## Experimental Study of Muon-Catalyzed Fusion in Low-Density Deuterium-Tritium Gas

W. H. Breunlich, M. Cargnelli, P. Kammel, J. Marton, P. Pawlek, J. Werner, and J. Zmeskal Osterreichische Akademie der Wissenschaften, A-1090 Vienna, Austria

and

K. M. Crowe and J. Kurck Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

and

A. Janett and C. Petitjean Schweizerisches Institut für Nuklearforschung, CH-5234 Villigen, Switzerland

and

R. H. Sherman

Material Science and Technology, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

and

H. Bossy and W. Neumann

Physik Department, Technische Universitat München, D-8046 Garching, West Germany

(Received 12 June 1984)

High rates for mesomolecular processes were found in a study of neutron spectra from muon-catalyzed dt fusion in low-density D/T mixtures. An interpretation is given in terms of a reaction-kinetics model which includes hyperfine effects. The hyperfine components of the  $d\mu t$  formation rates, first separated in this experiment, are large from 30 to 300 K. An unexpected temperature dependence for the transition rate between  $\mu t$  hyperfine states is found.

PACS numbers: 36.10.Dr, 25.30.-c

The fascinating idea of muon-catalyzed fusion was advanced more than 35 years ago, but gained new interest only recently, when substantial progress was achieved in our understanding of mesomolecular processes in pure deuterium and deuterium/tritium mixtures. A resonance mechanism first observed in pure deuterium<sup>1-4</sup> gives rise to high formation rates of muonic  $d\mu d$  and  $d\mu t$ molecules, so that a single muon can catalyze numerous fusions in deuterium-tritium mixtures (see Gershtein *et al.*<sup>5</sup> for details). In an early lowdensity, low-tritium-concentration experiment,<sup>6</sup> high molecular formation and isotopic transfer rates were experimentally verified. Later, in an investigation with pressurized high-density targets, high neutron yields and resonant behavior of the molecular formation rates were found.<sup>7</sup> (A recent review is given by Ponomarev.<sup>8</sup>)

We have performed a study of muon-induced fusion in low-density deuterium/tritium gas at the Swiss Institute for Nuclear Research (SIN), measuring the yield and time distribution of d-t

fusion neutrons detected after a muon stopped in the target gas. This was the first investigation of the entire low-temperature region (30-300 K). As a result of the low gas density used (1% of liquid)hydrogen) we were able to observe directly for the first time the extremely fast components of muoninduced reactions, which show surprising behavior as tritium concentration and temperature are varied.

Our goal was to understand the molecular formation mechanism as well as the processes of the d- $\mu$ -t fusion cycle in their full complexity. We were particularly interested in hyperfine (hf) effects, because recently a dramatic hyperfine dependence of the molecular formation process has been discovered in pure deuterium.<sup>4</sup> hf effects were neglected in the analyses of the two published experiments about D/T mixtures.<sup>6,7</sup> However, we have pointed out<sup>9</sup> that consideration of hf effects is indispensable not only for a quantitative description of the d- $\mu$ -t kinetics, but also for the interpretation of these experiments in terms of basic processes.

We chose the following target conditions: low

temperature (the narrow thermal energy spread of the initial  $\mu t$  atoms guarantees a high sensitivity to resonant structures in the  $d\mu t$  formation); low target density (as most of the decisive rates are proportional to the gas density, we were able to resolve reaction rates corresponding to lifetimes of 1–10 ns in liquid hydrogen); and a wide range of tritium concentrations (to disentangle the  $d\mu t$  formation rates on D<sub>2</sub> and DT molecules<sup>5</sup>).

The measurements were performed in the  $\mu E4$ beam of the Swiss Institute for Nuclear Research, with a cylindrical gas target of 1% liquid density (volume  $\sim 1000 \text{ cm}^3$ ). The experimental setup was similar to that used in our previous experiments (see Ref. 4 for details). The gas pumping and storage system was situated inside a glovebox in the experimental area. Tritium and deuterium gas were passed through separate Pd filters before being mixed in the target cell. 70 kCi of tritium were used in this experiment.<sup>10</sup> A mass spectrometer was connected directly to the target cell, and gas samples were taken before and after each run to determine the proportions of the different isotopic molecules present. The target cell was surrounded by plastic scintillators, which had an overall efficiency for detecting electrons from muon decay of about 75%. Two neutron detectors with a total efficiency of  $\sim 2\%$  for 14-MeV neutrons, the signature of *d*-*t* fusion, were used.

The electronic muon stop signal (rate  $\sim 10 \text{ kHz}$ ) consisted of 10% stops in the D/T gas and 90% stops in the windows and walls of the target cell, respectively. The lifetime of these latter muons was short ( $\tau_{Ag} \sim 85$  ns), because silver was used for the inner surface and the entrance windows of the target. In typically 10 h about 30000 fusion neutrons were collected for each temperature.

