

Charge transfer from the ground state of muonic hydrogen to ${}^4\text{He}$ at room temperature

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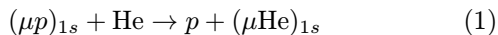
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Abstract. The present status of research of muon transfer from the ground state of muonic protium to ${}^4\text{He}$ is reviewed. The analysis of a recent measurement in a triple gas mixture of $\text{H}_2+{}^4\text{He}+\text{Ne}$ at 15 bar and room temperature is presented and the result is compared to the existing experimental and theoretical rates. The average muon transfer rate from protium to ${}^4\text{He}$ determined from all lifetime measurements is $\lambda_{p{}^4\text{He}} = (0.42 \pm 0.04) \times 10^8 \text{ s}^{-1}$.

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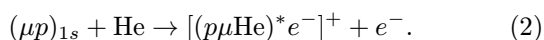
1 Introduction

The pioneering paper on direct muon transfer by Gershtein [1] in 1963 predicted that the direct transfer rate $\lambda_{p{}^4\text{He}}^{\text{dir}}$ (about 10^5 s^{-1}) from the ground state of muonic hydrogen to helium



is several orders of magnitude smaller than to higher Z elements such as carbon or oxygen (about 10^{11} s^{-1}) [1,2]. The reason is the absence of crossing and pseudo crossing points in the potential curves between the systems $\mu p + \text{He}^{++}$ and $p + (\mu\text{He})^+$. A first attempt to measure the muon transfer rate from hydrogen to helium [3] yielded a rate of the predicted order of magnitude. About ten years later and using a more refined approximation, Matveenko and Ponomarev [4] calculated the transfer rates from the hydrogen isotopes, protium and deuterium, to the helium isotopes ${}^3\text{He}$ and ${}^4\text{He}$. Their rates were about ten times higher than the rate calculated by Gershtein. In a second attempt to measure the transfer rate to helium [5], only an upper limit could be determined from the experimental data, which did not contradict the predictions.

In 1981, a new transfer mechanism was proposed by Aristov *et al.* [6] (Gatchina group), namely *via* the formation of a metastable muonic hydrogen-helium molecule in the $(2p\sigma)$ excited state



This process leads to much higher transfer rates ($\sim 10^8 \text{ s}^{-1}$). According to Aristov *et al.*, the molecule

should decay with a rate of about 10^{12} s^{-1} , either by Auger effect or by emitting an X-ray of about 7 keV, leaving a hydrogen and a muonic helium atom in its ground state. Somewhat later, the same Gatchina group corroborated their theoretical predictions by calculating the formation rate ($\lambda_{p\mu\text{He}}$) of the molecule using different assumptions and approximations [7,8]. In addition, several experiments obtained muon transfer rates from hydrogen to ${}^4\text{He}$ [9] and from deuterium to ${}^3\text{He}$ and/or to ${}^4\text{He}$ [10,11] of order 10^8 s^{-1} , indirectly confirming the new transfer mechanism. The first direct evidence for the formation of a muonic deuterium-helium molecule was obtained in 1988 by Matsuzaki *et al.* [12], who observed the 7 keV decay X-rays of the molecule $(d\mu^4\text{He})^*$ in a liquid $\text{D}_2+{}^4\text{He}$ mixture and measured their time distribution.

Two other experiments were performed to determine the muon transfer rate from hydrogen to ${}^4\text{He}$ [13,14]. The experiment by von Arb *et al.* [14], in contrast to the experiments measuring the lifetime of the $(\mu p)_{1s}$ atom [9,13], was based on a yield measurement of the 7 keV line. The result was an order of magnitude lower than the rates from the lifetime measurements. In 1993, a new series of experiments was performed by Ishida *et al.* [15] in binary liquid mixtures of $\text{D}_2+{}^4\text{He}$, $\text{D}_2+{}^3\text{He}$ and $\text{H}_2+{}^4\text{He}$. Surprisingly, the 7 keV X-ray was not observed in the $\text{H}_2+{}^4\text{He}$ mixture, whereas it could be measured in the mixtures with deuterium. This fact was explained the same year by Kino and Kamimura [16]. They predicted the existence of a third decay channel of the respective mesomolecules, namely the direct particle decay. Since then, the decay rates have been calculated by several authors [17–21]. They all show, that this dissociative decay channel is strongly isotope-dependent, the rate increasing with decreasing reduced mass of the two heavy particles. By including this third

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decay channel, the strong disagreement between the yield measurement [14] and the lifetime measurements [9,13] found an explanation.

