First observation of radiative photons associated with the $\mu^−$ transfer process from $(t\mu^−)$ to $^3$He through an intermediate $(t\,^3\text{He}\,\mu^−)$ mesomolecule

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Abstract

For the first time, we have observed the 6.76 keV radiative transition photons associated with the $\mu^−$ transfer process from $(t\mu^−)$ atoms to $^3$He nuclei through intermediate $(t\,^3\text{He}\,\mu^−)$ mesomolecule formation in a solid T$_2$ target. The radiative decay branching ratio of the $(t\,^3\text{He}\,\mu^−)$ mesomolecule and the muon transfer rate were determined and compared with theoretical values. We also studied an accumulation process of $^3$He atoms in a solid T$_2$ target by observing the neutron decay rates originating from $t$-$t$ muon-catalyzed fusions. Their time dependence indicates a sudden $^3$He bubble formation in the solid T$_2$ at an atomic concentration of 130 ppm.

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A negative muon transfer process from muonic hydrogen atoms to helium nuclei is one of the important subjects related to the muon catalyzed d–t fusion (µCF) process in a hydrogen target system containing a helium impurity. In the µCF process, negative muons stopped in a D–T mixture induce spontaneous d–t nuclear fusions without any additional devices, and behave as catalysts for repeated µCF cycles during the muon lifetime. The muon mass is 207 times larger than the electron mass, and the size of muonic hydrogen atoms, (dµ−) and (tµ−), are, therefore, smaller than those of electronic hydrogen atoms by the same amount. These small muonic atoms of neutral charge can approach the other hydrogen nuclei, d or t, and induce d–t nuclear fusions without experiencing a Coulomb repulsive force. After d–t fusion producing an α-particle (4He) and a neutron (n), most of the negative muons are liberated to participate in the next µCF cycle: d + t + µ− → α + n + µ− + 17.6 MeV. However, a small fraction of the negative muons are captured by the α-particle in the reaction, thereby terminating the µCF cycle. This fraction is called the α-sticking probability, and is the most important factor in limiting the number of fusion neutrons from a single negative muon, which places a limit on energy production by applying the µCF phenomena [1]. In the actual µCF process, helium impurities gradually accumulate in the D–T target. The major components are 3He nuclei originating from the tritium β-decay; the other is 4He nuclei of the d–t fusion product. The muon loss process due to muon transfer from muonic hydrogen atoms to the accumulated helium nuclei is an important problem to understand the actual µCF process in the D–T mixture, while muon sticking to an α-particle is the major muon loss in the µCF cycle.

In 1986, we succeeded for the first time in observing the 6.85 keV radiative transition photons associated with the µ− transfer from (dµ−) atoms to 4He nuclei through the intermediate (d 4He µ−) mesomolecule at KEK-MSL, using a liquid-deuterium target with 4He impurity dissolved by pressurizing the liquid-deuterium surface with the 4He gas [2]. The muon transfer process from (dµ−) atoms to 4He nuclei through the intermediate (d 4He µ−) mesomolecule is described as

\[ (dµ−) + 4He → (d 4He µ−)^* \]
\[ → d + (^3He µ−) + γ(6.85 \text{ keV}). \]  \tag{1}

The 6.85 keV photon corresponds to a radiation photon originating from the transition from the excited state to the unbound ground state of the mesomolecule. The 6.85 keV photon observation provides direct evidence of the theoretically predicted µ− transfer mechanism through the intermediate (d 4He µ−) mesomolecule. Aristov et al. have proposed a description of the µ− transfer process from muonic hydrogen atoms to helium nuclei via the formation of intermediate mesomolecules, and calculated the energy levels and the formation rates of mesomolecules [3]. Kravtsov et al. have calculated the photon energy spectra originating from deexcitations of the excited mesomolecules formed by hydrogen and helium isotopes, and predicted asymmetrically energy-broadened photon line shapes, which reflect the potential energy curves of the transitional states [4]. The photon energy, the asymmetric line shape and the transfer rate obtained in our experiment were in good agreement with theoretical values [2]. On the other hand, the direct transfer probability from the (dµ−) atoms to the 4He nuclei, (dµ−) + 4He → d + (^4He µ−), was calculated to be small: \( \sim 105 \text{ s}^{-1} \) [5]. We have continued further experimental studies on the muon transfer mechanism from hydrogen to helium in the systems of d–3He, d–4He and p–4He at KEK-MSL [6]. The transfer rates and the radiative decay branching ratios were obtained and compared with theoretical values [7–9]. A particle-emitting decay mode of the excited mesomolecules has been proposed theoretically in addition to radiative photon emission, which has explained well the isotope dependence of the radiative decay branching ratios of mesomolecules [6]. Several experiments have also been performed at PSI for these systems to obtain the photon energy spectra, transfer rates and radiative decay branching ratios [10,11].

