Discovery of Temperature-Dependent Phenomena of Muon-Catalyzed Fusion in Solid Deuterium and Tritium Mixtures

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A systematic experimental study on muon-catalyzed fusion was conducted using a series of solid deuterium and tritium mixtures. A variety of conditions were investigated, i.e., tritium concentrations from 20% to 70%, and temperatures from 5 to 16 K. With decreasing temperature, we observed an unexpected decrease in the muon cycling rate (λ_c) and an increase in the muon loss probability (W). The origins of these observed changes were interpreted by the temperature-dependence in the $dt\mu$ formation process for λ_c and that in the muon reactivation process after muon-to-α sticking for W.

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A negative muon catalyzes a nuclear fusion in a deuterium and tritium mixture: $d + t + \mu \rightarrow dt\mu \rightarrow \alpha + n + \mu + 17.5$ MeV. After this reaction, the muon is relieved and induces another fusion reaction. This cycling reaction of muon-catalyzed fusion (μCF) is repeated until the muon is lost from the cycle by either muon decay or muon capture to a higher-Z particle, like the fusion product (α). In the study of μCF, there still exists an unsolved problem of the $dt\mu$ formation mechanism at low temperature below 100 K. It is widely accepted that a $dt\mu$ molecule can be regarded as being resonantly formed in a two-body collision between $t\mu$ and D_2: $t\mu + D_2 \rightarrow [(dt\mu)dee]$ [1]. This resonant formation mechanism has succeeded to explain a high muon cycling rate, which is predominantly determined by the $dt\mu$ formation rate. A theoretical calculation based on this mechanism [2] predicted the $dt\mu$ formation rate to decrease steeply with decreasing temperature below 100 K. This was due to the existence of a threshold energy in $dt\mu$ formation, which lay below 10 meV [3]. In a study prior to the present work [4], we conducted a series of experiments at temperatures of 20 K for a liquid and 16 K for a solid with various tritium concentrations (C_t) from 0.1 to 0.7. Our results concerning the muon cycling rate (λ_c) as well as other experimental results [5,6] are not consistent with the theoretical prediction [2].

There is another discrepancy between the conventional resonant $dt\mu$ formation mechanism and the experimental result. Let us consider the density dependence of the muon cycling rate (λ_c), which is normalized by the target density in order to remove any trivial density dependence. Because the conventional resonant $dt\mu$-formation mechanism has been based on a two-body collision, λ_c is predicted to be constant with the density. However, as shown in Fig. 1, where the abscissa, ϕ, shows the atomic-number density of the D-T target normalized by the liquid hydrogen one, λ_c increases with the density, and this tendency holds good even for liquid and solid data. Thus, the conventional resonant $dt\mu$-formation mechanism can explain neither *the temperature dependence,* in which the muon cycling rate does not decrease below 100 K, nor *the density dependence,* in which the muon cycling rate increases with increasing density.

Several theoretical approaches [8–10] have been provided to solve these problems. The idea of a three-body collision during $dt\mu$ formation was proposed by

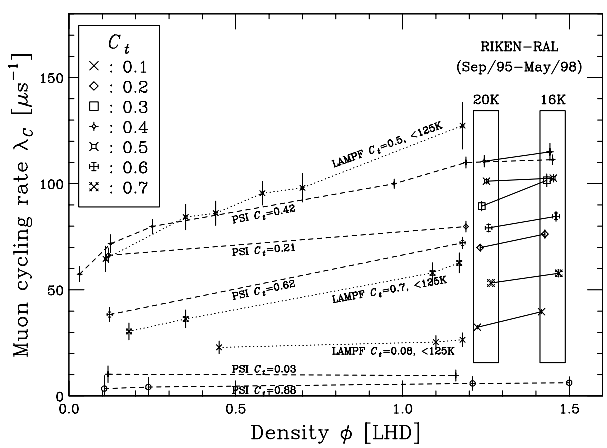

FIG. 1. Target density dependence of the muon cycling rate [4,6,7]. The abscissa shows the atomic-number density of the D-T target normalized by the liquid-hydrogen one (4.25×10^{22} cm^{-3}).

