



First measurement of the temperature dependence of muon transfer rate from muonic hydrogen atoms to oxygen

E. Mocchiutti^{a,*}, A. Adamczak^b, D. Bakalov^c, G. Baldazzi^d, R. Benocci^{e,f}, R. Bertoni^e, M. Bonesini^{e,g}, V. Bonvicini^a, H. Cabrera Morales^a, F. Chignoli^e, M. Clemenza^{e,g}, L. Colace^{h,i}, M. Danailov^{a,j}, P. Danev^c, A. de Bari^{k,l}, C. De Vecchi^l, M. De Vincenzi^{h,m}, E. Furlanetto^{a,n}, F. Fuschino^{d,o}, K.S. Gadedjisso-Tossou^{a,p,q}, D. Guffanti^{a,r}, K. Ishida^s, C. Labanti^{d,o}, V. Maggi^{e,f}, R. Mazza^e, A. Menegolli^{k,l}, G. Morgante^{d,o}, M. Nastasi^{e,g}, J. Niemela^p, C. Pizzolotto^a, A. Pullia^{t,u}, R. Ramponi^{t,v}, L.P. Rignanese^d, M. Rossella^l, N. Rossiⁿ, M. Stoilov^c, L. Stoychev^{a,p}, L. Tortora^h, E. Vallazza^a, G. Zampa^a, A. Vacchi^{a,n,s}

^a Sezione INFN di Trieste, via A. Valerio 2, Trieste, Italy

^b Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, PL31342 Kraków, Poland

^c Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences, blvd. Tsarigradsko ch. 72, Sofia 1142, Bulgaria

^d Sezione INFN di Bologna, viale Berti Pichat 6/2, Bologna, Italy

^e Sezione INFN di Milano Bicocca, Piazza della Scienza 3, Milano, Italy

^f Dipartimento di Scienze dell'Ambiente e della Terra, Università di Milano Bicocca, Piazza della Scienza 1, Milano, Italy

^g Dipartimento di Fisica G. Occhialini, Università di Milano Bicocca, Piazza della Scienza 3, Milano, Italy

^h Sezione INFN di Roma Tre, Via della Vasca Navale 84, Roma, Italy

ⁱ Dipartimento di Ingegneria, Università degli Studi Roma Tre, Via V. Volterra 62, Roma, Italy

^j Sincrotrone Elettra Trieste, SS14, km 163.5, Basovizza, Italy

^k Dipartimento di Fisica, Università di Pavia, via A. Bassi 6, Pavia, Italy

^l Sezione INFN di Pavia, Via A. Bassi 6, Pavia, Italy

^m Dipartimento di Matematica e Fisica, Università di Roma Tre, Via della Vasca Navale 84, Roma, Italy

ⁿ Dipartimento di Scienze Matematiche, Informatiche e Fisiche, Università di Udine, via delle Scienze 206, Udine, Italy

^o INAF-OAS Bologna, via P. Gobetti 93/3, Bologna, Italy

^p The Abdus Salam International Centre for Theoretical Physics, Strada Costiera 11, Trieste, Italy

^q Laboratoire de Physique des Composants à Semi-conducteurs (LPCS), Département de physique, Université de Lomé, Lomé, Togo

^r Gran Sasso Science Institute, via F. Crispi 7, L'Aquila, Italy

^s Riken Nishina Center, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

^t Sezione INFN di Milano, via Celoria 16, Milano, Italy

^u Dipartimento di Fisica, Università degli Studi di Milano, via Celoria 16, Milano, Italy

^v INFN-CNR, Dipartimento di Fisica, Politecnico di Milano, piazza Leonardo da Vinci 32, Milano, Italy

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ABSTRACT

We report the first measurement of the temperature dependence of muon transfer rate from muonic hydrogen atoms to oxygen between 100 and 300 K. Data were obtained from the X-ray spectra of delayed events in a gaseous target, made of a H₂/O₂ mixture, exposed to a muon beam. This work sets constraints on theoretical models of muon transfer and is of fundamental importance for the measurement of the hyperfine splitting of muonic hydrogen ground state as proposed by the FAMU collaboration.

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1. Introduction

We present the results of a systematic experimental investigation of the temperature dependence of the muon transfer rate from the ground-state muonic hydrogen atom μp to oxygen. A

* Corresponding author.

