molecular complex $[(d\mu t)xee]$ in which $(d\mu t)^+$ plays a role of one of the nuclei. The process is resonant, because the energy released upon formation of $d\mu t$ plus the μt kinetic energy must match the ro-vibrational (νK) excitation energy of $[(d\mu t)xee]$ in the final state. This is only possible due to the existence of a state $(J, v) = (1, 1)$ in $d\mu t$ which is bound very loosely (in the muonic scale) with the binding energy smaller than the dissociation energy of deuterium molecules.

Theoretical calculations [7,8,9,10,11] predict strong enhancement of the formation rate $\lambda_{d\mu t}$ at μt energy of order 1 eV (Fig 1), but direct experimental

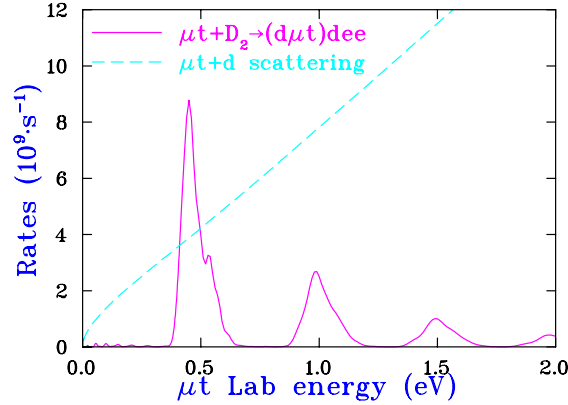


Fig. 1. Resonant molecular formation rate in $\mu t + D_2$ collisions calculated for a 3 K target [7,8,9]. The rates are normalized to the liquid hydrogen density and averaged over μt hyperfine states. Also shown is the μt elastic scattering rate on the d nucleus [12]

information on such epithermal resonance is scarce, let alone its detailed structure. Traditionally, μCF experiments in general, and molecular formation rate measurements in particular, have been performed in a bulk target made of mixture of hydrogen isotopes, in which complex chains of muon induced reactions take place, making the interpretation difficult and model-dependent. In addition, the resonant energies ~ 1 eV is difficult to access with the target thermal energy, since it would require a target of several thousand degrees, a formidable task when working with tritium.

At TRIUMF we have developed a new target system which uses multilayer of solid hydrogen thin films [13,14]. The use of μt beam obtained from the thin film target provides some unique advantages in tackling the epithermal resonances experimentally: (1) formation process can be isolated, to large extent, from the rest of the cycle, (2) epithermal energies are directly accessible due to the available beam energy, and (3) μt time of flight across the drift distance provide information of the resonance energies. Obviously, many technical challenges had to be overcome to make use of the new method.

3 A beam of muonic hydrogen atoms

The basic processes involved in creating a beam of muonic tritium atoms [15] can be categorized into four step [16]: *atomic formation*, *acceleration*, *extraction*, and *moderation*. When a muon is stopped in a thin solid hydrogen target consisting of protium (1H_2) doped with a small amount ($c_t \sim 0.1\%$) of tritium, muonic protium atom (μp) is mostly formed (*atomic formation*). The muon is quickly transferred from proton to triton [17] to form μt , the latter more tightly bound due to the reduced mass difference. In the reaction, the μt gains about 45 eV of recoil kinetic energy, and is thus *accelerated*. The μt then slow down from the col-

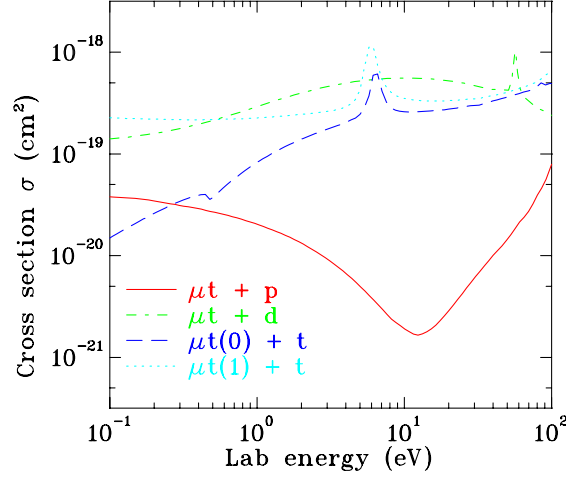


Fig. 2. Scattering cross sections for μt with a hydrogen isotope nuclei from Refs. [12,18], showing the Ramsauer-Townsend minimum at around 10 eV for $\mu t + p$. $\mu t(F) + t$ cross sections plotted include both elastic and spin exchange reactions, where $\mu t(0)$ is the singlet state and $\mu t(1)$ is the triplet state

