First measurement of the temperature dependence of muon transfer rate from the $\mu p$ atoms to oxygen

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We report the first measurement of the temperature dependence of muon transfer rate from $\mu p$ atoms to oxygen between 100 and 300 K. Data were obtained from the X-ray spectra of delayed events in gaseous target $H_2/O_2$ exposed to a muon beam. Based on the data, we determined the muon transfer energy dependence up to 0.1 eV, showing an 8-fold increase in contrast with the predictions of constant rate in the low energy limit. This work set constraints on theoretical models of muon transfer, and is of fundamental importance for the measurement of the hyperfine splitting of $\mu p$ by the FAMU collaboration.

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We present the results of a systematic experimental investigation of the energy dependence of the muon transfer rate from the ground-state muonic hydrogen atom $\mu p$ to oxygen. A precise knowledge of this process is of key importance for the final objective of the FAMU Collaboration, which is a measurement of the hyperfine splitting (HFS) of the $1S$ state of $\mu p$ by means of laser spectroscopy [1, 2].

In the experiment planned by the FAMU collaboration, a laser tuned to the hyperfine-splitting resonance energy induces the singlet-to-triplet transitions between the total-spin states $F = 0$ and $F = 1$ of the $1S$ muonic hydrogen, which is formed and then thermalized in a gaseous $H_2$ target. After the laser excitation, the $\mu p(F = 1)$ atom quickly de-excites back to the $F = 0$ state in collisions with protons bound in the surrounding $H_2$ molecules. As a result, the HFS energy is converted into additional $\mu p$ kinetic energy up to 0.1 eV with a wide distribution due the simultaneous rotational transitions in the hit $H_2$ molecules.

A clear and strong signal from the “laser-accelerated” neutral $\mu p$ atoms is necessary in order to perform a precise HFS measurement. Our idea is to use an admix-

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ture of a high-Z element in the H$_2$ target and register characteristic X-rays from the de-excitation of the high-Z muonic atom formed when the muon is transferred from $\mu p$. Since muon transfer takes place both from the thermalized and laser-accelerated $\mu p$ atoms, it is crucial to choose a higher-Z element which is characterized by a strong energy dependence of the low-energy muon-transfer rate below about 0.1 eV.

According to the PSI experiments [3–5], oxygen is a good candidate. Although these experiments were performed only at room temperature, an unexpected high value of the muon transfer rate was found for the epithermal 1S $\mu p$ atoms. The presence of energetic $\mu p$ atoms is due to the acceleration via atomic cascade [6, 7] after the negative muon capture in hydrogen and $\mu p$ formation in excited Coulomb states ($n \approx 14$). A strong dependence of the muon transfer rate to oxygen on collision energy was then pointed out in the quantum-mechanical calculations [8, 9].

The rough two-step function model of the energy-dependent muon transfer rate to oxygen, which was fitted to the PSI data [5], is inappropriate for the precise HFS measurement. Therefore, we decided to measure the muon transfer rate to oxygen from $\mu p$ atoms thermalized at different temperatures in a wide temperature interval (104–300 K). The obtained results allows us to test the available theoretical calculations of the muon transfer rate and will be used to choose optimal conditions for the HFS measurement.

The FAMU apparatus is built around a pressurized and thermalized gas target contained in an aluminium vessel kept at thermal equilibrium and surrounded by X-rays detectors. The FAMU experimental method requires a detection system suited for time resolved X-ray spectroscopy [2]. Eight scintillating counters based on Lanthanum Bromide crystals read by photomultipliers are used. Detectors output signals are registered to measure both energy and time spectrum of the recorded events. The experiment took place at the RIKEN muon facility of the Rutherford Appleton Laboratory (UK) [10]. Muons are produced in bunches with a repetition rate of 50 Hz. More details about the FAMU apparatus can be found in [11].

The data used in this analysis were acquired using a gas mixture of H$_2$ and O$_2$ with the oxygen concentration of 190 ppm. The target was filled at room temperature at 41 bar and subsequently brought to six different temperatures, from 300 to 104 K degrees. The lowest temperature was significantly higher than the oxygen condensation point for the mixture, 54 K, obtained from [12]. Each temperature was kept stable for a data taking time of three hours.