E-mail address: Emiliano.Mocchiutti@ts.infn.it (E. Mocchiutti).

precise knowledge of this process is of key importance for the final objective of the FAMU Collaboration – the measurement of the hyperfine splitting (HFS) of the $1S$ state of μp 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 singlet-to-triplet transitions between the total-spin states $F = 0$ and $F = 1$ of $1S$ 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 μp kinetic energy up to 0.12 eV with a wide distribution due to the simultaneous rotational transitions in the affected H_2 molecules.

A clear and strong signal from the “laser-accelerated” neutral μp atoms is necessary in order to perform a precise HFS measurement. Our idea is to use an admixture of a high- Z element in the H_2 target and detect characteristic X-rays from the de-excitation of the high- Z muonic atom formed when the muon is transferred from μp . Since muon transfer takes place both from the thermalized and laser-accelerated μp atoms, it is crucial to choose a higher- Z element which is characterized by a strong energy dependence of the 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 unexpectedly high value of the muon transfer rate was found for the $(\mu p)_{1S}$ atoms not yet thermalized (“epithermal”). Energetic μp atoms are due to the acceleration via atomic cascade [6,7] after the negative muon capture in hydrogen and μp formation in excited Coulomb states ($n \approx 14$). A strong dependence of the muon transfer rate to oxygen to the collision energy was also anticipated in the quantum-mechanical calculations reported in Ref. [8,9].

The PSI data on the energy-dependent muon transfer rate to oxygen were fitted with a two-step function of the collision energy [5]. This function, however, is inappropriate for modeling the HFS measurement because (a) the fit is too rough an approximation to the data and (b), most important, it provides no information about the energy dependence of the transfer rate in the energy range below the threshold of 0.12 eV (corresponding to temperatures ~ 1000 K), which in fact is the range of experimental interest. This information can only be obtained by measurements of the muon transfer rate to oxygen from μp thermalized atoms in a sufficiently wide temperature range below 1000 K, as performed in the present work.

In thermal equilibrium at temperature T , the observable transfer rate $\Lambda_{pO}(T)$ is expressed in terms of the function that describes the dependence of the transfer rate on the collision energy E by means of the relation

$$\Lambda_{pO}(T) = \int_0^{\infty} dE f_{MB}(E; T) \lambda_{pO}(E), \quad (1)$$

where

$$f_{MB}(E; T) = 2\sqrt{E/\pi} (k_B T)^{-3/2} e^{-E/(k_B T)} \quad (2)$$

is the Maxwell-Boltzmann distribution, with k_B the Boltzmann constant. The main contribution to the integral in the right-hand side of Eq. (1) comes from the region around $E = k_B T$; therefore the measurement of $\Lambda_{pO}(T)$ in the range between 100 and 300 K will provide important information about the behavior of $\lambda_{pO}(E)$ for energies in and around the range $0.01 < E < 0.05$ eV.

The obtained results allow us to test the available theoretical calculations of the muon transfer rate and will be used to choose optimal conditions for the FAMU HFS measurement.

2. Experiment

The FAMU apparatus, used for the transfer rate measurement, is built around a pressurized and thermalized gas target contained in an aluminium vessel kept at thermal equilibrium and surrounded by X-ray detectors. The FAMU experimental method requires a detection system suited for time resolved X-ray spectroscopy [2]. Eight scintillating counters are used, based on 1” Lanthanum Bromide crystals and read by Hamamatsu high speed photomultipliers. Data are acquired and processed by 500 MHz CAEN V1730 digitizers, within the framework of the FAMU Data Acquisition System [10]. Output signals from the detectors provide measurements of both the energy and time spectrum of the recorded events. In this configuration, the detectors provide a good resolution, about 8.5% (FWHM) at 133 keV, and an excellent time resolution, signal rise time of about 12 ns [11]. The experiment was performed at the RIKEN muon facility of the Rutherford Appleton Laboratory (UK) [12], using muons 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 an oxygen concentration c_O of 190 ppm. The target was filled at room temperature to 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 Ref. [13]. A thermally insulated vertical tube provided external connection to the target. After filling, the target was sealed by a valve placed just outside the cryogenic vessel. The portion of tube between the target and the valve had a volume about ten times smaller than the target itself. Due to gravity, the temperature gradient inside the tube ranged from room temperature, at the valve, to the gas target temperature, at the vessel. The variation of density inside the target was sufficiently small that any associated systematic effects were negligible.