lisions with the rest of the target nuclei (mostly protons), until it reaches about 10 eV. At these energies, $\mu t + p$ elastic scattering cross section drops dramatically due to the Ramsauer-Townsend effect, making the rest of the target nearly transparent (Fig 2). The μt atom is thus *extracted* from the layer into vacuum. The energy of the emitted μt , which is close to the Ramsauer-Townsend minimum of ~ 10 eV [19], can be controlled to some extent by placing an additional layer, for example of deuterium, on top of the emission layer (*moderation*). Creation of muonic deuterium (μd) is possible in a similar manner with a deuterium-doped protium target: in fact, the emission of muonic hydrogen atoms was first discovered in this system [21]. See Refs. [22,23,24] for our measurements involving muonic deuterium. Recently, we also observed emission of muonic protium atoms from pure hydrogen layer [25], though emission mechanism is expected to be completely different [26].

4 Resonant formation of muonic molecules $d\mu t$

Figure 3 (a) illustrates the principle of our molecular formation measurements using the atomic beam method [2,3,4,5]. A beam of $5 \times 10^3 \mu^-/\text{s}$ of momentum 27 MeV/c from the TRIUMF M20B channel was degraded in a 51 μm gold target support, and stopped in the upstream (US) emission layer. The μt beam, obtained as described above, was slowed via elastic scattering in a D_2 moderation layer from some 10 eV to near 1 eV to better match the resonance energies. The μt , after a few μs time of flight (TOF), is collided with a reaction layer in downstream (DS), separated by the drift distance of 17.9 mm in vacuum. For-

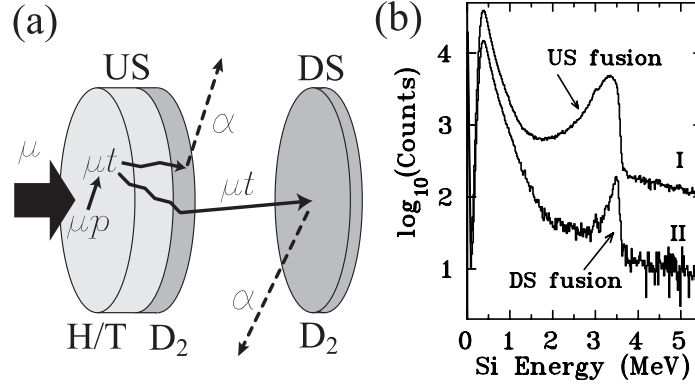


Fig. 3. (a) Schematic of the thin film target for the $d\mu t$ formation measurements consisting of the emission, the moderation and the reaction layers, which are prepared by rapidly freezing hydrogen isotopes on the gold foils (not shown) held in vacuum at 3.5 K. [13] The layer thickness ($3.43 \text{ mg}\cdot\text{cm}^{-2}$, $96 \text{ }\mu\text{g}\cdot\text{cm}^{-2}$, and $21 \text{ }\mu\text{g}\cdot\text{cm}^{-2}$, respectively) were measured off-line via α particle energy loss (see Fig. 5) (b) Measured Si energy spectra with prompt (I: $t > 0.02 \text{ }\mu\text{s}$) and delayed (II: $t > 1.5 \text{ }\mu\text{s}$) time cuts. Fusion in DS reaction layer is separated from that in US D_2 due to the μt TOF across the vacuum

mation of $d\mu t$ molecules is detected by observing 3.5 MeV α particles produced in the fusion reaction, $d + t \rightarrow \alpha + n$, which follows the formation. Si detectors placed in the vacuum enables the measurement α with high energy resolution. The background can be determined accurately by “turning off” the DS fusion reactions using the target without the DS layer. This ability to control a specific process, without affecting the rest, is an advantage of the thin film method. (In conventional targets, changing the target conditions would affect many processes simultaneously).