As for the t–3He system, the µ− transfer mechanism can be expressed similarly as

\[ (tµ−) + ^3He → (^3He µ−)^* \]
\[ → t + (^3He µ−) + γ \text{ (radiative photon)}. \]  \tag{2}

The muon transfer rates from (tµ−) atoms to ^3He nuclei accumulated in the D–T target and their temperature dependence were obtained by the LAMPF group in their fusion neutron data analysis of d–t µCF studies [12]. However, so far, no direct experiment has been
carried out to investigate the $\mu^-$ transfer mechanism in the $t$-$^3$He system by observing the radiative photon, in spite of its importance for understanding the realistic d-t µCF process with the existence of a small $^3$He impurity. Theoretical studies on the $t$-$^3$He system have been made to predict the transfer rate [13], the radiative photon spectrum [4] and the decay rate of the $(t^3$He $\mu^-)$ mesomolecule [8,9].

We recently performed a $t$-$t$ µCF experiment using a solid T$_2$ target as a part of the d-t µCF research program at the RIKEN-RAL Muon Facility. A preliminary result on the $t$-$t$ µCF experiment has been published [14]. In the experiment, we also succeeded in observing radiative photons associated with the $\mu^-$ transfer process from $(t\mu^-)$ atoms to $^3$He nuclei through the intermediate $(t^3$He $\mu^-)$ mesomolecule, where the $^3$He impurity originates from the tritium $\beta$-decay and accumulates in the solid T$_2$ target. We obtained the radiative decay branching ratio of the $(t^3$He $\mu^-)$ mesomolecule and the muon transfer rate from $(t\mu^-)$ atoms to $^3$He nuclei. In this Letter we report on the experimental results concerning the muon transfer process in the $t$-$^3$He system.

We conducted the present experiment using the same experimental set-up installed for a series of d-t µCF studies [15] at Port 1 of the RIKEN-RAL pulsed Muon Facility located at the Rutherford Appleton Laboratory in the UK. The T$_2$ target gas was produced at the Department of Radioisotopes of JAERI [16], and installed in an in-situ tritium gas-handling system [17]. The isotope enrichment of T was 99.1% and the remaining component was H of 0.9%. The target gas was purified by passing it through a palladium filter to remove any $^3$He impurity originating from tritium $\beta$-decay just before the measurements. The T$_2$ gas was introduced to the target cell cooled by a helium-flow cryostat and solidified at 16 K [17]. The solid T$_2$ target with a volume of 0.55 cm$^3$ was formed by the T$_2$ gas: a volume of 0.558 liter at STP and an inventory of 53.7 TBq (1450 Ci). The target cell, made of cupro-nickel alloy, was a cylinder of $\varnothing$14 mm $\times$ 14 mm with a beryllium window of 0.5 mm thickness for a low-energy photon observation. The inside surface of the cell was covered by a silver foil of 0.2 mm thickness to absorb any muon beam-induced photon background from the target wall. The target was positioned at the magnetic field center of a superconducting Helmholtz coil. A magnetic field of 2.4 T converged the incoming $\mu^-$ beam towards the target cell and prevented most of the $\mu$–$e$ decay electron background from reaching the photon detector, thereby reducing the photon background. For photon detection, a Si(Li) X-ray detector (70 mm$^2$ $\times$ 3.5 mm) was placed perpendicular to the $\mu^-$ beam at a distance of 13.3 cm from the target. In order to detect the t–t fusion neutron, two calibrated NE213 liquid scintillators ($\varnothing$2 inch $\times$ 2 inch) were positioned at a distance of 84 cm downstream of the target. Lead bricks of 5 cm thickness were placed in front of the neutron counters to eliminate the $\mu$–$e$ decay electron background. The $\mu$–$e$ decay electrons originating from the solid T$_2$ target were detected by segmented plastic scintillation counters located at the backward and forward directions from the target. A decay $\mu^-$ beam with a momentum of 54.5 MeV/c was extracted by the superconducting muon channel [18] and injected to the T$_2$ target. About 34 muons were stopped in the target during every muon pulse with a double-pulsed time-structure (100 ns total pulse width and 230 ns pulse separation at 50 Hz). It should be emphasized here that a pulsed muon beam is essential for a photon detection experiment under a huge white-noise type radiation background associated with the Bremsstrahlung of tritium $\beta$-ray decay. A vital improvement of the signal-to-noise ratio in the delayed photon spectrum was obtained by opening the observation time window synchronously with the muon pulse. We used the solid T$_2$ target in the experiment so that almost all the quantity of T$_2$ gas could be collected in the target cell for achieving the maximum experimental yields. $^3$He removal from the solid T$_2$ target was carried out by either a T$_2$ gas-purification using a tritium gas-handling system, or by forming it through the T$_2$ liquid phase from the gaseous T$_2$. In the course of the d-t µCF experiments, we found that $^3$He accumulated in the solid D-T target, but did not do so in the liquid target, by observing the time dependence of the fusion neutron decay rates after $^3$He removal [19]. Immediately following solid T$_2$ formation, data taking was started, and continued for 60 hours; the final $^3$He atomic concentration reached to 385 ppm in the target. Since the data taking was made in an event-by-event mode [20], all of the data of the photon, neutron and decay electron could be analyzed off-line as a function of time ($\tau$) after $^3$He removal, so that the data at the
A typical delayed photon spectrum is shown in Fig. 1. We have clearly observed for the first time a characteristic radiative photon at an energy of (6.76 ± 0.06) keV with a line width of (0.21 ± 0.14) keV at FWHM. These values are in good agreement with the predicted values of the photon spectrum in the t–3He system [4]. The observed photon has an asymmetric line shape with a tail at the low energy side; this feature has also been predicted by theory [4]. Therefore, this photon can be considered to originate from the radiative decay of the excited (t^3He µ^-) intermediate mesomolecule formed in the µ^- transfer process from (tµ^-) atoms to the accumulated ^3He nuclei in the solid T2 target. In addition to the radiative photon, we have observed Kα (8.2 keV) and Kβ (9.6 keV) muonic X-ray lines, which originate from the (αµ^-) atoms formed by the µ^- to ^4He sticking in the t-t µCF cycles: \[ t + t + µ^- → (αµ^-) + 2n + 11.3 \text{ MeV} \] [14]. The solid curve in the figure is a typical fitting result using single Gaussian line shapes for Kα and Kβ lines, and an asymmetric Gaussian line shape for the radiative photon with different Gaussian line widths at the low and high-energy sides.