The previously performed experiments on the muon transfer from protium to ^4He at room temperature are reviewed in more detail in Section 2, whereas Section 3 presents a new measurement of the transfer rate to ^4He , using the same lifetime method as Jacot-Guillarmod *et al.* [13].

2 Previous measurements

The first experiment, which indirectly confirmed the muon transfer mechanism *via* the formation of the molecule, was performed by Bystritsky *et al.* [9] at JINR (Dubna). They were using triple gas mixtures of $\text{H}_2 + ^4\text{He} + \text{Xe}$. In a series of three runs with different xenon gas densities, the relative atomic concentrations of ^4He were changed between 4.7% and 47%, at total gas pressures between 23–55 bars. The time distribution of the muonic xenon X-rays around 3.8 MeV and the decay electrons were measured using plastic scintillators, NaI and stilbene detectors. From the combined analysis of the different time spectra and the change of the lifetime of the $(\mu p)_{1s}$ atom, owing to the different helium gas concentrations, the total muon transfer rate to helium $\lambda_{p^4\text{He}}$ was deduced. This total rate is the sum of direct transfer $\lambda_{p^4\text{He}}^{\text{dir}}$ (1) and transfer *via* molecular formation $\lambda_{p\mu\text{He}}$ (2). Its value $\lambda_{p^4\text{He}} = (0.36 \pm 0.10) \times 10^8 \text{ s}^{-1}$ is in good agreement with the assumption of a molecular charge exchange mechanism [6]. The method employed by Bystritsky *et al.* has the advantage that the measured rate is independent from the transfer rate to the third gas component, in this case xenon. However, the detectors used had poor energy resolutions which made a proper background subtraction difficult. In addition, possible impurities contained in the target gas such as nitrogen or oxygen were not detectable.

The second muon transfer experiment from protium to ^4He was performed at PSI (Villigen) by the Fribourg group [13]. Again, triple gas mixtures were employed in these measurements. However, in contrast to the method employed by the Dubna group, the muon transfer rate to the third component, in this case argon, had to be precisely known. Since for elements with $Z > 2$ no element-specific calculation of the muon transfer rate exists and the available experimental results differ widely between each other, the transfer rate to argon had to be remeasured. Therefore, the muon transfer rate to argon was determined from two measurements in binary $\text{H}_2 + \text{Ar}$ mixtures with different argon gas concentrations. From the lifetimes of the $(\mu p)_{1s}$ atoms, deduced from the time distributions of the Lyman series X rays in argon between 640–840 keV measured by germanium detectors, a mean transfer rate to argon of $\lambda_{p\text{Ar}} = (1.44 \pm 0.04) \times 10^{11} \text{ s}^{-1}$ was determined, in agreement with Placci *et al.* [22]. The transfer rate to helium was then determined from the time distributions of the muonic argon X-rays measured in two triple gas mixtures. The total pressure was 15 bar, with atomic concentrations of helium of 20% and 33%, respectively. The total

mean transfer rate to ^4He , $\lambda_{p^4\text{He}} = (0.88 \pm 0.09) \times 10^8 \text{ s}^{-1}$, obtained by using the above given transfer rate to argon, was a factor of two higher than the one quoted by Bystritsky *et al.* [9]. The advantage of the method used by the Fribourg group is the high energy resolution of the germanium detectors, leading to a large peak-to-background ratio and a reliable subtraction of the background underneath the muonic argon X-rays. In addition, impurities in the target gas can easily be identified at a level of a few ppm and corrected for. The disadvantage is the high sensitivity to uncertainties in the rate $\lambda_{p\text{Ar}}$ and in the argon concentration in the triple mixture, since the transfer rates to argon and helium differ by about three orders of magnitude.