An accurate determination of the muon transfer rate as a function of kinetic energy requires measurements under steady-state conditions, when the kinetic-energy distribution of μp atoms is well known. The distribution of μp initial energy, after the muon capture in H_2 and subsequent atomic cascade, is broad and can cover hundreds of eV depending on the target density [6,7,14,15]. The shape of this distribution is not well known. Therefore, we have measured the muon transfer rate only after the slowing down and thermalization of μp atoms, when their kinetic energies are described by the Maxwell-Boltzmann distribution for a given target temperature.

Data were taken with a target number density which enabled fast thermalization of μp atoms (many times faster than the muon lifetime) and rapid quenching of the initial statistical population of the spin state $F = 1$. For H_2 gas at 41 bar and 300 K, the Monte Carlo simulation gives a thermalization time of 150 ns and a quenching time of 10 ns [16]. In order to observe the characteristic X-rays from muonic oxygen over long times, the oxygen concentration for a given target density cannot be too high. For $c_O = 190$ ppm at this temperature and pressure, the average muon transfer rate from the thermalized μp atoms to oxygen is $0.78 \times 10^6 \text{ s}^{-1}$ (using the experimental rate of $8.5 \times 10^{10} \text{ s}^{-1}$ at liquid-hydrogen density [5]), which is comparable with the muon decay rate. This choice enabled us to observe the muon-transfer process from thermalized μp atoms for several microseconds.

Muonic oxygen atoms are formed in excited Coulomb states, which 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 μp thermalization time. In

this way the large background of X-ray emissions due to the interaction between muons and all the elements of the target – mostly aluminium, nickel, gold, and carbon – was strongly suppressed and could be neglected.

Under these conditions, at a given temperature, T , the variation of the number $N_{\mu p}$ of μp atoms in the target in the time interval dt is given by:

$$dN_{\mu p}(t) = -N_{\mu p}(t)\lambda_{\text{dis}}(T) dt, \quad (3)$$

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

$$\lambda_{\text{dis}}(T) = \lambda_0 + \phi [c_p \Lambda_{pp\mu} + c_d \Lambda_{pd}(T) + c_o \Lambda_{pO}(T)]. \quad (4)$$

Here $\lambda_0 = (4665.01 \pm 0.14) \times 10^2 \text{ s}^{-1}$ [17,18] is the disappearance rate of the muons bound to protons (that includes both muon decay and nuclear capture), $\Lambda_{pp\mu} = 2.01 \times 10^6 \text{ s}^{-1}$ [17] is the formation rate of the $pp\mu$ molecular ion in collisions of μp with a hydrogen nucleus (normalized to liquid hydrogen density, LHD, $N_0 = 4.25 \times 10^{22} \text{ atom/cm}^3$) and $\Lambda_{pO}(T)$ is the muon transfer rate from μp to oxygen atoms. The muon transfer rate $\Lambda_{pd}(T)$ from μp to deuterium, bound mostly in HD molecules, varies from $8.65 \times 10^9 \text{ s}^{-1}$ at 100 K to $8.20 \times 10^9 \text{ s}^{-1}$ at 300 K (normalized to $2.125 \times 10^{22} \text{ HD molecules/cm}^3$). The rate Λ_{pd} for muon transfer to bare deuterium nuclei is practically constant at the lowest energies [19]. The energy-dependent electron screening in hydrogenic molecules [20] leads to the above appreciable change of $\Lambda_{pd}(T)$ in the considered temperature interval. 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 $c_d = (1.358 \pm 0.001) \times 10^{-4}$ [21].¹ 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 [22]. The μd atoms, which are created via the muon transfer from μp atoms to the small natural admixture of deuterium, have an initial energy of about 42 eV. They are never thermalized since a deep Ramsauer-Townsend minimum in the cross section of scattering $\mu d + H_2$ is present at 7 eV [23]. As a result, for target densities corresponding to 300 K hydrogen at 41 bar, the mean kinetic energy of μd atoms is 26 eV and only a very small fraction of these atoms is thermalized. The muon transfer rate Λ_{dO} from μd atoms to oxygen is negligible at energies $\gtrsim 1$ eV, as confirmed by the absence of corresponding X-ray spectra from muonic oxygen at short times in the experiment performed in a pure D_2 target with a small admixture of SO_2 [24]. Therefore, a contribution to the X-ray spectra due to the muon transfer from μd atoms to oxygen is neglected in the present analysis. Our Monte Carlo simulation, which used the experimental results of Ref. [24] for the rate Λ_{dO} at thermal, epithermal and higher energies, has shown that this contribution is smaller than 1% for times up to several microseconds.