An accurate determination of the muon transfer rate as a function of kinetic energy requires measurements in steady-state conditions, when the kinetic-energy distribution of $\mu p$ atoms is well known. The distribution of $\mu p$ initial energy, after the muon capture in H$_2$ and subsequent atomic cascade, is broad and can cover hundreds eV depending on target density [6, 7, 13]. The shape of this distribution is not well known. Therefore, we have measured the muon transfer rate at times after the slowing down and thermalization of $\mu p$ atoms, when their kinetic energies are described by the Maxwell-Boltzmann distribution for a given target temperature. The data set was taken with a target number density which enabled fast thermalization of $\mu p$ atoms (≤ 150 ns) and rapid quenching of the initial population of the spin state $F = 1$ (≈ 10 ns) [14]. The 190 ppm concentration of O$_2$ in our gas mixture was chosen in order to have a long (≥ 150 ns) mean life time of $\mu p$ atoms. This allowed us a long-time observation of the muon-transfer process from $\mu p$ atoms thermalized at the chosen temperature.

The muonic oxygen atoms are formed in excited Coulomb states. They promptly de-excite by emitting characteristic X-rays. A measurement of the muon transfer rate is performed by studying the time evolution of muonic-oxygen lines. The muon-transfer process was studied at times beyond the end of the muon pulse and much larger than the prompt emission and $\mu p$ thermalization time. This way the large background of X-ray prompt emission due to the interaction between muons and all the elements of the target — mostly aluminium, nickel, gold, and carbon — was strongly suppressed and thus was neglected.

At such times, at temperature $T$, the variation of the number $N_{\mu p}$ of $\mu p$ atoms in target in the time interval $dt$ is:

$$dN_{\mu p}(t) = -N_{\mu p}(t)\lambda_{\text{dis}}(T)dt,$$  

(1)

where $\lambda_{\text{dis}}(T)$ is the total disappearance rate of the muonic hydrogen atoms at temperature $T$, given by:

$$\lambda_{\text{dis}}(T) = \lambda_0 + \phi [c_p \Lambda_{pp\mu} + c_d \Lambda_{pd}(T) + c_O \Lambda_{pO}(T)],$$  

(2)

Here $\lambda_0 = (4665.01 \pm 0.14) \times 10^2$ s$^{-1}$ [15, 16] is the rate of disappearance of the muons bounded to proton (that includes both muon decay and nuclear capture), $\Lambda_{pp\mu} = 2.01 \times 10^8$ s$^{-1}$ [15] is the formation rate of the $pp\mu$ molecular ion in collision of $\mu p$ with a hydrogen nucleus (normalized to liquid hydrogen density, LHD, $N_0 = 4.25 \times 10^{22}$ atom/cm$^3$), $\Lambda_{pd}(T) \approx (1.6 \pm 0.3) \times 10^{10}$ s$^{-1}$ denotes the muon transfer rate from $\mu p$ to deuterium as function of temperature [17], and $\Lambda_{pO}(T)$ is the muon transfer rate from $\mu p$ to oxygen atom. The number density of the atoms in the target gas is $\phi = (4.869 \pm 0.003) \times 10^{-2}$ in LHD units, and $c_p$, $c_O$, and $c_d$ are the number concentrations of hydrogen, oxygen, and deuterium respectively. We used hydrogen with measured natural deuterium abundance $c_d = (1.358 \pm 0.001) \times 10^{-4}$ [18]. The oxygen concentration was $c_O = (1.90 \pm 0.04) \times 10^{-4}$ and $c_p = 1 - c_O - c_d$. Let us note that the rate of nonresonant $pp\mu$ formation is practically constant below 0.1 eV [19]. Small changes of the muon transfer rate $\Lambda_{pd}(T)$ are due to the electron-screening corrections, which have been calculated according to Ref. [20].

In this analysis, only the steady-state delayed X-ray events due to the thermalized atoms are considered. The
only unknown variable, the transfer rate \( \Lambda_{pO}(T) \), is determined using Eqs. (1) and (2) by fitting numerically the oxygen X-rays time evolution at temperature \( T \) and leaving \( \Lambda_{pO} \) a free parameter. Details about the method can be found in Ref. [21].

