New Insights in Muon-Catalyzed *dd* Fusion by using Ortho-Para Controlled Solid Deuterium

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For the first time, we observed the dependence of the $dd\mu$ formation rate and the $d\mu$ hyperfine-transition rate on the ortho-para state in muon-catalyzed fusion in the solid D_2 state, and found that the effect is even opposite to a recent theoretical prediction. We also determined the back-decay rate and the hyperfine-transition rate via scattering in solid state by using the ortho-para dependence. A theory to describe properly our experimental result is called for to understand the nature of muon-catalyzed fusion in the solid state.

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In muon-catalyzed fusion (μCF), a muon plays an essential role as a catalyst to enhance the fusion rate through resonantly forming a muonic molecule, such as a $dd\mu$. The $d\mu$ hyperfine-transition process is also important because it is directly related to resonant molecular formation, especially for dd-μCF. Figure 1 shows the resonant formation process in the dd-μCF schematically. The $d\mu$ hyperfine-transition process from $F = 3/2$ to $F = 1/2$ is decomposed into two components, which are a transition by atomic scattering ($\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$) and a transition via molecular formation with a subsequent back decay ($\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$). The first experiment to investigate the hyperfine transition [1–7] observed about a 1.4 times smaller rate than the rate predicted by conventional theories, and concluded that the conventional theories overestimated the hyperfine-transition rate [7]. Voropaev *et al.* [8] evaluated both $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$, but their evaluation totally depended on a theoretical estimation of the temperature dependence. Recently, Lauss *et al.* [9] observed the hyperfine-transition rate in a liquid H/D mixture. There, by using the fact that $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ is proportional to the atomic density of deuterium and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ is proportional to the molecular density of deuterium, they separately determined the two components ($\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$) by changing the H/D mixture ratio.

However, there has been no experiment in solid deuterium. In the case of μCF with a target of mixed hydrogen isotopes (H/D, D/T, H/D/T, and others), the increase in the related processes may make the system more complicated, and sometimes disturbs our understanding of the observed phenomena. On the contrary, we adopted one of the simplest systems, so that the experiment is efficient for understanding new features such as the dependence of the μCF processes on the ortho-para state of deuterium, where the ortho deuterium is a state with a coupled nuclear spin $I = 0, 2$ and a rotational state $J =$ even, and the para deuterium is a state with $I = 1$ and $J =$ odd.

An experiment using a solid deuterium target is also important because it can provide crucial information to solve a long-standing puzzle concerning the resonant formation rate. The formation rate in the solid state was predicted to be negligibly small because the kinetic energy of a muonic deuterium atom was expected to be smaller than the resonant energy. However, recent experiments [10–12] which used solid deuterium targets found that the resonant formation rate is comparable to that in the liquid and gas states. To explain this surprising difference, some theoreticians [13–15] proposed several models concerning the solid-state effects, such as subthreshold resonant formation with simultaneous phonon creation/annihilation, which exists only in the solid state. Because these theories suggested a different ortho-para dependence of the formation rate, we had measured the ortho-para dependence of the fusion proton yield in a previous experiment [16,17]. The result could be explained only by a nonthermalization effect [13], which is caused by a steep decrease in the $d\mu$ scattering cross section at low energy in the solid state. However, fusion

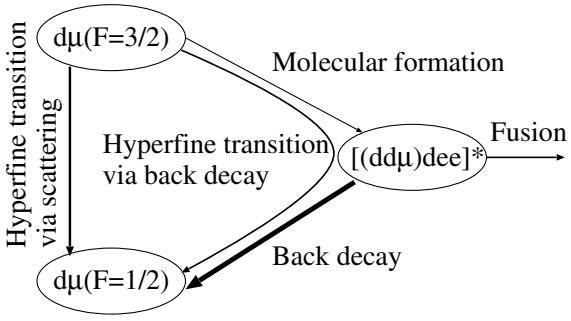

FIG. 1. Diagram for the molecular-formation process and the hyperfine-transition process in dd-μCF.

proton yield depends not only on the resonant formation rate, but also on the $d\mu$ hyperfine-transition rate, both of which were expected to have the ortho-para dependence. The fusion proton yield thus does not reflect the ortho-para dependence of the resonant formation process directly. In order to measure separately the hyperfine-transition rate and the molecular-formation rate, we performed the present measuring fusion proton time spectrum at TRIUMF while taking advantage of the good time resolution. The experiment enabled us to test the present theoretical prediction directly.