By using the same method, the Fribourg group remeasured the muon transfer rate from hydrogen to ^4He in a triple gas mixture with methane (CH_4) as the third gas component [23]. This measurement was performed at a total pressure of 40 bar, with an atomic concentration of helium of 33%. Again, the transfer rate to carbon of the methane molecule had first to be measured in binary mixtures $\text{H}_2 + \text{CH}_4$. The shapes of the measured time distributions of the muonic carbon X-rays showed that the transfer rate $\lambda_{p\text{C}}$ was strongly dependent on the kinetic energy of the muonic hydrogen atom, increasing with decreasing energy. Hence, the obtained precision for the transfer rate from thermalized μp 's to carbon using the late delayed part of the time distribution was limited. Therefore, only an upper limit for the total transfer rate to helium, namely $\lambda_{p^4\text{He}} < 0.6 \times 10^8 \text{ s}^{-1}$, could be given. This upper limit contradicted the transfer rate obtained by the same Fribourg group in the triple mixture with argon [13].

This contradiction and the strong energy dependence observed in the transfer rate to carbon [23] as well as to oxygen [24] made the true value of the transfer rate to argon questionable. To clarify the situation, the transfer rate to argon was remeasured under improved experimental conditions [25]. Contrary to the former $\text{H}_2 + \text{Ar}$ measurements [13], the high statistics accumulated in the time distributions of the muonic argon X-rays revealed a dependence of the transfer rate on the energy of the μp atom. The obtained muon transfer rate from thermalized hydrogen to argon turned out to be $\lambda_{p\text{Ar}} = (1.63 \pm 0.09) \times 10^{11} \text{ s}^{-1}$, *i.e.* about 15% higher than in the previous experiments [13]. As a consequence, the recalculated muon transfer rate to helium obtained from the measurement in the triple mixture $\text{H}_2 + ^4\text{He} + \text{Ar}$ [13] yielded a considerably lower value, namely $\lambda_{p^4\text{He}} = (0.51 \pm 0.19) \times 10^8 \text{ s}^{-1}$ [25], in agreement with theory [6–8] and other experiments [9, 23].

In contrast to the experiments using the lifetime method [9,13,23], von Arb *et al.* [14] determined the transfer rate to ^4He by measuring X-ray yields. Specifically, in a binary gas mixture of $\text{H}_2 + ^4\text{He}$ with an atomic concentration of helium of 33% and a total pressure of 5 bar, the yields of the muonic Lyman series of hydrogen around 2 keV and of the 7 keV decay X-rays of the $(p\mu^4\text{He})^*$ molecule were measured. For this purpose, two xenon gas scintillation proportional counters were used.

Taking into account the direct ground state transfer (1), by using the rate $\lambda_{p^4\text{He}}^{\text{dir}} = 0.055 \times 10^8 \text{ s}^{-1}$ calculated by Matveenko and Ponomarev [4], the formation rate of the $(p\mu^4\text{He})^*$ molecule was determined. The obtained rate $\lambda_{p\mu^4\text{He}} = (0.032 \pm 0.013) \times 10^8 \text{ s}^{-1}$ is in strong disagreement with theory [6–8] and experiments [9, 13]. Although this method does not depend on a transfer rate to a third element and does not measure the lifetime of the $(\mu p)_{1s}$ atom, the obtained formation rate decisively depends on the rates of the possible decay modes of the muonic hydrogen-helium molecule. Since at this time only two decay channels were known, namely the radiative and the Auger decay, but not the particle decay - which in addition turns out to be the main channel in this case - the analysis leads to a wrong result. By using the three decay rates as calculated by Kravtsov *et al.* [18], a muon transfer rate $\lambda_{p^4\text{He}} = (0.44 \pm 0.16) \times 10^8 \text{ s}^{-1}$ is obtained. This result is in good agreement with other experiments and with theory. One may mention here that, depending on the approaches used in the calculation of the rates of the decay channels [21], the rate $\lambda_{p^4\text{He}}$ can become as high as $(0.68 \pm 0.26) \times 10^8 \text{ s}^{-1}$.