The measured neutron shows a simple exponential decay time spectrum with a single component and a continuous recoil proton energy spectrum up to 9 MeV. A quantitative analysis of the observed energy spectrum is complicated because it is overlapped by two neutron-energy components from the t-t µCF reaction [14]. On the other hand, the single component of the neutron time spectrum, called the neutron disappearance rate, gives information about the muon transfer loss process from (tµ^-) atoms to the accumulated ^3He nuclei, because the active muons contributing to the t-t µCF cycle are taken away by the muon transfer process and the neutron disappearance rate increases according to the ^3He atomic concentration in the solid T2 target.

The neutron disappearance rate, \( \lambda_n(τ) \), in the solid T2 at time τ after the ^3He removal is described as follows, by assuming that the total muon loss is composed of two major components of the muon transfer and muon sticking processes in the t-t µCF cycle:

\[
\lambda_n(τ) = \lambda_0 + Wφλc + φC_{He}(τ)λ_{t^3He}\mu.
\]

where \( \lambda_0, W, \phi, λ_c, C_{He}(τ) \) and \( λ_{t^3He}\mu \) are the free-muon decay rate \((0.455 \times 10^6 \text{s}^{-1})\), effective sticking probability in the t-t µCF cycle, T2 target density normalized to the liquid-hydrogen density \( (4.25 \times 10^{22} \text{ atoms/cm}^3) \), t-t fusion cycling rate independent of the ^3He impurity, ^3He atomic concentration in the solid T2 target at time τ and muon transfer rate from (tµ^-) atoms to ^3He nuclei, respectively. We have observed a time-dependent change of \( \lambda_n(τ) \), as shown in Fig. 2. The \( \lambda_n(τ) \) for the solid T2 target shows three interesting features: (1) a linear increase for τ = 0–20 h, (2) a gradual decrease for τ = 20–35 h and (3) nearly constant for τ = 35–60 h. A similar behavior with a different time scale has been observed for the D-T solid target \((C_t = 70\%)\), as also shown in the figure. These interesting phenomena are considered to originate from the ^3He accumulation effect in the solid hydrogen target. On the other hand, such a time dependence of \( \lambda_n(τ) \) has never been observed in a liquid D-T target. This fact means that the ^3He accumulates in the solid T2 and D-T targets, but does not do so in liquid targets, because the ^3He is released to the gaseous space in the target [19].
can neglect here the $^3$He concentration dissolved in the liquid target proportional to the partial pressure in the gaseous space according to Henry’s law. The calculated $^3$He atomic concentrations in the solid at $\tau = 20$ h for $T_2$ and $\tau = 30$ h for D–T ($C_T = 70\%$) are approximately 130 ppm. After exceeding this atomic concentration, the observed $\lambda_n(\tau)$ gradually decreases, indicating a decrease in the effective $^3$He atomic number contributing to the muon transfer process in the solid hydrogen. This suggests that the distributed $^3$He atoms collect together at this specific atomic concentration and create $^3$He bubbles at the interstitial sites of the solid hydrogen lattice. However, further experimental and theoretical studies are required to clarify this phenomenon.

