Diffusion of Muonic Deuterium in D₂ Gas

J. B. Kraiman,⁽¹⁾ W. H. Breunlich,⁽²⁾ M. Cargnelli,⁽²⁾ G. Chen,⁽¹⁾ P. P. Guss,⁽¹⁾ F. J. Hartmann,⁽³⁾
P. Kammel,⁽²⁾ J. Marton,⁽²⁾ C. Petitjean,⁽⁴⁾ J. J. Reidy,⁽⁵⁾ R. T. Siegel,⁽¹⁾ W. F. Vulcan,⁽¹⁾ R. E. Welsh,⁽¹⁾ H. Woolverton,⁽⁵⁾ A. Zehnder,⁽⁴⁾ and J. Zmeskal⁽²⁾

⁽¹⁾College of William and Mary, Williamsburg, Virginia 23185

⁽²⁾Institut fuer Mittelenergiephysik, Oesterreichische Akademie der Wissenschaften, A-1020 Vienna, Austria

⁽³⁾Physik-Department, Technische Universität München, D-8046 Garching, West Germany

⁴⁾Paul Scherrer Institute, CH-5232 Villigen, Switzerland

⁽⁵⁾University of Mississippi, Oxford, Mississippi 38667

(Received 1 May 1989)

Diffusion of muonic deuterium (μd) atoms in D₂ at 300 K was studied at pressures of 0.1-10 bars by measuring the distributions of time intervals between entry of negative muons into the gas and detection of photons emitted upon the arrival of μd atoms at foils spaced along the muon beam axis. The results indicate an approximately Maxwellian velocity distribution with a mean energy of 1.8 ± 0.1 eV for the μd atoms, and μd scattering cross sections in agreement with theory.

PACS numbers: 36.10.Dr

Knowledge of the behavior of muonic hydrogen atoms $(\mu p, \mu d, \text{ and } \mu t)$ in matter is important to studies of the weak interactions of muons and the muon-catalyzed fusion of light nuclei. Experimental efforts to establish this behavior were carried out for many years at CERN, 1 along with some related work by others. 2 We have modified the CERN technique and extended it to D_2 pressures lower by 2 orders of magnitude and with reduced statistical uncertainties. Our results differ substantially from earlier work.^{1,2}

The experimental method involved stopping muons in purified D_2 gas filling the regular interstices between multiple parallel planar foils arrayed normal to the incident muon beam (Fig. 1). Scintillation counters detected incident muons, some of which stopped in the gas and formed μd muonic atoms in highly excited states, which then rapidly (\approx ns) (Ref. 3) deexcited to the 1S state. The subsequent motion was determined by the initial velocity distribution of the μd atoms and by the elastic and inelastic scattering of the μd atoms by D₂ molecules. In the case of those μd 's which survived to reach a foil surface, the muons transferred to excited atomic states, bound to nuclei of the foil surface material. During the following processes of atomic deexcitation and nuclear absorption a characteristic "transfer photon spectrum" was emitted. This spectrum included muonic x rays, electronic x rays, and nuclear deexcitation γ rays. The time distributions of such delayed signals relative to the incident muon signals were then accumulated and analyzed.

Our modifications of the CERN experiments¹ included use of (i) a low- (34 MeV/c) momentum beam in the μ E4 area at the Paul Scherrer Institute meson factory to optimize stopping of the μ^{-} in the D₂ gas, (ii) an array of four intrinsic Ge photon detectors providing highenergy resolution to improve photon-line identification and signal-to-noise ratio, and (iii) foils of thin (9 μ m) low-vapor-pressure plastic (Kynar, $C_2H_2F_2$) film⁴ coated on both sides with 100 ± 10 Å of Au to minimize background from muon stops in the surface layer material. The fifty foils used were each 10 cm in diameter, and were mounted with $\pm 5\%$ uniformity in spacing. Deuterium pressures of 188, 375, 750, or 1520 mbar were used with 0.23-cm foil spacing, and 188, 375, and 750 mbar with 0.46-cm spacing. The D_2 was at 300 K and was continuously circulated through a Pd purifier during the data runs.