3 New Measurement

Considering the puzzling situation in the past and the difficulties of the previous measurements, we decided to remeasure the total transfer rate to helium by employing the triple gas mixture method with neon as the third gas component. Using this method, we are independent from theoretical predictions on the decay of the $(p\mu^4\text{He})^*$ molecule. Neon as the third gas component has the advantage that its transfer rate from muonic protium ($\lambda_{p\text{Ne}}$) is energy independent and about one order of magnitude smaller than the one to argon [26]. Hence, higher neon gas concentrations can be employed, which in turn can be determined with better accuracy.

The experiment was performed at the $\mu E4$ channel at PSI. The same target apparatus was used as in our previous experiments [13, 23, 25, 26]. More details can be found in reference [13]. The target was filled with a triple gas mixture of $\text{H}_2 + 33.4\% ^4\text{He} + 0.5370\% \text{Ne}$ (atomic concentrations) at a pressure of 14.97 ± 0.07 bar and a temperature of 293.6 ± 1.3 K. This corresponds to an atomic density of liquid hydrogen of $\phi = 1.296 \pm 0.008\%$. Two germanium detectors were used to measure the muonic X-rays of the Lyman series of neon between 207 keV and 270 keV. The energy and time resolutions of the detection system were determined using the $\mu\text{Ne}(2-1)$ transition as a reference. The 66 cm^3 Ge detector had a 0.9 keV energy and 8 ns time resolution, the 75 cm^3 detector 1.1 keV and 9 ns, respectively. The electronic setup is similar to the one described by Knowles *et al.* [27]. With a primary proton beam of 1 mA, negative muons with a momentum of approximately 31 MeV/c were focused onto the target center as a $2.2 \times 2.2 \text{ cm}^2$ beamspot. With a pile up rejection circuit of $\pm 5 \mu\text{s}$ gate length, typically 140 counts/s were measured in both germanium detectors.

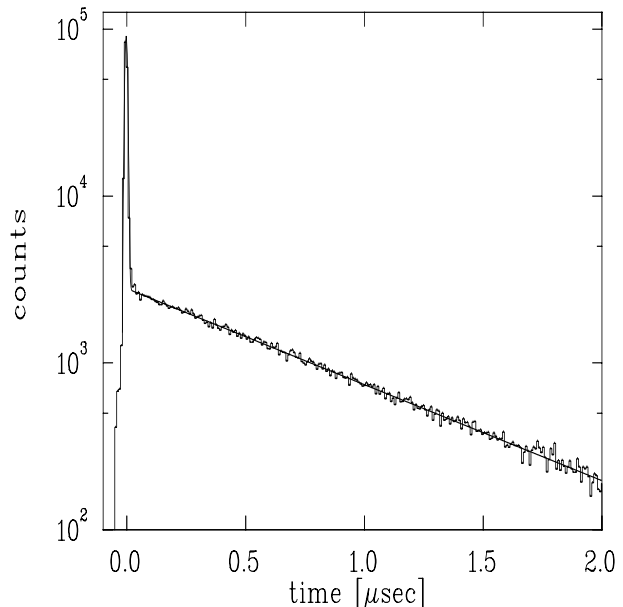


Fig. 1. Background subtracted time spectrum of the sum of the $\mu\text{Ne}(2-1)$, $\mu\text{Ne}(3-1)$ and $\mu\text{Ne}(4-1)$ transitions, measured by the 66 cm^3 Ge detector. The lifetime $\tau_{\mu p_{1s}}$ was determined by using a single exponential in the time interval [0.1–2.0 μs].