We present here data from runs in which the tritium concentration was high  $(c_t = 88\%)$ . The complexity of mesomolecular processes<sup>8, 9</sup> is drastically reduced in this regime, since most of the muons are captured directly by tritium nuclei. Figure 1 shows data from one neutron detector for various target temperatures. These time spectra of d-t fusion neutrons detected after a muon stop were obtained with an energy cut at 8.5-MeV neutron energy to exclude t-t fusion neutrons, and neutron/gamma discrimination, resulting in a detection efficiency for 14-MeV neutrons of  $(0.38 \pm 0.04)$ %. Capture neutrons due to wall stops were suppressed by a factor of  $\sim 300$  by requiring the detection of a muon decay electron in one of the electron detectors surrounding the target in a delayed time window  $(0.3-3.7 \ \mu s)$  after the fusion neutron. In spite of



FIG. 1. Time spectra of d-t fusion neutrons detected after muon stop ( $\rho = 1\%$  of liquid hydrogen,  $c_t = 88\%$ , bin width 16 ns). Neutron energy threshold of 8.5 MeV and detection of delayed decay electron were required. Data normalized to equal numbers of muon stops; neutron statistics typically 10 000 per spectrum.

the low target density it was possible to obtain clean spectra of fusion neutrons starting immediately at the moment when the muon stops in the target. This is demonstrated by the characteristic shape of the energy spectra of the 14-MeV neutrons. As a further test, we increased the suppression of capture neutrons relative to fusion neutrons by a factor of  $\sim 8$  by requiring a delayed coincidence signal from two electron telescopes instead of a single detector (the rate of accidental events due to lowenergy gamma radiation was significantly lower in the telescopes).

The spectra in Fig. 1 show two distinct components with different decay rates. The amplitudes of the two components are not very sensitive to temperature variations, but the decay rate of the short-lived component increases strongly with increasing temperature. This behavior cannot be understood within the simple model used in previous works,<sup>5-7</sup> which provides only one time constant at high  $c_t$ .

To interpret our data we use an extended model of the muon-induced fusion kinetics, which includes hyperfine effects both in the molecular formation and in the various hyperfine transitions (see Fig. 2; for more details see also Ref. 9). In agreement with recent theoretical<sup>8</sup> estimates, the thermalization time of the muonic  $\mu t$  atoms was neglected. According to Ref. 9 the hyperfine components  $\lambda_{dt\mu}^F$  of the molecular  $d\mu t$  formation rate are expected to have quite different resonant behavior (*F* is the total spin of the  $\mu t$  atom). The observed rates are a combination of the  $d\mu t$  formation rates in collisions of  $\mu t$  atoms with DT and D<sub>2</sub> molecules, respectively.<sup>9</sup> The hyperfine transition can be induced by the processes

$$\mu t (F=1) + t \rightarrow \mu t (F=0) + t \quad (\text{rate } \lambda_t^{\mu t}),$$

$$\mu t (F=1) + d \rightarrow \mu t (F=0) + d \quad (\text{rate } \lambda_t^{\mu t})$$
(1)

leading to an overall hyperfine transition rate

$$\lambda_{\rm hf} = c_t \lambda_t^{\mu t} \to c_d \lambda_d^{\mu t}. \tag{2}$$



FIG. 2. Extended model of the kinetics of muon-catalyzed fusion including hyperfine effects.  $c_d$  and  $c_t$ denote atomic concentration of deuterium and tritium, respectively. All rates normalized to liquid hydrogen density,  $\rho_0 = 4.2 \times 10^{22}$  cm<sup>-3</sup>. Details of the  $d\mu d$  and  $t\mu t$ fusion branches are omitted for clarity.

A high, temperature-independent value for  $\lambda_t^{\mu t} = 9 \times 10^8 \text{ s}^{-1}$  is predicted theoretically<sup>11</sup>; no experimental results yet exist. The rate  $\lambda_d^{\mu t}$  is usually neglected.<sup>12</sup>

This model adequately describes the observed data, as can be seen in Fig. 3, where we have fitted the 300- and 30-K data with the exact solution of the reaction sequence of Fig. 2. The buildup effect seen in the first 50 ns, which is interesting in itself, was included by a rate  $\lambda_a$ . These fits (Fig. 3 and Table I) indicate a surprising behavior of the observed rates.

For the hyperfine components  $\lambda_{dt\mu}^F$  of the  $d\mu t$ formation rate, first isolated in this experiment, high rates are found over the entire temperature range studied, with no dramatic temperature dependence. The formation rate  $\lambda_{dt\mu}^1$  from the upper hyperfine state of the  $\mu t$  atom, the highest mesomolecular formation rate yet observed, approaches  $10^9 \text{ s}^{-1}$ . This experimental separation of the hyperfine components  $\lambda_{dt\mu}^F$  provides a stringent test of the theoretical understanding of the resonance mechanism for  $d\mu t$  formation. While recently calculated values for the rates<sup>13</sup> are in reasonable agreement with the data of Ref. 7, they do not agree even qualitatively with the more detailed information obtained in the present experiment.

For the hyperfine transition rate  $\lambda_{hf}$  an unexpected temperature behavior is found. At 300 K we get



FIG. 3. Fit of time spectra of fusion neutrons with generalized model of reaction kinetics ( $\rho = 1\%$ ,  $c_t = 88\%$ , temperature 300 and 30 K). The different contributions to the fusion neutron yield (solid curve), which in terms of the model result from molecular formation from the F = 1 and  $F = 0 \ \mu t$  states, are indicated by the dashed curves.