Menshikov *et al.* [8]. They hypothesized molecular formation in a three-body system, $t\mu + D_2 + D_2'$, in which the resonant molecular formation is enhanced compared with that in a two-body system by the third particle, D_2', which takes away the excess energy of molecular formation. This idea allows molecular formation below the above-mentioned threshold energy, which is lower than 10 meV, and thus can explain the temperature dependence. The contribution of the third particle may also explain the density dependence. Faifman introduced a phenomenological three-body effect [11], and pointed out that low-temperature data were able to be explained by the transition energy from $t\mu$ to $dt\mu$ shifting from 0.601 to 0.596 eV. On the other hand, based on the idea of a three-body collision, Fukushima calculated the molecule formation rate in a solid by taking into account the phonon excitation as a condensed-matter effect [12]; also, a sharp increase in λ_c was predicted below about 10 K. Although these theoretical studies were based on the three-body (many-body) effect, their results concerning the temperature dependence at low temperature were different from each other due to the difference in the treatment of this effect.

From the standpoint of experiments, the density dependence had been paid attention. Although some data were provided at low temperatures [4,6], no systematic study was performed with respect to the temperature dependence before. Especially, no study was conducted below 10 K, although the temperature dependence of λ_c was predicted to be conspicuous with decreasing temperature below 10 K [12]. Thus, by investigating the temperature dependence in a solid, new information on the three-body effect in a $dt\mu$ formation was expected to be derived.

There is another unsolved problem concerning the dt-μCF study. A discrepancy between theory and experiment exists concerning the effective muon-to-α sticking probability at high density [4,13,14]. Some theoretical studies treated this problem with an accurate calculation for muon reactivation from an $(\alpha\mu)^+$ ion [13,14]. However, there is still a wide gap. Although the muon reactivation process is supposed to be dominated by high-energy reactions against the dissociation energy of $(\alpha\mu)^+$, there exists further room for investigating both theoretically and experimentally. A temperature-dependence observation is one of the experimental contributions to this problem.

The present study was conducted at the RIKEN-RAL Muon Facility [15]. An advanced tritium gas-handling system, which had an *in situ* ^{3}He-removal capability with a palladium filter [16], was applied to prepare a D-T mixture gas with a tritium concentration from 0.2 to 0.7. Passing through a palladium filter, the D-T gas obtained the chemical equilibrium state. By controlling the cryogenic system, the target cell was adjusted to a temperature from 5 to 16 K within an error of about 0.1 K, and the D-T gas was condensed into the cell. A Rh-Fe thermometer, connected to the coldhead of the cryostat, was adopted to monitor the target temperature; also, the vapor pressure of the target gas was used as a sensitive sensor above 10 K. By utilizing an intense pulsed muon beam, x rays associated with the α-sticking phenomenon were clearly observed, and the α-sticking K_α x-ray yield (Y_X) was determined in the same way as described in Refs. [4,17]. Fusion neutrons were also observed by a system reported separately [18]. In determining the neutron disappearance rate (λ_n) and the fusion neutron yield (Y_n), the effect of target sublimation induced by β-ray heating [19] and the accumulation effect of ^{3}He originating from tritium β decay [20] were taken into account. Sublimation transformed the target form, and this transformation became slower with decreasing temperature. Concerning the effect of ^{3}He accumulation, the muon transfer rate from $t\mu$ to ^{3}He increased by more than twice with decreasing temperature from 16 to 5 K; this was qualitatively consistent with an extrapolation which was smoothly associated with the theoretical prediction above 15 K [21]. Although each of these phenomena attracted our interest, we concentrated on the temperature dependence of more fundamental parameters in the μCF cycle, such as the muon cycling rate (λ_c) and the total muon loss probability (W). The effect of sublimation was taken into account to determine the number of stopping muons in the target. The effect of ^{3}He accumulation was corrected, and was eliminated from the following discussion. Because the thermal conductivity of solid hydrogen was sufficiently good against β-ray heating