When a muon captured by hydrogen has reached the ground state $(\mu p)_{1s}$, the muonic hydrogen atom disappears by free muon decay with the rate $\lambda_0 = 4.5516 \times 10^5 \text{ s}^{-1}$, by formation of a $pp\mu$ molecule ($\lambda_{pp\mu}$) or by muon transfer to ^4He ($\lambda_{p^4\text{He}}$), to Ne ($\lambda_{p\text{Ne}}$) or to impurities (λ_{pi}). Due to the use of natural hydrogen, which contains about 150 ppm of deuterium, the muon can also transfer from protium to deuterium with a rate λ_{pd} . Although one has to take into account the muon transfer from protium to deuterium, the muons directly captured in deuterium can be neglected. Therefore, the measured disappearance rate $A_{\mu p_{1s}}$ is the sum of the different rates,

$$A_{\mu p_{1s}} = \frac{1}{\tau_{\mu p_{1s}}} = \lambda_0 + \phi(c_{^4\text{He}}\lambda_{p^4\text{He}} + c_p\lambda_{pp\mu} + c_d\lambda_{pd} + c_{\text{Ne}}\lambda_{p\text{Ne}} + c_i\lambda_{pi}). \quad (3)$$

Here, $\tau_{\mu p_{1s}}$ is the lifetime of the $(\mu p)_{1s}$ atom, c_j the atomic concentration of element j, and $\lambda_{p^4\text{He}}$ the sum of $\lambda_{p^4\text{He}}^{\text{dir}}$ (1) and $\lambda_{p\mu^4\text{He}}$ (2). The normalized rates $\lambda_{pd} = (1.68 \pm 26) \times 10^{10} \text{ s}^{-1}$ [28], $\lambda_{pp\mu} = (2.34 \pm 17) \times 10^6 \text{ s}^{-1}$ [29], and $\lambda_{p\text{Ne}} = (0.0849 \pm 18) \times 10^{11} \text{ s}^{-1}$ [26] are used in this work. The contribution of the decay channels $\lambda_{pp\mu}$ and λ_{pd} in (3) is less than 7%, such that their energy dependences can be neglected. The transfer rate $\lambda_{p\text{Ne}}$ is energy independent [26]. The time spectra of the muonic neon X-rays measured in the triple gas mixture $\text{H}_2 + ^4\text{He} + \text{Ne}$ shows no deviation from the shape of a single exponential (see Fig. 1). Hence, from a fit of the delayed part of this time distribution, the lifetime $\tau_{\mu p_{1s}}$ of the $(\mu p)_{1s}$ atom can be extracted. Using (3), the total muon transfer rate to ^4He $\lambda_{p^4\text{He}}$ is then determined.

Specifically, the time distributions of the muonic neon X-rays were analyzed using background subtracted time

Table 1. Measured lifetimes of the $(\mu p)_{1s}$ atoms in the triple gas mixture $H_2 + ^4He + Ne$.

transition	66 cm ³ detector	75 cm ³ detector
$\mu Ne(2-1)$	758.7 ± 4.5 ns	753.6 ± 4.2 ns
$\mu Ne(3-1)$	740.2 ± 9.2 ns	756.6 ± 8.7 ns
$\mu Ne(4-1)$	752.0 ± 7.7 ns	747.2 ± 6.9 ns
weighted mean	754.5 ± 3.6 ns	752.6 ± 3.3 ns
$\mu Ne(\text{sum spectrum})$	755.2 ± 3.3 ns	754.0 ± 3.4 ns