Because the time between muon stop and the fusion α detection (which we call *fusion time*) is dominated by the μt TOF, it provides a measure on molecular formation energy, as long as the energy loss of μt , due to elastic scattering before the formation in the DS D_2 , is small. Figure 4 illustrates the simulated correlation between the fusion time and the energy at which molecular formation takes place, when there is little μt energy loss. A thin DS layer ($\sim 1 \text{ }\mu\text{m}$) was thus used in the measurement to minimize the energy loss and maximize the time-energy correlation.

A great deal of efforts were made to understand the systematics of this new method. Fig. 5 and Table 1 give some such examples. The detail of the analysis can be found in Refs. [29,30]. The resulting DS fusion time spectrum and its comparison with Monte Carlo (MC) simulations are shown in Fig. 6, which clearly establishes the resonance structure. From the time-of-flight analysis of 2036 ± 116 DS fusion events, a formation rate consistent with $0.73 \pm (0.16)_{\text{meas}} \pm (0.09)_{\text{model}}$ times the theoretical prediction of Faifman *et al.* [9] was obtained (the first error

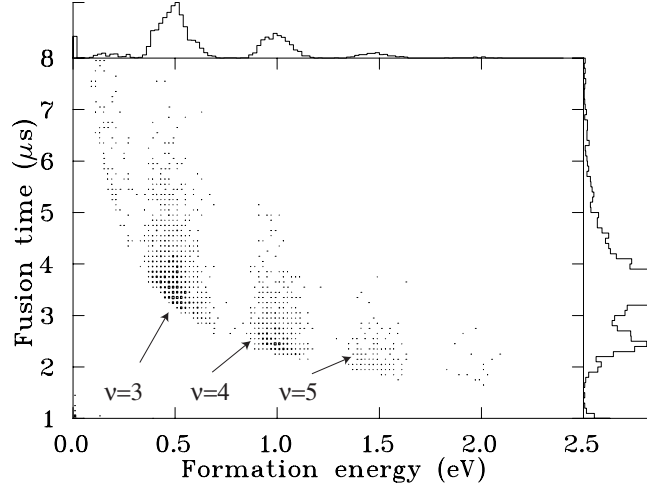


Fig. 4. Simulated correlations between the fusion time and the energy at which molecular formation takes place when there is no μt energy loss in DS layer. Corresponding vibrational excitation in the $[(d\mu t)dee]$ system are also shown

refers to the measurement uncertainty including the statistics and the second is that in MC modeling). The resonance energies were determined from the fit to be $0.940 \pm (0.036)_{meas} \pm (0.080)_{model}$ times the theoretical value [9]. Thus, for the first time, the existence of epithermal resonances in $d\mu t$ molecular formation was directly confirmed, and their energies measured. For the largest peak at the resonance energy of 0.423 ± 0.037 eV, our results correspond to the peak rate of $(7.1 \pm 1.8) \times 10^9 \text{ s}^{-1}$. This is more than an order of magnitude larger than the rates at lower energies, experimentally demonstrating the prospect for high

Table 1. Estimated effects on the formation rate scaling parameter S_λ by the systematic uncertainties in the MC modelling

MC error source	$\Delta S_\lambda / S_\lambda$ %
μ beam size	1.2
Nonuniform μ stopping (GEANT)	1.8
μt TOF drift distance	2.6
$\mu p \rightarrow \mu t$ transfer, $p\mu p$ formation	5.6
$\mu t + d$, $\mu t + t$ scattering, layer thickness	5.7
$\mu t + p$ RT minimum energy	1.1
Resonance Doppler widths in solid	8.0
Solid state and low energy processes	5.0
(subthreshold resonances, slow thermalization, μt energy after resonant scattering)	
Total (in quadrature)	12.9

cycling μCF in a high temperature target of several thousand degrees. If one assumes the energy levels of the $[(d\mu t)\text{dee}]$ molecule, which have a similar structure to those of ordinary hydrogen, our results imply sensitivity to the binding of energy of the loosely bound $(d\mu t)_{11}$ state with an accuracy comparable to the vacuum polarization and other QED corrections, opening up a new possibility of precision spectroscopy in a quantum few body system.