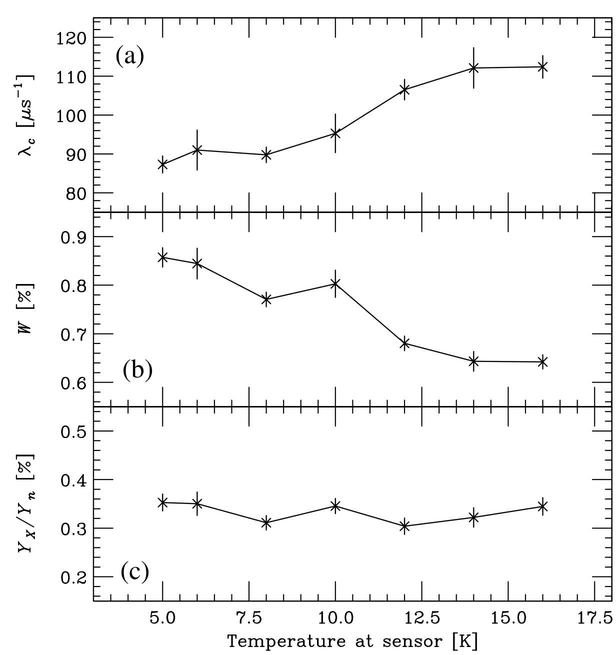

FIG. 2. Temperature dependence of (a) the muon cycling rate (λ_c), (b) the muon loss probability (W), and (c) the ratio of Y_X to Y_n at the tritium concentration of 0.4.

[22], the temperature gradient between the cooler interior of the target cell and the hotter solid-target center, of which typical thickness was a few millimeters, was supposed to be negligible.

The muon cycling rate (λ_c) and the total muon loss probability (W), which includes the effects of α-sticking, dd-μCF and tt-μCF, are associated with two observable quantities, λ_n and Y_n:

$$Y_n = \phi \lambda_c / \lambda_n, \quad (1)$$

$$\lambda_n = \lambda_0 + W \phi \lambda_c, \quad (2)$$

where λ_0 is the muon decay rate (0.455 μs^{-1}).

In the present work, λ_n and Y_n were determined by neutron detection, and Y_X was determined by x-ray detection. The λ_c and W were uniquely determined by λ_n and Y_n using Eqs. (1) and (2). Figure 2 shows the temperature dependence of (a) λ_c, (b) W, and (c) the ratio of Y_X to Y_n, in the case of $C_t = 0.4$. As can be clearly seen in these figures, λ_c and W have some dependence on the temperature, while the ratio of Y_X to Y_n is almost constant with the temperature. A similar temperature dependence was also confirmed at other tritium concentrations. In the following, we examine this result in detail.

The reciprocal of the muon cycling rate is the sum of the muon staying time in $d\mu$ ($\tau_{d\mu}$) and in $t\mu$ ($\tau_{t\mu}$): $1/\lambda_c = \tau_{d\mu} + \tau_{t\mu}$. They are represented by the muon transfer rate from $(d\mu)_{1s}$ to $(t\mu)_{1s}$ (λ_{dt}), and the $dt\mu$ formation rate ($\lambda_{dt\mu}^{F=0} \equiv \lambda_{dt\mu}^{F=0,\,D_2} C_d + \lambda_{dt\mu}^{F=0,\,DT} C_t$) [4]:

$$\tau_{d\mu} = \frac{q_{1s} C_d}{\lambda_{dt} C_t}, \qquad \tau_{t\mu} = \frac{3}{4} \frac{1}{\lambda_{t\mu}^{10} C_t} + \frac{1}{\lambda_{dt\mu}^{F=0} C_d}, \quad (3)$$