Our experiment was conducted at the TRIUMF M9B muon-beam channel. The experimental setup and the target-preparation method were almost the same as those of the previous experiment [16,17]. Figure 2 shows a schematic experimental arrangement around the target. A 32-MeV/c negative muon beam was injected into a thin solid film target (thickness, 280 μm; diameter, 38 mm). To determine the dd-μCF parameters, fusion protons were detected by two pairs of silicon surface barrier detectors, whose total solid angle was 1.68×10^{-1} sr. We also detected the characteristic x rays for various atomic states [μAl M_α, $e(\mathrm{Ag}\mu)K_\alpha$, $e(\mathrm{Ag}\mu)K_\beta$, $e(\mathrm{Ag})K_\alpha$, $e(\mathrm{Ag})K_\beta$, and others] to determine the ratio of the muon-beam yield in the target in to that in the target out by using a Si(Li) detector whose solid angle was 1.28×10^{-2} sr. We took data at 3.5 ± 0.1 K for 66.7%-ortho (normal) deuterium, 99.7 ± 0.4%-ortho deuterium, and an empty to estimate the background level. The ortho-rich deuterium was prepared by flowing deuterium gas into an ortho-para converter filled with a catalyst (Al_2O_3 70% + Cr_2O_3 30%: 14 K). Its ortho-para ratio was measured by observing the thermal conductivity [18].

Based on the kinetics model commonly used for dd-μCF with a dense target [1,2], we analyzed the fusion proton time spectrum with a two-exponential fitting function, which was the same as that for our previous experiment [16,17]. In principle, there should be no ortho-para dependence of parameters summarized in Table I, we fixed these parameters in the fitting procedure. Figure 3 is our fusion proton time spectrum for ortho (99.7%-ortho) deuterium and that for normal (66.7%-ortho) deuterium. It was shown that both the amplitude (N_{fast}) and the slope (λ_{fast}) of a fast component for the ortho deuterium were smaller than those for normal deuterium. By fitting these spectra, we extracted the total molecular-formation rate from a $d\mu$ hyperfine state, $F = 3/2$ ($\tilde{\lambda}_{dd\mu}^{3/2}$), which mainly contributes to N_{fast} and the total hyperfine-transition rate from $F = 3/2$ to $F = 1/2$ ($\tilde{\lambda}_{d\mu}^{3/2\,1/2}$) which mainly contributes to λ_{fast}.

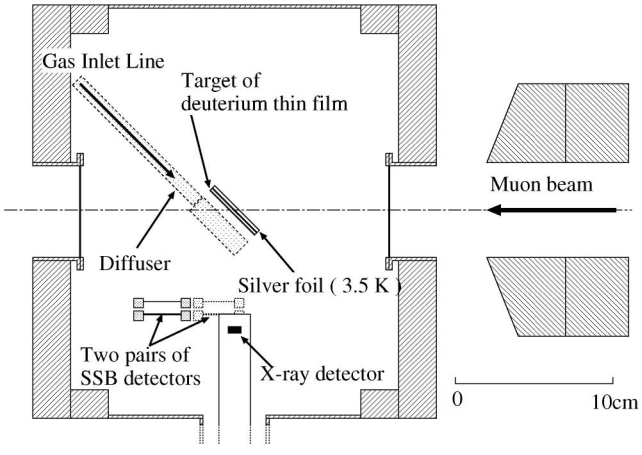

FIG. 2. Schematic view of the experimental arrangement around the target. The diffuser used to spray deuterium gas onto a silver foil is removed before data taking.

Table II gives our results of $\tilde{\lambda}_{dd\mu}^{3/2}$ and $\tilde{\lambda}_{d\mu}^{3/2\,1/2}$ for ortho deuterium and normal deuterium. Because our result for normal deuterium agreed well with the other experimental results [11,12], our fitting procedure was confirmed to be reasonable. The rates $\tilde{\lambda}_{dd\mu}^{3/2}$ and $\tilde{\lambda}_{d\mu}^{3/2\,1/2}$ in ortho deuterium became smaller by 25.7 ± 2.2% and 17.9 ± 2.7%, respectively, compared to those in normal deuterium. A clear ortho-para dependence of the dd-μCF parameter was observed for the first time. Our new data reproduced our previous result [16,17] with much better statistics. This is shown by comparing the parameter $R_p = Y_p^{\mathrm{ortho}}/Y_p^{\mathrm{norm}} - 1$, where Y_p^{ortho} is the fusion proton yield per muon in ortho deuterium , and Y_p^{norm} is that in normal deuterium. In this experiment, the parameter R_p was -0.104 ± 0.046, which agrees well with the previous value of -0.100 ± 0.117 [16,17].