The electronic logic system was a variant of one previ-



FIG. 1. Experimental setup for the diffusion experiment. Plastic scintillators S_1, S_2, S_3 are beam telescope counters. Scintillators V_1, V_2 veto charged particles. The foils are 10 cm in diameter.

ously used in other work.⁵ Standard techniques were used to digitize the times and pulse heights of those signals from the intrinsic Ge detectors which occurred in an interval from 100 ns before to 4 μ s after the incoming muon trigger $S_1S_2S_3$ (see Fig. 1). These trigger signals occurred at a rate of about 16 kHz, reduced to 9 kHz by a "second muon protection" gate. Pulse heights digitized corresponded to energies from 50 to 600 keV in all Ge detectors and also 600 keV-8 MeV in the largest Ge detector (GMX in Fig. 1). A data acquisition system based on a Microvax-II computer provided on-line information and stored events on magnetic tape.

During a preliminary run a study was made of the spectra of photons emitted when the muon transferred to the Au surface layers. It is known that the x-ray spectrum from muons stopping in solid Au targets⁶ is rich in photons from transitions among circular states with principal quantum number n and angular momentum quantum number l=n-1. However, the experiment of Pfeiffer, Springer, and Daniel⁷ showed that the muonic K- and L-series spectra following transfer from H_2 to Ar gas are consistent with an *l* distribution immediately after transfer which is weighted towards low l. Such a distribution will suppress higher circular transitions during the muonic cascade. Indeed, we observed that after transfer from D₂ to Au, muonic x rays from higher circular transitions were present with only 5%-10% yields, and were thus of limited usefulness as indicators of muon transfer. However, the 356-keV nuclear γ ray emitted from ¹⁹⁶Pt with $35\% \pm 5\%$ yield⁸ following muon absorption in Au (mean life ≈ 70 ns) provided transfer signals of good intensity and signal-to-noise ratio ($\approx 4:1$) over the delay-time region studied, which included 40-2000 ns following the incident muon trigger.

The time distribution of the nuclear γ rays used as transfer signals was a disadvantage in that it widened the effective time response for transfer detection beyond the 12-ns (FWHM) instrumental resolution. At the lower pressures used the transfer event rates tended to restrict the width of the minimum statistically useful time bin to 40 ns, so the information loss due to the 70-ns response time was not a serious limitation. Moreover, the spectrum of these γ 's was quite insensitive to the largely unknown details of the process whereby the muons were transferred from μd atoms to the gold surface layer. (For example, after transfer the initial population of states in muonic Au may depend on the velocity of the incident μd , which could affect the subsequent muonic x-ray spectra.) Since $\approx 95\%$ of the muons would in any event reach the muonic 1S state,⁹ any variation in the cascade process had little effect on the subsequent nuclear absorption of the muons in ^{197}Au .

Several tests were performed on the integrity of the transfer-photon data. For example, the delayed photon spectra were searched for carbon and fluorine muonic x rays, the presence of which might indicate that some

 μd 's had penetrated the Au and transferred to the plastic foils. No delayed fluorine muonic x rays were seen under any experimental condition. However, some delayed muonic carbon 3-1 and 4-1 x rays (carbon 2-1 was obscured by a background line) were seen during a test run at a D₂ pressure of 7.8 bars, though not at 2.5 bars and lower. It was deduced that the carbon muonic x rays originated in a surface layer of carbon which is about 5-10 Å in thickness and which rapidly forms on metallic Au surfaces exposed to air.¹⁰ If some μd 's were sufficiently slowed before impact by scattering, those μd 's would be unable to penetrate the carbon surface layer and would transfer to it rather than to the underlying Au. All data runs were therefore made below 1.6 bars to avoid such effects.

Interpretation of the time distributions involved a procedure whereby initial assumptions were made concerning the velocity distribution and the scattering processes. The best values of parameters involved in these assumptions were then evaluated by nonlinear least-squares fits to the experimental data, as described below. In the experimental time distributions a complicated interplay occurs between the effects of scattering and those of the initial velocity distribution. This interplay allows ambiguities of interpretation, particularly at high pressure (> 5 bars). The chances for such ambiguities to develop during the analysis were reduced by taking data under seven different pressure-spacing conditions, and then analyzing the data taken under all conditions as a single comprehensive data set.