where the formation rates of $dd\mu$ and $tt\mu$ are negligibly small in comparison with the $dt\mu$ case, and thus their effects are neglected. At 16 K, the parameters are assumed to take the following values: $q_{1s} = 1/(1 + a_q C_t)$, ($a_q = 6.8$), $\lambda_{dt\mu}^{F=0,\,D_2} = 350\ \mu s^{-1}$, $\lambda_{dt\mu}^{F=0,\,DT} = 160\ \mu s^{-1}$ [4], $\lambda_{dt} = 280\ \mu s^{-1}$ [5], and $\lambda_{t\mu}^{10} = 1300\ \mu s^{-1}$ [23], where q_{1s} is the fraction of $d\mu$ reaching the $1s$ state from the initial Coulomb capture, and $\lambda_{t\mu}^{10}$ is the hyperfine transition rate of $t\mu$ from $F = 1$ to $F = 0$. We consider two possible origins of the temperature dependence: one is the temperature dependence of $\lambda_{dt\mu}^{F=0,\,D_2}$; the other is that of λ_{dt}. The observed change in λ_c is so big that only the rate-determining term, such as $\lambda_{dt\mu}^{F=0,\,D_2}$ or λ_{dt}, can cause this change. In addition, each of them describes a process after $d\mu$ or $t\mu$ thermalizes, and is potentially sensitive to the temperature. It was confirmed only above 20 K that λ_{dt} was almost constant with the temperature [7,24]. Thus, we dealt with the λ_{dt} case as one option.

Comparing these two cases, the C_t dependence of λ_c at 5 K is expected to be different from each other. As can be clearly seen in Fig. 3(a), the case assuming a change in $\lambda_{dt\mu}^{F=0,\,D_2}$ reproduces the experimental result much better than the λ_{dt} case. The best-fitted result was obtained when $\lambda_{dt\mu}^{F=0,\,D_2}$ decreased by about 30% from the value at 16 K, provided that the other parameters were fixed. This result is qualitatively consistent with the phenomenological three-body calculation adopted 0.596 eV as the transition energy from $t\mu$ to $d\mu$ [11], and shows an opposite tendency to a prediction in Ref. [12].

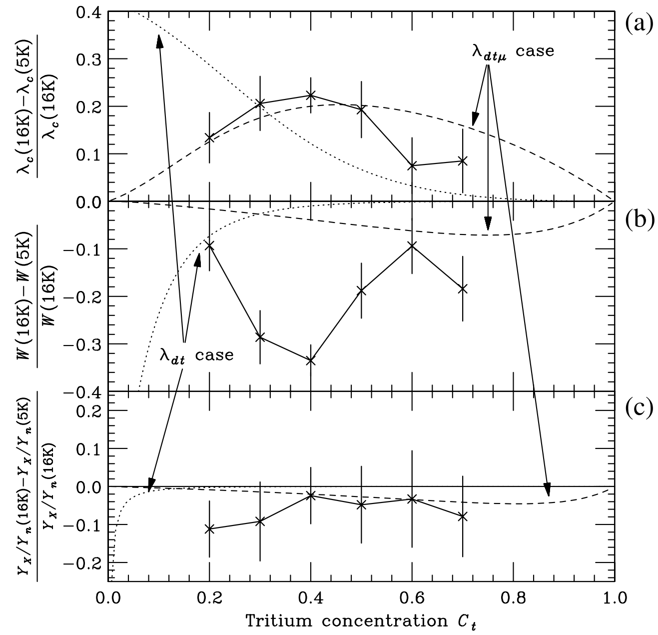

FIG. 3. Tritium concentration dependence of the ratio of change in (a) the muon cycling rate (λ_c), (b) the muon loss probability (W), and (c) the ratio of Y_X to Y_n, which were determined by $[\lambda_c(16\ K) - \lambda_c(5\ K)]/\lambda_c(16\ K)$, and so forth. According to the dashed lines, $\lambda_{dt\mu}^{F=0,\,D_2}$ decreases by about 30% (360 $\mu s^{-1} \rightarrow$ 240 μs^{-1}), and according to the dotted lines, λ_{dt} decreases by about 60% (280 $\mu s^{-1} \rightarrow$ 110 μs^{-1}).