To explain that the experiments [10–12] do not show the steep decrease of the resonant formation rate in the solid state, which was predicted theoretically, several possible theories on solid-state effects, such as the decrease in the scattering cross section (Adamczak [13]), the $d\mu$ acceleration by rotational deexcitation (Gurin [14]), the molecular formation with phonon creation (Menshikov [15]), and others, were proposed. Our result did not show the lack of strong resonant formation from $F = 3/2$ in the ortho deuterium case predicted by Gurin and Menshikov [14,15]. Recently, Adamczak *et al.* tried to explain our result by developing a Monte Carlo simulation code while considering almost all of the expected solid-state effects, such as phonon creation/annihilation

TABLE I. Parameters that were fixed in our fitting of the proton time spectrum. β_F is the branching ratio of the $dd\mu$ fusion process with neutron emission for the $d\mu$ hyperfine state F, and ω_d is the He sticking probability. The formation rate from $F = 1/2$ ($\lambda_{dd\mu}^{1/2}$) is in units of $\mu\mathrm{s}^{-1}$; the density (ϕ) is in units of LHD (liquid hydrogen density).

$\beta_{3/2}$	$\beta_{1/2}$	$\lambda_{dd\mu}^{1/2}$	ω_d	ϕ
0.58	0.53	0.044	0.122	1.43
Expt. [19]	Expt. [20]	Expt. [1–6]	Expt. [19]	Expt. [21]

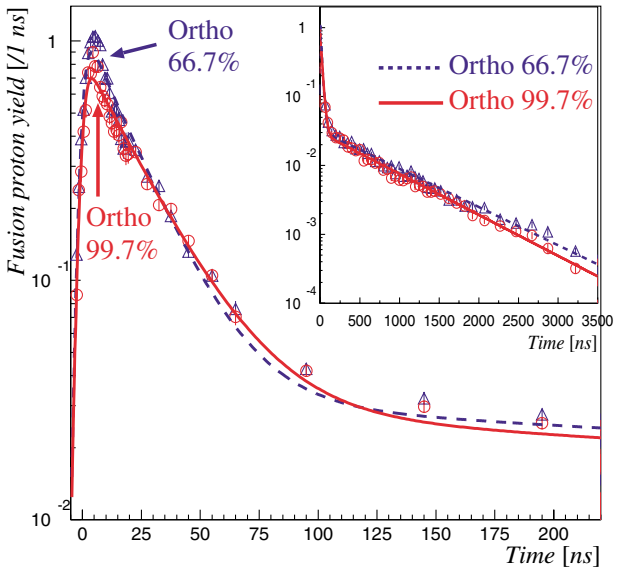

FIG. 3 (color online). Fusion proton time spectrum normalized by taking the amplitude for the slow component being the resonant formation rate from $F = 1/2$ (μs^{-1}) for 66.7%-ortho deuterium (data points, triangle; fitted result, dotted line) and that for 99.7%-ortho deuterium (data points, circle; fitted result, solid line) are plotted. The upper-right inset is for a different time range. The small difference of the slope with a slow component between the normal and ortho deuterium results from an impurity difference.

effects, rotational deexcitation, and other effects [22]. As written in their paper [22], our previous data [16,17] supported their calculation, which predicts a somewhat lower fusion yield for targets with a more ortho deuterium molecule. However, in the present experiment, we confirmed that there was certainly an ortho-para dependence of the resonant formation rate and the $d\mu$ hyperfine-transition rate. To check whether Adamczak's new theory reproduces it, we extracted the rate for the fast component ($\lambda_{\rm fast}$) of the fusion proton time spectrum. In the experiment, the rate $\lambda_{\rm fast}$ for normal deuterium was 52.7 ± 1.1 μs^{-1}, and the rate $\lambda_{\rm fast}$ for ortho deuterium was 43.4 ± 1.0 μs^{-1} ($17.6\% \pm 2.6\%$ decrease). The experimental value of the $\lambda_{\rm fast}$ in ortho deuterium was confirmed to be smaller than that in normal deuterium with good accuracy. However, Adamczak's value of $\lambda_{\rm fast}$ is larger than that in normal deuterium [22], and a more detailed theory that includes the proper solid-state effect is necessary.

Now we discuss our experimental result of the hyperfine-transition rate. By using our data in solid deuterium, the $d\mu$ hyperfine-transition rate due to scattering ($\lambda_{d\mu}^{3/2\,1/2\,\rm scat}$) and the rate of back decay after $dd\mu$ molecular formation ($\lambda_{d\mu}^{3/2\,1/2\,\rm back}$) can be determined in the following way.