It should be mentioned here that the solid tritium in a cylindrical cavity is known to form a uniform and stable distribution with a certain time constant due to the sublimation effect induced by the $\beta$-decay radiation heating [21]. In the d–t $\mu$CF experiments with solid targets of high tritium concentrations [19], we monitored the change of stopping muon numbers in the target by the $\mu$–e decay, and found that the D–T solid formed the stable distribution at 16 K within one hour after the solidification. In the present experiment, we also confirmed that the stable distribution of $T_2$ solid target completed within one hour. On the contrary, the observed changes of $\lambda_n$, as shown in Fig. 2, occur with a longer time scale than expected from the sublimation effect, and the phenomena cannot be explained by the sublimation effect.

The observed increase of $\lambda_n(\tau)$ due to the $\mu^-$ transfer process at time $\tau$ after $^3$He removal is described as

$$\lambda_n(\tau) = \phi C_{^3\text{He}}(\tau) \lambda_{^3\text{He} \mu^\pm}.$$  \hspace{1cm} (4)$$

where $\lambda_n(0) = 0.882 \pm 0.030 \, \mu\text{s}^{-1}$ is the neutron disappearance rate at $\tau = 0$. The $C_{^3\text{He}}(\tau)$ is simply expressed as $C_{^3\text{He}}(\tau) = C_T \lambda_T \tau$, where $C_T$ and $\lambda_T$ are the tritium concentration ($C_T = 0.991$) of the solid $T_2$ target and the tritium decay rate ($1.54 \times 10^{-4}$ day$^{-1}$), respectively. We can therefore expect a linear increase of $\lambda_n(\tau)$ against the time $\tau$ after $^3$He removal.

By taking a linear increasing region of the observed $\lambda_n(\tau)$ ($\tau = 0$ to 20 hours) and assuming that all of the $^3$He atoms accumulate in the solid $T_2$ target, the muon transfer rate, $\lambda_{^3\text{He} \mu^\pm}$, has been obtained to be $(4.6 \pm 0.4) \times 10^9$ s$^{-1}$ at 16 K.

The radiative photon yield from the ($t^3\text{He} \mu^-$) mesomolecule, $Y_{^3\text{He} \mu^\pm}$, can be expressed as

$$Y_{^3\text{He} \mu^\pm}(\tau) = \varepsilon_{^3\text{He} \mu} \frac{\lambda_n(\tau) - \lambda_n(0)}{\lambda_n(\tau)},$$ \hspace{1cm} (5)$$

where $\varepsilon_{^3\text{He} \mu}$ and $(\lambda_n(\tau) - \lambda_n(0))/\lambda_n(\tau)$ are the radiative decay branching ratio of the ($t^3\text{He} \mu^-$) mesomolecule and the muon transfer loss ratio at time $\tau$ after $^3$He removal, respectively. The radiative photon yield, corrected for the detection efficiency of the Si(Li) detector, was normalized to the stopping muon number in the target. Our measurement showed a good correlation between the time dependence of the radiative photon yield and that of the muon transfer loss ratio determined by the neutron disappearance rate shown in Fig. 2. The ratio of the radiative photon yield to the muon transfer loss ratio, $\varepsilon_{^3\text{He} \mu}$ in Eq. (5), was calculated at every 4 hours period and was reasonably constant within the statistical error (typically $\pm 10\%$). By combining the averaged ratio $(0.947 \pm 0.033)$ with the systematic error ($\pm 0.06$), the radiative decay branching ratio was obtained to be $(0.95 \pm 0.07)$.  