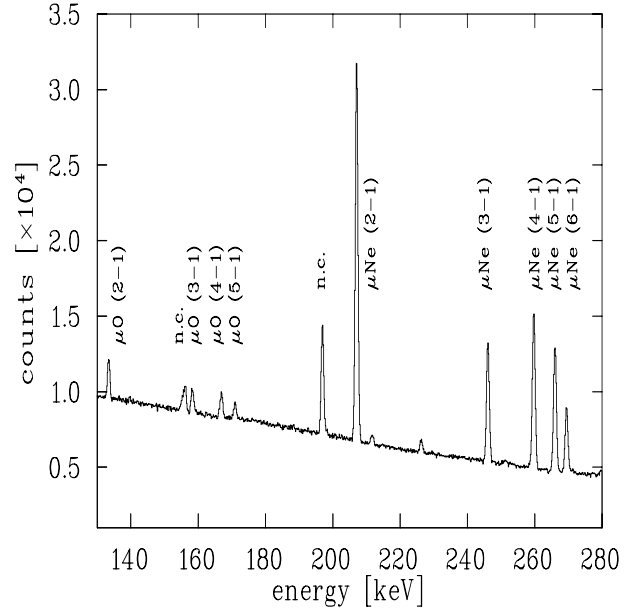
spectra [30]. For both detectors sufficient statistics was accumulated for the $\mu Ne(2-1)$, $\mu Ne(3-1)$ and $\mu Ne(4-1)$ lines, such that their time spectra could be analyzed individually. The results obtained by fitting the delayed part [0.1–2.0 μs] are displayed in Table 1. The measured lifetimes of the $(\mu p)_{1s}$ atoms from the different muonic neon transitions agree with each other. Hence, in order to improve the accuracy, the time spectra of the Lyman series of each detector were summed up, yielding the lifetimes $\tau_{\mu p_{1s}} = 755.2 \pm 3.3$ ns for the 66 cm³ detector and $\tau_{\mu p_{1s}} = 754.0 \pm 3.4$ ns for the 75 cm³ detector. These values are in good agreement with each other and also with the mean values of the individual transitions (see Tab. 1). For the determination of the transfer rate to helium, the averaged value $\langle \tau_{\mu p_{1s}} \rangle = 754.6 \pm 2.5$ ns will be used.

Figure 2 shows the energy spectrum of the delayed photons between 120 keV and 300 keV. The highest transition observed in the muonic Lyman series of neon is the $\mu Ne(6-1)$ line. Apart from several nuclear gamma rays resulting mainly from nuclear capture of muons by elements of the target walls, one notices the presence of the first four transitions of the Lyman series of muonic oxygen. The time constant determined from the analysis of their time distributions, $\tau_{\mu p_{1s}} = 763 \pm 26$ ns, corresponds to the lifetime of the $(\mu p)_{1s}$ atoms deduced from the muonic neon X-rays. In addition, the relative intensities of the $\mu O(n-1)$ lines have a structure which is characteristic for muon transfer to oxygen [30, 31]. The delayed muonic oxygen X-rays are consequently due to muon transfer to oxygen present in the target gas as an impurity.

The transfer to this impurity is taken into account by the last term of equation (3). In the present case, it can be written as

$$c_O \lambda_{pO} = \frac{I_O}{I_{Ne}} c_{Ne} \lambda_{pNe}, \quad (4)$$

where I_O and I_{Ne} are the number of muons transferred to oxygen and to neon, respectively. They are determined by summing up the delayed Lyman series intensities of oxygen and neon, corrected for efficiency. The value $c_O \lambda_{pO}$ can thus be obtained without knowing neither the concentration of the impurity nor the transfer rate to oxygen. Its contribution in equation (3) is less than 4%. In the present case of oxygen, the transfer rate is known [24] such that the oxygen concentration can be determined. It amounts to about 46 ppm, *i.e.* less than 1% of the neon concentration. No other impurities have been found.

**Fig. 2.** Energy spectrum of the delayed muonic oxygen and neon Lyman series. The nuclear capture lines (n.c.) do not participate on the decay of the $(\mu p)_{1s}$ atoms.

The total muon transfer rate from protium to 4He is calculated from the measured $(\mu p)_{1s}$ lifetime by using (3) and (4). One obtains

$$\lambda_{p^4He} = (0.43 \pm 0.01_{stat.} \pm 0.04_{syst.}) \times 10^8 \text{ s}^{-1}.$$

One notes, that the systematical error dominates the statistical one. The main contribution to the former is not due to the uncertainty in the oxygen contamination, but to the uncertainty in the transfer rate to neon and the uncertainty in the neon density. Contributions due to the errors of total pressure, helium concentration, and λ_{pd} and $\lambda_{pp\mu}$ are small.