The rate $\lambda_{d\mu}^{3/2\,1/2\,\rm back}$ is described as [23]

$$\lambda_{d\mu}^{3/2\,1/2\,\rm back} = \sum_S \lambda_{dd\mu}^{3/2\,S} \frac{\Gamma_{S(1/2)}}{\tilde{\lambda}_f + \sum_{F'} \Gamma_{SF'}}, \qquad (1)$$

where $\tilde{\lambda}_f$ ($\rm ns^{-1}$) is the total fusion rate, $\lambda_{dd\mu}^{FS}$ is the rate of molecular formation from F to the $dd\mu$ spin state S, and Γ_{SF} ($\rm ns^{-1}$) is the rate of back decay from S to F. The observed formation rate, $\tilde{\lambda}_{dd\mu}^{3/2}$, is also written by combining the above parameters and the nonresonant molecular-formation rate ($\lambda_{nr} = 0.044$ [23]) as

$$\tilde{\lambda}_{dd\mu}^{3/2} = \lambda_{nr} + \sum_S \lambda_{dd\mu}^{3/2\,S} \frac{\tilde{\lambda}_f}{\tilde{\lambda}_f + \sum_{F'} \Gamma_{SF'}}. \qquad (2)$$

Then the relation between $\tilde{\lambda}_{dd\mu}^{3/2}$ and $\lambda_{d\mu}^{3/2\,1/2}$ is given by

$$\tilde{\lambda}_{d\mu}^{3/2\,1/2} = \lambda_{d\mu}^{3/2\,1/2\,\rm scat} + \frac{\Gamma_{(1/2)(1/2)}}{\tilde{\lambda}_f} (\tilde{\lambda}_{dd\mu}^{3/2} - \lambda_{nr}), \qquad (3)$$

where we assume that the formation from $F = 3/2$ to $S = 1/2$ is dominant [24]. This assumption is validated as follows. At the lowest temperature, the ground state for the rotational level (J) of a deuterium molecule becomes dominant because the rotational energy is 86 K, even for the first excited state ($J = 1$). Ortho deuterium thus corresponds to the $J = 0$ state, the para deuterium

TABLE II. Our experimental result for the dependence of the total molecular-formation rate from the hyperfine state, $F = 3/2$ ($\tilde{\lambda}_{dd\mu}^{3/2}$), and the total hyperfine-transition rate from $F = 3/2$ to $1/2$ ($\tilde{\lambda}_{d\mu}^{3/2\,1/2}$) on the ortho-para state of deuterium is summarized. The other experimental results for normal (66.7%-ortho) deuterium [11,12] are also summarized for a comparison.

$\tilde{\lambda}_{dd\mu}^{3/2}$ (μs^{-1})	66.7%-ortho D_2	99.7%-ortho D_2
This work at 3.5 K	2.868 ± 0.060	2.131 ± 0.043
Knowles's result [11] at 3 K	$2.71 \pm 0.07 \pm 0.32$	
Demin's result [12] at 5.5 K	2.587 ± 0.079	···
$\tilde{\lambda}_{d\mu}^{3/2\,1/2}$ (μs^{-1})	66.7%-ortho D_2	99.7%-ortho D_2
This work at 3.5 K	36.14 ± 0.84	29.68 ± 0.71
Knowles's result [11] at 3 K	$34.2 \pm 0.8 \pm 0.1$	···
Demin's result [12] at 5.5 K	31.2 ± 1.1	···

TABLE III. Comparison of the $d\mu$ hyperfine-transition rate due to scattering [$\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ (μs^{-1})], the $d\mu$ hyperfine-transition rate via back decay [$\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ (μs^{-1})], and the back-decay rate [$\Gamma_{(1/2)(1/2)}$ (ns^{-1})] between experiments (this work; solid D_2, other experiments [8,9]; liquid D_2) and theories (liquid D_2) [24–26].

	$\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$	$\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$	$\Gamma_{(1/2)(1/2)}$
This expt. (sol.)	11.5 ± 4.2	24.7 ± 4.9	2.8 ± 0.6
Lauss's expt. (liq.) [9]	26.3 ± 3.0	11.0 ± 3.0	0.9 ± 0.3[a]
Voropaev's expt. (liq.) [8]	23.6 ± 0.4	12.9 ± 0.4	···
Theories (liq.)	~ 36 [b]	~ 14 [c]	~ 1.5 [d]

[a]We calculated this rate by combining the experimental data of $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ [9], $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ [9], and $\tilde{\lambda}_f$ [5–7].
[b]Reference [26].
[c]Reference [25].
[d]Reference [24].

corresponds to the $J = 1$ state, and the $J \geq 2$ state almost disappears. The resonant transition from $F = 3/2$ to $S = 3/2$, which needs a deuterium molecule with the $J = 2$ state [23], thus disappears at the lowest temperature.