3. Data analysis

In our analysis, only the steady-state delayed X-ray events due to the thermalized atoms are considered. The only unknown variable is the transfer rate $\Lambda_{pO}(T)$, which is determined using Eqs. (3)

and (4) by numerically fitting the time evolution of the oxygen X-rays at temperature T and leaving Λ_{pO} a free parameter. Details about the method can be found in Ref. [25].

The experimental sample used in this work consists of about 2.6×10^6 muon triggers, and corresponds to $\approx 7.8 \times 10^7$ reconstructed X-rays. The time, in nanoseconds, associated to each X-ray is relative to the trigger generated by the accelerator and beam control system. In this time reference frame, the two muon spill bunches peak at about 530 and 850 ns. Each muon spill bunch consists of about 10^3 muons.

Data were taken at six target temperatures: 104, 153, 201, 240, 272, and 300 K. Two sensors were used to measure the target temperature. The cryogenic system was able to keep the temperature stable by limiting fluctuations to about 10 mK/h, within the systematic errors of the temperature sensors. More details about the cryogenic system can be found in [11]. Each temperature was kept stable for an acquisition time of three hours.

X-ray signals were identified and reconstructed using a fitting procedure on the detector waveforms. A clean data sample was obtained by applying light selection criteria based on the reduced χ^2 of the waveform fit and on the distance between two consecutive reconstructed signals. The reduced χ^2 selection ($\bar{\chi}^2 < 100$) was used to reject not-converged fit and poorly reconstructed events. The requirement on the distance between two consecutive signals to be greater than 30 ns was chosen by testing the reconstruction algorithm by means of a GEANT4 simulation. The simulation showed that when two peaks are separated by more than 30 ns the software reconstruction efficiency and accuracy (correct energy reconstruction) is better than 99.9%. Selection efficiencies and fractional live time were estimated and taken into account in the analysis of the time evolution of the oxygen lines. 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. [25]. For each temperature, the energy spectrum of delayed events was studied as a function of time. The delayed time window – from ≈ 1200 to 10000 ns from the trigger – was split into 20 bins of increasing width, the narrowest being ≈ 140 ns. The time resolution of the reconstructed events was better than 1 ns, hence any migration effect on neighboring 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. The background was estimated for each time and temperature bin using the data taken with a pure H_2 -gas target and with the same selection criteria and same gas condition, but with a smaller statistical sampling.

Fig. 1 shows the energy spectra at 104 K in two different time bins. These energy spectra were obtained by summing the calibrated energy spectra of each single LaBr detector. Dotted lines represent the background (mostly induced by electrons from muon decay) estimated using a target of pure hydrogen gas. Due to the lower statistics, the fluctuations in the background spectra were higher, hence, the spectra were smoothed using a Gaussian kernel algorithm [26]. The background was normalized to the data, for each time bin and energy spectrum, in the region between 250 and 350 keV. In the figure, the signal after background subtraction is drawn as a shaded area. The tails towards lower energy are due to energy leakage from the LaBr crystal. This effect was studied by means of GEANT4 simulations and corresponds to X-rays hitting the crystals on the border, with an incoming direction almost parallel to the surface. The total number of detected oxygen X-rays in the range 100 to 200 keV varied from about ten thousands to few hundreds, depending on target temperature and time bin. Systematic errors due to the normalization of the background were estimated by using a statistical approach on the number of

¹ A sample bottle of the same batch of pure hydrogen used to produce the mixture was provided by the gas supplier. This sample gas was tested using a precise mass spectrometer to determine the deuterium abundance. Unfortunately, the same study could not be done with the gas mixture used during the acquisition to determine better the oxygen abundance.