TABLE I. Fit results using extended kinetic model including hyperfine effects. (Fit range 0.048-4.0  $\mu$ s, 239 data points). The errors given in parentheses include contributions from fit and absolute normalization, and correspond to one standard deviation. Important fixed parameters (all rates normalized to liquid density):  $\lambda_{dt} = 2 \times 10^8 \text{ s}^{-1}$ ,  $\lambda_{\mu\mu} = 3 \times 10^6 \text{ s}^{-1}$ ,  $\omega_s = 0.01$ .

Temperature (K)	Rates $(10^6 \text{ s}^{-1})$			
	$\lambda^0_{dt\mu}$	$\lambda^1_{dt\mu}$	$\lambda_{hf}$	Events
30	30(3)	834(90)	642(27)	9700
300	45(4)	891(100)	317(13)	11 000

an upper limit

$$\lambda_t^{\mu t} \le \lambda_{\rm hf} / c_t = (360 \pm 15) \times 10^6 \, {\rm s}^{-1}, \tag{3}$$

which is about one third of the theoretically predicted value.<sup>11</sup> [This limit is deduced from Eq. (2). Making no assumption about  $\lambda_d^{\mu t}$  gives rise to the inequality.] Thus, the theoretical description<sup>11</sup> disagrees with our observed rates, both in magnitude and in temperature dependence. Refined theoretical models, including the possibility of a resonant mechanism for hyperfine transitions of  $\mu t$ atoms,<sup>14</sup> have to be considered now.

Thus, the first experimental observation of strongly different components in the time spectra of *d*-*t* fusion neutrons at high tritium concentrations is an interesting discovery, which demonstrates the complexity of the muon-induced fusion cycle. Our unexpected results prove that low-density experiments are essential if one is to understand the basic processes of muon catalysis. An interpretation of our data is given in terms of a model including hyperfine effects, which is an extension of our model used successfully for pure deuterium.<sup>4</sup> In the D/Tsystem, however, the results obtained are in significant disagreement with theoretical calculations on molecular formation<sup>13</sup> and hyperfine transition<sup>11</sup> rates. Therefore, different theoretical explanations of our data should be considered too.

Finally, we emphasize the preliminary nature of the present analysis, where additional data taken at

low  $c_t$  was not included. Because of the increased complexity of the muon-induced kinetics at low  $c_t$  (see Fig. 2 and Ref. 8) these latter data, which indicate that the neutron time spectra strongly depend on the tritium concentration, require further analysis.

Support by the following institutions is gratefully acknowledged: the Austrian Academy of Sciences, the Austrian Science Foundation, the Swiss Institute for Nuclear Research, the German Federal Ministry for Science and Technology, and the U.S. Department of Energy under Contract No. AT03-81ER40004. We are indebted to Professor Blaser and Professor Lintner for their continuous support and encouragement. We especially thank the Swiss Institute for Nuclear Research technical staff for their expert assistance.

<sup>1</sup>S. I. Vinitskii *et al.*, Zh. Eksp. Teor. Fiz. **74**, 849 (1978) [Sov. Phys. JETP **47**, 444 (1978)].

<sup>2</sup>V. M. Bystritskii *et al.*, Zh. Eksp. Teor. Fiz. **76**, 460 (1979) [Sov. Phys. JETP **49**, 232 (1979)].

<sup>3</sup>P. Kammel et al., Phys. Lett. 112B, 319 (1982).

<sup>4</sup>P. Kammel et al., Phys. Rev. A 28, 2611 (1983).

<sup>5</sup>S. S. Gershtein *et al.*, Zh. Eksp. Teor. Fiz. **78**, 2099 (1980) [Sov. Phys. JETP **51**, 1053 (1980)].

<sup>6</sup>V. M. Bystritskii *et al.*, Zh. Eksp. Teor. Fiz. **80**, 1700 (1981) [Sov. Phys. JETP **53**, 877 (1981)].

<sup>7</sup>S. E. Jones *et al.*, Phys. Rev. Lett. **51**, 1757 (1983).

<sup>8</sup>L. I. Ponomarev, Atomkernenergie/Kerntechnik **43**, 175 (1983).

<sup>9</sup>P. Kammel *et al.*, Atomkernenergie/Kerntechnik **43**, 195 (1983).

<sup>10</sup>The tritium was provided by the U.S. Department of Energy.

<sup>11</sup>A. V. Matveenko *et al.*, Zh. Eksp. Teor. Fiz. **59**, 1593 (1971) [Sov. Phys. JETP **32**, 871 (1971)].

 $^{12}$ Note, however, our recent experimental results for a similar process [W. Bertl *et al.*, Atomkernenergie/Kerntechnik **43**, 184 (1983)] and possible theoretical explanations (Ref. 8).

<sup>13</sup>M. Leon, Phys. Rev. Lett. **52**, 605 (1984).

<sup>14</sup>Compare the theoretical explanation for hyperfine transitions in H/D mixtures mentioned in Ref. 8.