Because the parameters $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$, $\Gamma_{(1/2)(1/2)}$, $\tilde{\lambda}_f$, and λ_{nr} are basically independent of the deuterium molecular state, such as the ortho-para state, we can determine $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ and $\Gamma_{(1/2)(1/2)}/\tilde{\lambda}_f$ by the observed ortho-para dependence of $\tilde{\lambda}_{dd\mu}^{3/2}$ and $\tilde{\lambda}_{d\mu}^{3/2\,1/2}$. Moreover, $\Gamma_{(1/2)(1/2)}$ is determined experimentally by combining our result of $\Gamma_{(1/2)(1/2)}/\tilde{\lambda}_f$ and the rate $\tilde{\lambda}_f$ provided by other experiments [5–7].

A comparison of $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$, $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$, and $\Gamma_{(1/2)(1/2)}$ between experiments and theories is summarized in Table III. In the liquid state, although the other experiments [8,9] already confirmed that $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ was well reproduced by theory [25], $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ was overestimated by the corresponding theory by Adamczak *et al.* [26].

In the solid state, we experimentally determined $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ for the first time, and these results were found not to be so much different from those in the liquid state. However, a theoretical prediction in the solid state is not available even in the recent theory by Adamczak *et al.* [22]. It is interesting whether each of $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$ and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ is properly reproduced by the theory or not.

In conclusion, we clearly observed the ortho-para dependence of the molecular-formation rate and the hyperfine-transition rate by separately determining these rates with much more statistics. Our experimental data are not reproduced, even by the latest theory. We also separately determined two components of the $d\mu$ hyperfine-transition rate ($\lambda_{d\mu}^{3/2\,1/2\,\mathrm{scat}}$ and $\lambda_{d\mu}^{3/2\,1/2\,\mathrm{back}}$) in solid dd-μCF for the first time. Detailed theoretical prediction in the solid state is called for to understand the solid-state phenomena observed by our experiment for both the resonant formation process and the hyperfine-transition process in the dd-μCF.

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[1] P. Kammel *et al.*, Phys. Lett. **112B**, 319 (1982).
[2] P. Kammel *et al.*, Phys. Rev. A **28**, 2611 (1983).
[3] M. Cargnelli, Ph.D. thesis, Technical University of Vienna, Vienna, 1986.
[4] N. Nägele *et al.*, Nucl. Phys. **A439**, 397 (1989).
[5] J. Zmeskal *et al.*, Phys. Rev. A **42**, 1165 (1990).
[6] J. Zmeskal *et al.*, Muon Catal. Fusion **1**, 109 (1987).
[7] W. Breunlich *et al.*, Muon Catal. Fusion **5/6**, 149 (1990/1991).
[8] N. Voropaev *et al.*, Hyperfine Interact. **118**, 135 (1999).
[9] B. Lauss *et al.*, Hyperfine Interact. **118**, 79 (1999).
[10] G. Marshall *et al.*, Hyperfine Interact. **82**, 529 (1993).
[11] P. Knowles *et al.*, Hyperfine Interact. **101/102**, 21 (1996).
[12] D. Demin *et al.*, Hyperfine Interact. **101/102**, 13 (1996).
[13] A. Adamczak, Hyperfine Interact. **119**, 23 (1999).
[14] G. Gurin and L. Menshikov, Hyperfine Interact. **118**, 147 (1999).
[15] L. Menshikov and V. Filchenkov, Hyperfine Interact. **101/102**, 207 (1996).
[16] A. Toyoda *et al.*, Phys. Lett. B **509**, 30 (2001).
[17] A. Toyoda, Ph.D. thesis, University of Tokyo, Tokyo, 2000.
[18] E. Grilly, Rev. Sci. Instrum. **24**, 72 (1953).
[19] D. Balin *et al.*, Phys. Lett. **141B**, 173 (1984).
[20] C. Petitjean *et al.*, Hyperfine Interact. **118**, 127 (1999).
[21] C. Petitjean *et al.*, Muon Catal. Fusion **2**, 37 (1988).
[22] A. Adamczak and M. Faifman, Phys. Rev. A **64**, 052705 (2001).
[23] A. Scrinzi *et al.*, Phys. Rev. A **47**, 4691 (1993).
[24] L. Menshikov *et al.*, Sov. Phys. JETP **65**, 656 (1987).
[25] M. Faifman *et al.*, Muon Catal. Fusion **2**, 247 (1988).
[26] A. Adamczak *et al.*, At. Data Nucl. Data Tables **62**, 255 (1996); **62**, 315 (1996); **62**, 329 (1996).