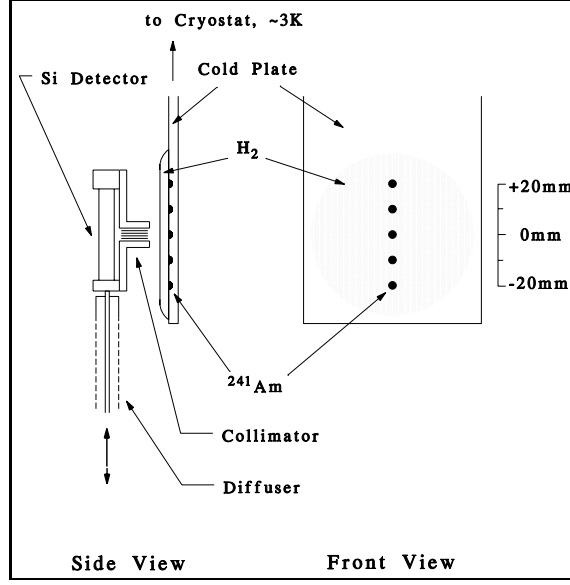


Fig. 5. Calibration measurement of solid hydrogen film layer thickness via energy loss of α particles [27,28]. Americium α source is embedded on the surface of the gold-plated copper substrate, onto which hydrogen thin film is deposited by releasing the gas through porous sintered metal (diffuser). Silicon detector, mounted on the vertically movable diffuser, measured the α particle energy loss in the film, which is converted the thickness using the stopping power

Recently μCF using a triple isotope mixture (H/D/T) target has drawn considerable interest [31], particularly because of resonances in the $d\mu t$ formation in the $\mu t + \text{HD}$ collisions, predicted to be even stronger than the $\mu t + \text{D}_2$ case. Our thin film target allows us to create μt collision with pure HD molecules, and time-of-flight method described here should give a direct test of the theoretical prediction. The data for the $\mu t + \text{HD}$ collision have been collected, and the result will be reported in our future publication [32].

5 μ - α sticking: yet another bottle-neck

Although our measurements of epithermal resonant formation indicates molecular formation may no longer be a bottle-neck at appropriate conditions, the

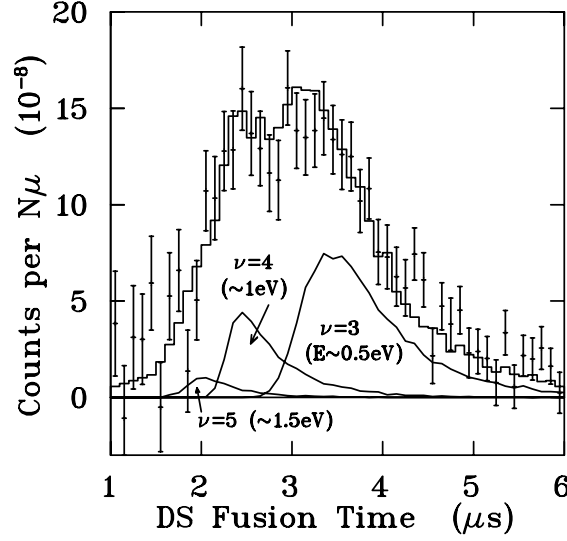


Fig. 6. Time-of-flight fusion spectrum (error bars) and simulation spectrum (histogram), normalized the number of incident muons N_μ . Also plotted are simulated contributions from different resonance peaks given by the time-energy correlated events

process of μ - α sticking still gives a stringent limit, independent of the cycling rate, on the number of fusions catalyzed by one muon. Intense efforts have been made for nearly two decades to understand this key process, but discrepancies persist between and theory and experiment (including latest PSI [33] and RIKEN-RAL [34] results), the experiment being systematically lower than the theory [35,36].

Theoretical calculations of sticking are challenging, due to the interplay of the Coulomb and strong interactions in a non-adiabatic few-body system, yet recent predictions, including the effects of nuclear structure and the deviations from the standard sudden approximation, now converge to a few percent [36]. They cannot, however, be readily compared to experiment because most measurements are primarily sensitive to *final sticking* (ω_s^{fin}), which is a combination of *initial sticking* (ω_s^0), the intrinsic branching ratio for $d\mu t \rightarrow \mu\alpha + n$, and *stripping* (R), collisional reactivation of the muon from $\mu\alpha$ in the target medium (*i.e.*, $\omega_s^{fin} \equiv \omega_s^0(1 - R)$).