Since the foil spacings were of the order of the μd scattering mean free path at the pressures used in this experiment, the diffusion approximation was not valid, and it was necessary to follow the history of each μd by a Monte Carlo calculation when developing a theoretical model to test against the data. Such Monte Carlo time distributions were compared to the experimental results using the program MINUIT.¹¹ The values of parameters for several different velocity distributions were varied, as was the value of a single parameter characterizing the scattering cross sections (see below).

The scattering was simulated in the Monte Carlo fitting procedure on the basis of the theoretical calculations of Bubak and Faifman¹² (BF) for the scattering of μd on deuterium *nuclei*. It was assumed (after trying some other forms for the scattering) that the scattering cross sections on *molecules* would be increased over the nuclear cross sections of BF by a factor of 2.1 (2.3) for total center-of-mass kinetic energies greater (less) than E', the "crossover energy," which was a free parameter in the data fit. It is to be expected that at energies above 1 eV the molecular factor approaches 2, but at lower energies other effects may develop. The Monte Carlo fit was then made to the best value of E' as well as to the best value of the mean energy E_m in the Maxwell velocity distribution. Both the choice of a Maxwell distribution and the above assumption about molecular scattering are somewhat arbitrary, but comparison with other trial velocity distributions (Gaussian, δ function, rectangular, etc.) as well as with other simple molecularscattering assumptions did not improve the fits. Thus there were only two physical parameters varied in the final fitting procedure. The Monte Carlo kinematics were appropriate to μd collisions with molecules, including the effects of μd hyperfine splitting and the thermal motion of the target molecules.

Figure 2 shows data sets for two of the experimental conditions along with the best-fit curves, made using MINUIT as described above. Note that both sets show a rising behavior at small times. This early-time behavior is caused by the experimental time response function of the Ge detection system for the Pt nuclear γ rays from μd transfer to Au. That response function was measured by stopping negative muons directly in a pure Au target of 0.52 g/cm² and 4 cm in diameter and registering the time distribution of the response function was then tak-



FIG. 2. Time distributions of delayed Pt nuclear γ rays with foil gaps of 0.23 cm at D₂ pressures of 1520 mbar (solid squares) and 375 mbar (open circles), with the effect of muon decay removed. The experimental data include statistical standard deviations. The solid lines are from the Monte Carlo fit to data from *all* conditions (see text), and were generated using the values for parameters E_m and E' as defined and given in the text. The Monte Carlo curves shown each include 40000 events.

en into account in the fit. During diffusion-data runs the background from direct stops in the Au-coated plastic foils was measured with the target vessel evacuated.

The results of the MINUIT analysis indicate values for μd -atom mean kinetic energy E_m of the (laboratory) Maxwell velocity distribution of $E_m = 1.8 \pm 0.1$ eV, and for the molecular-scattering crossover energy of $E' = 0.32 \pm 0.03$ eV, with a reduced $\chi^2 \approx 1.4$ for 171 degrees of freedom. E_m remained within the quoted limits even when various distributions (see above) which yielded relatively poor fits ($\chi^2 > 2$) were used. These values for E_m and E' were stable against removal from the analysis of the low-pressure and/or early-time data. We note that the CERN group¹ assumed 1 eV for the mean initial μd energy in analyzing their data, which was taken at higher pressure (14 bars).

We discuss first our conclusion that the velocity distribution of the μd atoms is approximately Maxwellian with a mean energy E_m as above. The traditional¹³ view is that the μd (or μp) deexcites from its initial excited state with $n \approx 14$ by a combination of external Auger effect (dominant at high n) and circular muonic x-ray emission at lower $n (\leq 5)$. Neither the Auger effect nor x-ray emission should alter the μd momentum appreciably. Recent theoretical studies¹⁴ predict that the formation energy should be ≈ 0.7 eV if the incident muon impacts a free deuterium atom (a molecule would seem more likely); another prediction¹⁵ is that after formation the μdD molecule has a mean laboratory kinetic energy E_m of 0.22 eV, with a rectangular distribution starting at $E_m = 0$, implying that the μd atom will have $E_m \approx 0.1$ eV after separation from the μdD molecule. An earlier paper¹⁶ predicts $E_m \approx 1.5$ eV and a roughly Maxwell shape. While one might be tempted to favor Ref. 16 because it agrees with our data, the conclusions in that and other theoretical papers are highly dependent on the stopping power of D₂ to negative muons in the keV region, on which there is no direct experimental information. We can conclude only that our observed E_m for the μd atoms is substantially higher than most predictions of the kinetic energy at initial formation in highly excited states. There is also the possibility¹⁷ that the μd gains kinetic energy during deexcitation as a result of Coulomb deexcitation of the μd in collisions with D_2 molecules, although it is uncertain whether such processes are of importance at these pressures.