The experimental sample, used in this work, consists of about \( 2.6 \times 10^6 \) muon triggers, and corresponds to \( \approx 7.8 \times 10^7 \) reconstructed X-rays.

The six target temperature were: 104, 153, 201, 240, 272, and 300 K. Two sensors [11] were used to measure the target temperature. The time needed to stabilize the system at a preset temperature varied from half hour to about an hour and a half, depending on the preset temperature and on external conditions.

X-rays signals were identified and reconstructed using a fitting procedure of the detectors waveforms. A clean data sample was obtained by applying light selection criteria on the reduced \( \chi^2 \) and on the distance between two consecutive reconstructed signals. Selection efficiencies and fractional live time were estimated and taken into account in the study of the oxygen lines time evolution. The combined efficiencies and fractional live time were about 95\%, constant above 2000 ns, and smoothly decreased to about 92\% at 1200 ns. A detailed discussion of the data selection and selection efficiencies can be found in Ref. [21]. For each temperature, the energy spectrum of delayed events was studied as function of time. The delayed time window from 1200 to 10000 ns from trigger was split into 20 bins of increasing width, the narrowest being \( \approx 140 \) ns. The time resolution of the reconstructed events was better than 1 ns, hence any effect on neighbour bins was negligible and no time deconvolution was needed.

An estimation of the background below the oxygen-line signal was the most important aspect of the data analysis. Eventually, the background was estimated for each time and temperature bin using the data taken with a pure \( \text{H}_2 \)-gas target and with the same selection criteria. Figure 1 shows the energy spectra at 104 K in two different time bins. Dotted lines represent the background (mostly induced by electrons from muon decay) estimated using a target of pure hydrogen gas. Signal after background subtraction is drawn as a shaded area. The tails towards lower energy are due to the detector behaviour [21]. The total number of detected oxygen X-rays from 100 to 200 keV varied from about ten thousands to few hundreds, depending on temperature and time bin. Fluctuations due to the normalization of the background and to the type of background subtraction were considered as a systematic error [21]. This systematic error was quadratically summed to the statistical error for each signal spectrum (see, e.g., numbers reported in Fig. 1), before performing the fit to the data that provides the transfer-rate measurement.

The time evolution of the oxygen line X-ray spectra, at the two temperature limits of 300 K (green triangles) and at 104 K (blue circles), is shown in Fig. 2. Each point represents the integrated signal, after the background subtraction and efficiency correction, divided by the time bin width. The fit was performed between 1200 ns, when the thermalized phase started, and 6500 ns from trigger, when the signal was still detectable above the background. It can be noticed that the slopes of the two dis-
tributions are significantly different, which correspond to the different transfer rates as functions of temperature.

The experimental results are presented in Tab. I. During measurements the temperature was kept stable, with less than 0.1\% variation around the mean value $T$ (column 1), to which the measured rate $\Lambda_{pO}$ (column 2) is referred.

TABLE I. Summary of transfer rate from muonic hydrogen to oxygen results. The first and second errors represent the statistical with background related systematics and other systematic uncertainties, respectively.

| Temperature $T$ [K] | $\Lambda_{pO}(T)$ [$10^{10}$ s$^{-1}$] |
|---------------------|---------------------------------|
| 104                 | $3.25 \pm 0.10 \pm 0.07$        |
| 153                 | $5.00 \pm 0.11 \pm 0.10$        |
| 201                 | $6.38 \pm 0.10 \pm 0.13$        |
| 240                 | $7.62 \pm 0.12 \pm 0.16$        |
| 272                 | $8.05 \pm 0.12 \pm 0.17$        |
| 300                 | $8.68 \pm 0.12 \pm 0.18$        |

Using the MINUIT package [22], we worked out a series of smooth fits $\Lambda_{pO}(T; P)$ to the FAMU experimental data that involve up to three parameters, and labelled them as fit $A$–$E$. The explicit form and the numerical values of the parameters are given in Tab. II. Of these fits we selected for further use the second degree polynomial fit $A$ for its simplicity and very good $\chi^2/ndf = 1.13/3$.