The total muon loss probability (W) is mainly composed of three terms,

$$W = \omega_s^{\rm eff} + W_{dd} + W_{tt}$$
$$= \omega_s^{\rm eff} + \tau_{d\mu} \times \frac{2}{3} C_{D_2} \lambda_{dd\mu}^{3/2} \beta_d \omega_d + \tau_{t\mu} C_t \lambda_{tt\mu} \omega_t. \quad (4)$$

The first term is the effective sticking probability, the second and third terms represent the muon loss due to dd-μCF and tt-μCF, respectively, where $\beta_d = 0.58$ is the branch ratio of ^{3}He + d channel in dd-fusion, and ω_d and ω_t are the muon sticking probabilities to ^{3}He in dd-μCF and ^{4}He in tt-μCF, respectively. As shown in Eq. (3), $\tau_{d\mu}$ and $\tau_{t\mu}$ depend on λ_{dt} and $\lambda_{dt\mu}^{F=0,\,D_2}$, respectively. Thus, a change in λ_c affects W indirectly through $\tau_{d\mu}$ or $\tau_{t\mu}$. In the case assuming a change in $\lambda_{dt\mu}^{F=0,\,D_2}$, the tt-μCF term in W is expected to increase; in the λ_{dt} case, the dd-μCF term increases. However, the expected increase in each case is not sufficient to explain the observed change in W, as shown in Fig. 3(b). At 16 K, the contribution of W_{dd} and W_{tt} to W are estimated to be about 1% and 13% at $C_t = 0.4$, respectively; with increasing C_t, W_{tt} increases and W_{dd} decreases [4]. Thus, this tendency cannot explain the observed profile that the change in W has the maximum at around $C_t = 0.4$. Therefore, the increase in W is expected to be due to an increase in ω_s^{eff}. The muon sticking process is divided into two processes, initial sticking and a subsequent reactivation process, $\omega_s^{\mathrm{eff}} = (1 - R)\omega_s^0$, where R is the reactivation probability and ω_s^0 is the initial sticking probability. Because the initial sticking is a process immediately after a nuclear reaction, it is implied that the reactivation process depends on the temperature.

Concerning the α-sticking process, we obtained other information by x-ray detection. The ratio of Y_X to Y_n gives information about the initial status of an α-sticking $(\alpha\mu)^+$ ion: $Y_X/Y_n = Y(K_\alpha) + Y_{dd}(K_\alpha) + Y_{tt}(K_\alpha)$, where $Y(K_\alpha)$ is the x-ray yield per dt fusion, and is proportional to ω_s^0: $Y(K_\alpha) = \gamma_{K_\alpha}\omega_s^0$. The coefficient is the x-ray yield per initial sticking. The second and the third terms stand for contributions from dd and tt-μCF, respectively: $Y_a(K_\alpha) = \gamma_{K_\alpha}(a)W_a/(1 - R_a)$, ($a = dd, tt$). In each side channel, $\gamma_{K_\alpha}(a)$ and R_a correspond to γ_{K_α} and R in the dt case, respectively. The dependences on C_t of $Y_{dd}(K_\alpha)$ and $Y_{tt}(K_\alpha)$ are similar to W_{dd} and W_{tt}, respectively, and their contributions to Y_X/Y_n are estimated to be about 0.03% and 7%, respectively, at $C_t = 0.4$ and 16 K [4]. Similar to the case of W, $Y_{dd}(K_\alpha)$ and $Y_{tt}(K_\alpha)$ were affected by a change in λ_c; however, their expected changes were negligibly small, as shown in Fig. 3(c). Thus, γ_{K_α} was almost constant with the temperature. Although both R and γ_{K_α} describe the sticking process, only R shows a temperature dependence, which is an open question. In order to explain this, an unknown mechanism should be introduced, such as reactivation after thermalization or during a thermalization process which depends on temperature.

In conclusion, let us summarize the present work. Temperature-dependent phenomena were clearly observed in solid D-T mixtures. With decreasing temperature, (1) λ_c decreased, (2) W increased, and (3) γ_{K_α} was constant. The origins of these observed change (1) and change (2) can be interpreted by the temperature dependence of the $dt\mu$ formation process for (1) and the muon reactivation process for (2), respectively. All of these results were obtained in a solid, and the density was almost fixed at 1.4–1.5. Thus, the observed temperature-dependent phenomena were considered not to be affected by the density dependence. In the present work, we considered the origin of temperature-dependent phenomena in λ_c and W independently. However, we cannot reject the existence of a correlation between λ_c and W. Thus, the origin of the temperature dependence leaves room for further discussion, and a theoretical study is especially anticipated.

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