Discrepancies in the experimental sticking results has been in part attributed [33] to the difficulties associated with the conventional neutron method, which include a high model dependence and the need for the absolute neutron calibration. A recent RIKEN-RAL experiment [34] has directly observed, with impressive statistics, X-rays from $\mu\alpha$ deexcitation following the muon sticking to excited states of $\mu\alpha$, taking advantage of the new intense pulsed muon beam which enabled the large signal enhancement, but unfortunately the determina-

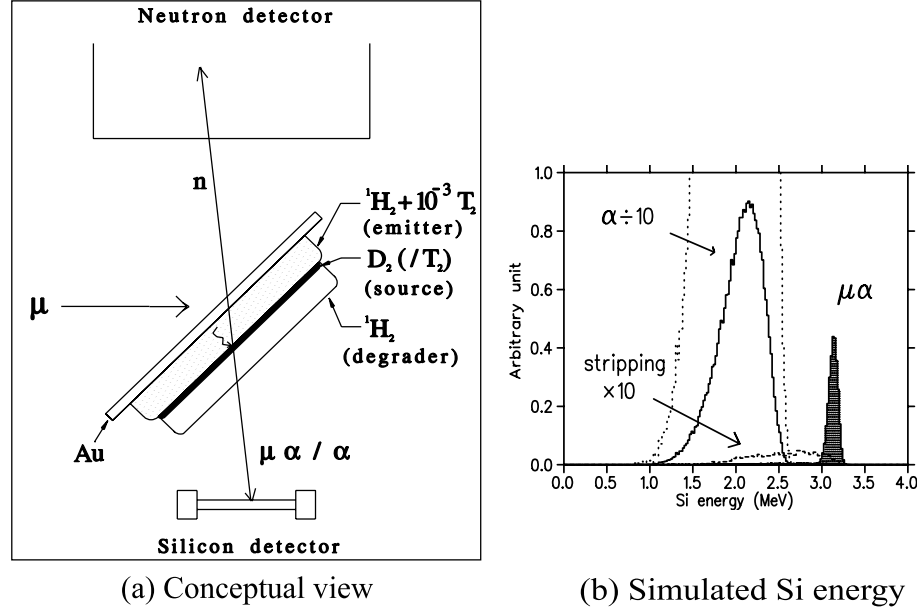


Fig. 7. Proposed direct measurement of $\mu\alpha$ sticking [37,38]. (a) The μt created in the *emitter* is stopped in the *source*, where fusion takes place producing α^{++} or $(\mu\alpha)^+$. The *degrader* separates the two species by their stopping powers (recall that $dE/dx \propto Z^2$). Collinear coincidence of the neutron with the charged events suppresses the background. (b) Simulated Si spectrum for the coincidence events, showing a clear separation between α and $\mu\alpha$ peaks

tion of the sticking probability has to rely on the models of $\mu\alpha$ cascade and stripping, which are yet largely untested.

We have proposed a new direct experiment of sticking using our multi-layer thin film target. The method is illustrated in Fig 7, and the details are given in Refs. [37,38]. The determination of sticking from the ratio $\mu\alpha/(\mu\alpha + \alpha)$ is simple and model independent. Since the stripping in the degrader is small, the measurement is sensitive to the initial sticking, while the stripping itself process can be systematically studied by varying the degrader thickness. Thus experimental separation of initial sticking and stripping will become possible for the first time.

6 Future prospects

Currently there is growing interest in the high energy physics community worldwide in developing a muon storage ring for a neutrino factory and/or for a muon collider [39]. Low energy muon science, including muonic atom and molecule

studies would greatly benefit, if these facilities are realized. Some of the experiments with solid thin films targets are presently limited by the statistics, due to small fraction of muon stops in the target layer, and increased background from muon stopping in the target support and the vacuum chamber [40]. We can expect substantial improvements in the accuracy, as well as entirely new types of experiments with the advent of high intensity muon sources.

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