The result obtained with our triple gas mixture with neon as the third gas component is in agreement with the other experiments using the same method, namely Bystritsky *et al.* [9], Jacot-Guillarmod *et al.* [25] and Piller *et al.* [23] (see Tab. 2).

4 Conclusion

The knowledge of the muon transfer rates from hydrogen isotopes to helium is important for the recycling process of the muon in muon catalyzed fusion (μCF). Since helium is produced both in fusion reactions and in tritium decay, the helium atoms act as muon scavengers either by direct muon capture in helium or by muon transfer from hydrogen isotopes to helium, hence limiting the fusion yield. In addition to the interest in the μCF cycle, the molecular formation rate and its decay channels are a challenge for theoreticians. Indeed, considerable theoretical efforts have been made to understand the transfer from (μp) to helium and to calculate transfer rates and temperature dependencies.

In the experiment the total transfer rate λ_{pHe} , which is the sum of direct muon transfer rate λ_{pHe}^{dir} (1) and the

Table 2. Experimental and theoretical muon transfer rates from hydrogen to ^4He at room temperature (in units of 10^8 s^{-1}). The total transfer rate $\lambda_{p^4\text{He}}$ is the sum of the direct transfer rate $\lambda_{p^4\text{He}}^{\text{dir}}$ (1) and the molecular formation rate $\lambda_{p\mu^4\text{He}}$ (2).

mixture	lifetime measurement	yield measurement	theory	theory
	$\lambda_{p^4\text{He}}$	$\lambda_{p^4\text{He}}$	$\lambda_{p\mu^4\text{He}}$	$\lambda_{p^4\text{He}}^{\text{dir}}$
$\text{H}_2 + ^4\text{He} + \text{CH}_4$	< 0.6 [23]			
$\text{H}_2 + ^4\text{He}$		$0.44 \pm 0.16^{\text{a}}$ [14]	0.44 [6] $0.34\text{-}0.35$ [7] $0.43\text{-}0.52$ [8]	0.055 [4] ~ 0.001 [1]
$\text{H}_2 + ^4\text{He} + \text{Xe}$	0.36 ± 0.10 [9]			
$\text{H}_2 + ^4\text{He} + \text{Ar}$	0.51 ± 0.19 [25]			
$\text{H}_2 + ^4\text{He} + \text{Ne}$	$0.43 \pm 0.04^{\text{b}}$ [this work]			
average	$0.42 \pm 0.04^{\text{c}}$,			

^aDepending on the molecular decay rates used, [17,18,21], the rate varies between (0.44 ± 0.16) , (0.59 ± 0.22) and (0.68 ± 0.26) , respectively.

^bThe error is the rms error of statistical and systematical uncertainties.

^cAverage of the lifetime measurements [9,25] and the value of this work.

molecular formation rate $\lambda_{p\mu\text{He}}$ (2), is determined. From the lifetime measurements, namely Bystritsky *et al.* [9], Jacot-Guillarmod *et al.* [25], and the present experiment (see Tab. 2), a mean value for the transfer rate from hydrogen to ^4He is calculated with the result

$$\lambda_{p^4\text{He}} = (0.42 \pm 0.04) \times 10^8 \text{ s}^{-1}.$$

The yield measurement by von Arb *et al.* [14] depends crucially on the theoretical predictions for the different decay channels of the mesomolecule. Using the branching ratios calculated by Kravtsov *et al.* [18], the transfer rate obtained by von Arb *et al.* is in good agreement with the lifetime measurements.

If one neglects the direct muon transfer process (1) - for which only theoretical predictions exist - and if only transfer *via* molecular formation is considered, the experimentally obtained averaged value for $\lambda_{p^4\text{He}}$ is in good agreement with the theoretical predictions of Aristov *et al.* [6] and Kravtsov *et al.* [8]. However, taking direct muon transfer as calculated by Matveenko and Ponomarev [4] into account - which may contribute up to 13% to the total rate - the experimental rate $\lambda_{p^4\text{He}}$ is in better agreement with the molecule formation rate calculated by Ivanov *et al.* [7].

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