$\Lambda_{pO}(T) \approx \Lambda_{pO}(T; P) = P_1 + P_2T + P_3T^2$. (3)

Figure 3 shows the experimental data together with the fit $A$. The error bars represent both errors reported in Tab. I quadratically summed. The analysis of systematic errors showed that the main source of uncertainty derives from the gas composition. Other sources of systematic effects, including temperature and pressure measurements, timing, parameters error propagation, were proved to be negligible [21]. Results are in perfect agreement with the PSI measurement at 294 K [5].

The main goal of our investigations is the quantitative determination of the muon transfer rate $\lambda_{pO}(E)$ to oxygen as function of the center-of-mass (CM) collision energy of $\mu p$ with the oxygen nucleus. Thus, we relate $\lambda_{pO}(E)$ to $\Lambda_{pO}(T)$ by

$$\Lambda_{pO}(T) = \int_0^{\infty} dE f(E; T) \lambda_{pO}(E),$$  (4) where $f(E; T)$ is the (unknown in the general case) density of kinetic energy distribution of the $\mu p$ atoms.

FAMU experimental set-up and event selection criteria guarantee that data are obtained at thermal equilibrium, when $f(E; T)$ is nothing but the Maxwell-Boltzmann distribution:

$$f(E; T) = f_{MB}(E; T) = 2\sqrt{E/\pi(k_BT)^{-3/2}} \exp(-E/k_BT).$$

This allows to compute $\Lambda_{pO}(E)$ as solution of the integral equation Eq. (4). Following the same path, we compute the approximant $\lambda_{pO}(E; p)$ to $\lambda_{pO}(E)$ (where $p$ denotes the set of parameters) from the equation

$$\Lambda_{pO}(T; P) = \int_0^{\infty} dE f_{MB}(E; T) \lambda_{pO}(E; p).$$  (5)

The specific choice of the functional form $A$ (3) of the fit $\Lambda_{pO}(T; P)$ yields the following explicit form of the approximant $\lambda_{pO}(E; p)$:

$$\lambda_{pO}(E) \approx \lambda_{pO}(E; p) = p_1 + p_2E + p_3E^2,$$

$$p_i = \pi_i P_i, \quad \pi_i = \frac{1}{(2i-1)!!} \left( \frac{2}{k_B} \right)^{i-1}. $$  (6)

The numerical values of the parameters are given in Tab. II (fit $A$).

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$$\lambda_{pO}(E) \approx \lambda_{pO}(E; p) = p_1 + p_2E + p_3E^2,$$

$$p_i = \pi_i P_i, \quad \pi_i = \frac{1}{(2i-1)!!} \left( \frac{2}{k_B} \right)^{i-1}. $$  (6)

The numerical values of the parameters are given in Tab. II (fit $A$).

TABLE II: Explicit form of five alternative fits of the temperature dependence of the muon transfer rate $\Lambda_{pO}(T; p)$, of the corresponding energy-dependence functions $\lambda_{pO}(E; p)$, related to the former through Eq. (5), and the chi-squared criterion $\chi^2/ndf$ for each of the fits $\Lambda_{pO}(T; p)$.

| Fit | Analytical expressions and $\chi^2$ | Optimal values of the parameters |
|-----|---------------------------------|-------------------------------|
| $A$ | $\lambda_{pO}(E; p) = \sum_{k=1}^{3} p_k E^{k-1}$ | $p_1 = (-1.32 \pm 0.61) \times 10^{10}$ s$^{-1}$ |

FIG. 3. Fit to the FAMU data (circles) using a parabolic dependence of the muon transfer rate on temperature $T$ (solid line). PSI measurement [5] is also shown (triangle).
The main uncertainties of the approximant $\Lambda_{pO}(E; p)$ are: (a) the statistical uncertainty $\sigma(E)$ due to the statistical uncertainties $\sigma_i$, $i = 1, \ldots, 6$ of the $i$-th experimental value $\Lambda_{pO}(T_i)$, ($\sigma_i$ being the square root of the sum of the errors given in Tab. I, squared), and (b) the model uncertainty, related to the arbitrariness in the choice of the fitting functions. The specific functional form of fit A (3) allows to put the statistical uncertainty $\sigma(E)$ in a closed form:

$$\sigma(E) = \sqrt{\sum_{i=1}^{6} (c_i(E))^2},$$

$$c_i(E) = \sum_{j,k=1}^{3} \pi_k E^{k-1} M_{kj}^{-1} T_i^{-1}, \quad (7)$$

$$M_{kj} = \sum_{i=1}^{6} \frac{T_i^{k+j-2}}{\sigma_i^2}.$$ 

To estimate the model uncertainty we considered fitting functions with very diverse behavior outside the range of interest: unrestricted non-singular behavior in the limit $E \to 0$, and from exponential decrease to power growth at large energies $E \gg 0.1$ eV (fits A–E in Tab. II). While providing very similar temperature dependence $\Lambda_{pO}(T; p)$ for $100 < T < 300$ K (see Fig. 4, left), the corresponding $\Lambda_{pO}(E; p)$ happen to differ by up to 20% (Fig. 4, right). Accordingly, we take half of the width of the envelope of the ensemble of curves A–E as estimate of the model uncertainty of the extracted energy dependence of the muon transfer rate to oxygen. Note that the diverging behavior of the various approximants A–E at $E > 0.1$ eV does not exceed a few percent at $T = 300$ K and decreases fast with decreasing temperature. Similarly, the contribution from the range $E < 0.01$ eV does not exceed 10% for $T \sim 100$ K (with the exception of fit B), and is much less at higher temperatures, so that the unphysical negative values of the transfer rate at very low energies can simply be discarded by truncating the integral at 0.01 eV. These considerations prove that the FAMU experimental data in the temperature range $100 < T < 300$ K provide a sufficiently firm ground for the quantitative determination of the energy dependence of the muon transfer rate to oxygen for energies $0.01 < E < 0.1$ eV. This very range is of importance for the experimental measurement of the hyperfine splitting in the ground state of muonic hydrogen.

The results of the present investigation are presented on Fig. 5. The best fit A, $\Lambda_{pO}(E; p)$ Tab. II, is plotted in the interval from 0.01 to 0.1 eV, together with the statistical and model uncertainty bands, as well as the theoretical functions from Refs. [8, 9] and the “two-step function” fit of Ref. [5]. The selected energy interval essentially covers the range of kinetic energy of both the $\mu p$ atoms accelerated after depolarization in collision with a target gas hydrogen molecule, and the atoms thermalized at 100 K. Our fit is not supposed to be valid at energies above 0.1 eV and below 0.01 eV; the reason is that the contributions to the measured transfer rate $\Lambda_{pO}(T; P)$ from the higher-energy tail of the MB distribution and from the lowest-energy interval, where this distribution steeply tends to zero, are very small. As long as the goal of the present paper was to experimentally determine the energy dependence of the muon transfer rate to oxygen, we did not take into account the available results of theoretical calculations in the fitting procedure, and added them to Fig. 5 just for comparison.

Figure 5 shows that the advanced quantum-mechanical three-body calculations of Refs. [8] and [9] do not re-
produce correctly the energy dependence of the rate of muon transfer to oxygen in the low collision energy limit, though Ref. [9], in which electron screening effects are accounted for in an approximate way, describes qualitatively the monotonous growth of the rate with energy up to 0.1 eV. The detailed analysis of the applicability of these theoretical approaches, however, is beyond the scope of the present work. As for the two-step function model [5], it is supposed to reflect the higher muon transfer rate from epithermal muonic atoms compared to the lower rate from the $\mu p$ atoms thermalized at $\sim 300$ K.

In the investigation of the temperature dependence of the muon-transfer process from the thermalized $\mu p$ atoms to oxygen, we have observed for the first time a strong monotonic rise of the rate $\Lambda_{\text{pO}}(T)$ in the temperature interval 104–300 K. This rise is well-fitted by a parabolic function of temperature. Based on the data, we have quantitatively determined the energy dependence of the transfer rate $\lambda_{\text{pO}}(E)$, which increases by a factor of about eight in the collision-energy interval 0.01–0.08 eV. Such a strong change enables us to employ the muon transfer rate to oxygen as a signature of the kinetic-energy gain of the $\mu p$ atom, in the planned FAMU measurement of the hyperfine splitting of 1S state of this atom.

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