Fig. 2. Time-dependent change of the fusion neutron disappearance rate, $\lambda_n(\tau)$, for a solid $T_2$ target in 2-hour bins after solidification. Similar data for a solid target of D–T mixture ($C_T = 70\%$) is also shown for a comparison.
The muon transfer process from (tµ−) atoms to the accumulated 3He nuclei are expressed as

\[
(t\mu^-) + ^3\text{He} \\
\rightarrow (t^3\text{He} \mu^-) \nonumber^* \nonumber \\
\rightarrow (t^3\text{He} \mu^-) + t + \gamma \quad \text{radiative decay,} \quad (6) \\
\rightarrow (t^3\text{He} \mu^-) + t + \text{K.E.} \quad \text{particle decay,} \quad (7) \\
\rightarrow (t^3\text{He} \mu^-) + t + e^- \quad \text{Auger emission decay} \quad (8)
\]

Three decay modes of the excited \((t^3\text{He} \mu^-)^*\) mesomolecule have been theoretically predicted. In the radiative decay mode, we have observed the characteristic radiative photons with an asymmetrically energy-broadened line shape to provide direct evidence of the predicted transfer mechanism through the intermediate \((t^3\text{He} \mu^-)\) mesomolecule [3]. The observed photon energy of \((6.76 \pm 0.06)\) keV and the line width of \((0.89 \pm 0.14)\) keV at FWHM in the present work are in good agreement with the theoretical values, reflecting the potential energy curve of the \((t^3\text{He} \mu^-)\) mesomolecule [4]. By taking into account of the particle decay mode, the isotope dependence of radiative decay branching ratios has been explained well by the reduced-mass effect for the mesomolecules formed in the d–\(^3\text{He},\) d–\(^4\text{He}\) and p–\(^4\text{He}\) systems [6]. As for the Auger emission decay mode, the calculated rates are about 25% of the radiative decay rates of the mesomolecules [8].

The radiative decay branching ratio is an important value to investigate the dissociation mode of the excited \((t^3\text{He} \mu^-)\) mesomolecule. The obtained value of \((0.95 \pm 0.07)\) can be compared with the theoretical values, 0.63 [9] and 0.58 [8], and shows a dominance of the radiative decay mode of the mesomolecule.

We have obtained the muon transfer rate from \((t\mu^-)\) atoms to \(^3\text{He}\) nuclei to be \((4.6 \pm 0.4) \times 10^9 \text{ s}^{-1}\) at a temperature of 16 K. The LAMPF group has calculated the transfer rates and the temperature dependence using the neutron data of their d–t µCF experiments. They have obtained a transfer rate of \((0.9 \pm 0.2) \times 10^9 \text{ s}^{-1}\) at the lowest temperature of 100 K [12]. Although these two experimental values were obtained at different temperatures, they seem to be comparable by taking into account the temperature dependence of \(\lambda_{t^3\text{He} \mu^-}\): it increases rapidly as the temperature decreases from 100 K to 16 K. From a theoretical point of view, the present value may be compared with the predicted value of \(4.6 \times 10^9 \text{ s}^{-1}\) at \(\varepsilon = 0.004 \text{ eV}\) for the simple-approach approximation with electron screening and averaged over the Maxwellian distribution by Kravtsov et al. [13]. The obtained transfer rate can be also considered to be the formation rate of the \((t^3\text{He} \mu^-)\) mesomolecule because the dissociation rates of the mesomolecules are much higher, by two orders of magnitude: \(~10^{11} \text{ s}^{-1} [8,9].\)

In summary, we have observed for the first time the 6.76 keV radiative photons associated with the muon transfer process from \((t\mu^-)\) atoms to \(^3\text{He}\) nuclei through the intermediate \((t^3\text{He} \mu^-)\) molecular formation in a solid T2 target. The observed features of the photon energy spectrum are in good agreement with theoretical predictions. We also have determined a radiative decay branching ratio of \((0.95 \pm 0.07)\) for the \((t^3\text{He} \mu^-)\) mesomolecule and a muon transfer rate of \((4.6 \pm 0.4) \times 10^9 \text{ s}^{-1}\) at 16 K. These values will be good objectives for theoretical studies on the muon transfer mechanism from \((t\mu^-)\) atoms to \(^3\text{He}\) nuclei.

In addition, we have also studied the \(^3\text{He}\) accumulation process in the solid T2 target by observing the t–t µCF neutron disappearance rates. Their time dependence after \(^3\text{He}\) removal seems to indicate a sudden \(^3\text{He}\) bubble formation in the solid T2 at an atomic concentration of 130 ppm.

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