The CERN group¹ used cross sections for μd scattering on deuterium *nuclei* in analyzing their experiment, and found $(8.0 \pm 2.0) \times 10^{-20}$ cm² for the $\mu d + d \rightarrow \mu d$ +d cross section, assumed independent of energy and hyperfine state. For the experiment reported here, taking into account the fitted values of 2.1 (2.3) for the molecular factor from our data above (below) the c.m. energy E' = 0.3 eV, we obtain agreement with Bubak and Faifman¹² in their prediction of $\mu d + d$ cross sections (for both hyperfine states) varying within the limits $(1.7-2.5) \times 10^{-19}$ cm² for c.m. energies 0.01-10 eV. The use in the data analysis of two separate regions for the molecular factor was suggested by two recent papers.¹⁸ Since we varied the pressure and thus the effects of scattering, further theoretical studies of the $\mu d + D_2$ scattering system, more complete in c.m. energy coverage and including estimates of theoretical uncertainty, would permit a fairly stringent test of the theory to be made by comparison with our experimental results.

We wish to express appreciation to D. Joyce and A. Scrinzi for help in the early stages of the experiment. We are grateful for the support of Professor J. P. Blaser and the staff of the Paul Scherrer Institute, and of the U.S. National Center for Supercomputing Applications. This work was supported in part by the Austrian Science Foundation, the German Bundesministerium für Forschung und Technologie, the Paul Scherrer Institute, and the U.S. National Science Foundation.

¹A. Alberigi-Quaranta *et al.*, Phys. Rev. **177**, 2118 (1969); A. Bertin *et al.*, Nuovo Cimento **72A**, 225 (1982).

²V. M. Bystritski *et al.*, Zh. Eksp. Teor. Fiz. **87**, 384 (1984) [Sov. Phys. JETP **60**, 219 (1984)].

³H. Anderhub et al., Phys. Lett. 101B, 151 (1981).

⁴Manufactured by Pennwalt Co., King of Prussia, PA 19406.

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

⁶F. J. Hartmann *et al.*, Z. Phys. **305**, 189 (1982).

⁷H. J. Pfeiffer, K. Springer, and H. Daniel, Nucl. Phys. A254, 433 (1975).

⁸H. J. Evans, Nucl. Phys. A207, 379 (1973).

⁹C. K. Hargrove *et al.* [Phys. Rev. Lett. **23**, 215 (1969)] found that in muonic ²⁰⁹Bi about $(7 \pm 2)\%$ of the muons cause nuclear excitation resulting in neutron emission during the cascade, and we estimate these radiationless transitions to occur at the 5% level in muonic Au.

¹⁰We are indebted to G. Barth of Surface Science Laboratories for discussions of the carbon layer.

¹¹F. James and M. Roos, MINUIT, CERN Program Library D506.

¹²M. Bubak and M. I. Faifman, Joint Institute for Nuclear Research, Dubna, Report No. E4-87-464, 1987 (unpublished).

¹³V. E. Markushin, Zh. Eksp. Teor. Fiz. **80**, 35 (1981) [Sov. Phys. JETP **53**, 16 (1981)]; E. Borie and M. Leon, Phys. Rev.

A 21, 1460 (1981).

¹⁴J. S. Cohen, Phys. Rev. A 27, 167 (1983).

¹⁵V. V. Balashov et al., Muon Catal. Fusion 2, 105 (1988).

¹⁶G. Ya. Korenman and S. I. Rogovaya, J. Phys. B 13, 641 (1980).

¹⁷L. I. Menshikov, Muon Catal. Fusion 2, 173 (1988).

¹⁸A. Adamczak and V. S. Melezhik, Phys. Lett. A **118**, 181 (1986); Muon Catal. Fusion **2**, 138 (1988).