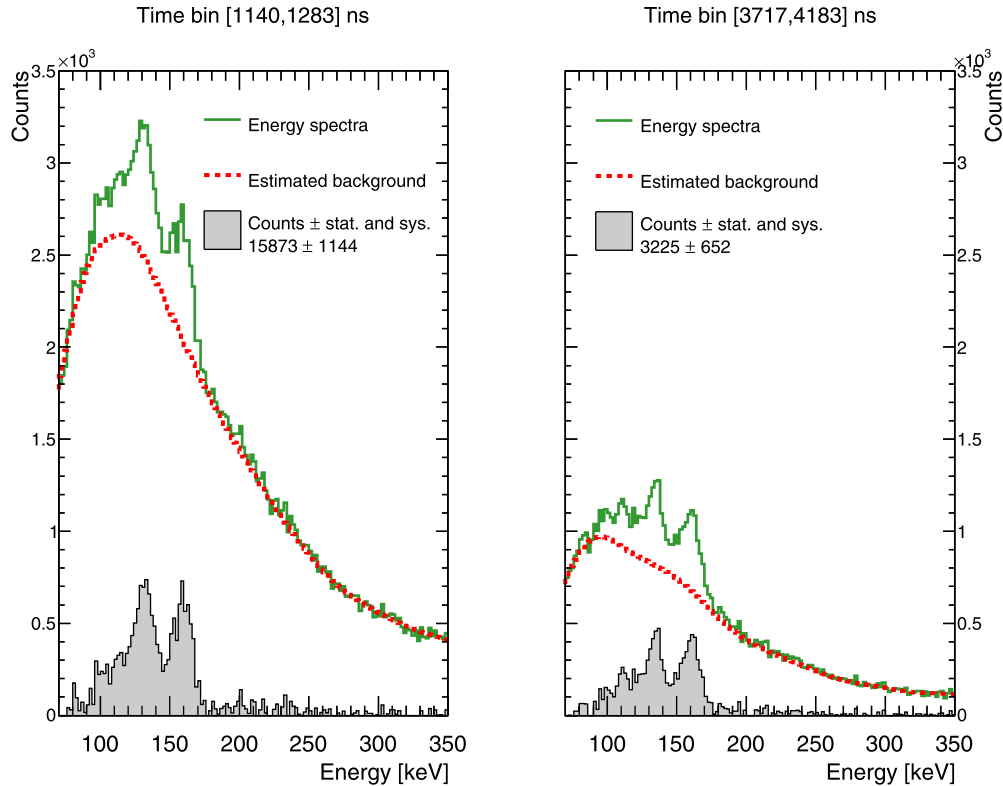


Fig. 1. Energy spectra at 104 K in two different time bins: [1140,1283] ns in the left panel and [3717,4183] ns in the right panel. Solid green line: energy spectra; dotted red line: estimated background; shaded area: signal after background subtraction. (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

both background and signal events in the normalization interval. A further consideration of systematic errors in the shape of the background was estimated by comparing the pure hydrogen background distribution with analytical functions fitted to the data and by using gas target mixture with different composition (i.e., CO₂ in hydrogen and Ar in hydrogen). The overall 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.

Notice that, due to the background subtraction, a time measurement cannot be associated to each oxygen x-ray, i.e., it is not possible to tag the single event. Hence, after background subtraction in a given time bin, there is not an average time measurement coupled to the signal spectrum.

The time evolution of the oxygen line X-ray spectra, at the two temperature limits of 300 K (red circles) and at 104 K (blue circles), is shown in Fig. 2 and reported in Table 1 for all the temperatures. Each point represents the integrated signal, after the background subtraction and efficiency correction, divided by the time bin width. Data points are centered in the time bins and error bars on the x axis represent the time bin width. To perform a fit of the data versus time, a step-like fit function was used: the fit function is an exponential function, which is integrated within each time bin that was defined for the collected data. Function and data can then be considered as two histograms, which can be compared directly. For the purposes of this quantitative comparison the question of choosing the correct time value within the bin is irrelevant. The fit was performed starting at ≈ 1200 ns, about 350 ns after the second muon spill in order to take into account only the thermalized phase, given a thermalization time of 150 ns. The upper limit of the time window was chosen according to the available statistics: data points were used if the measured integrated signal was greater than three times the associated error.

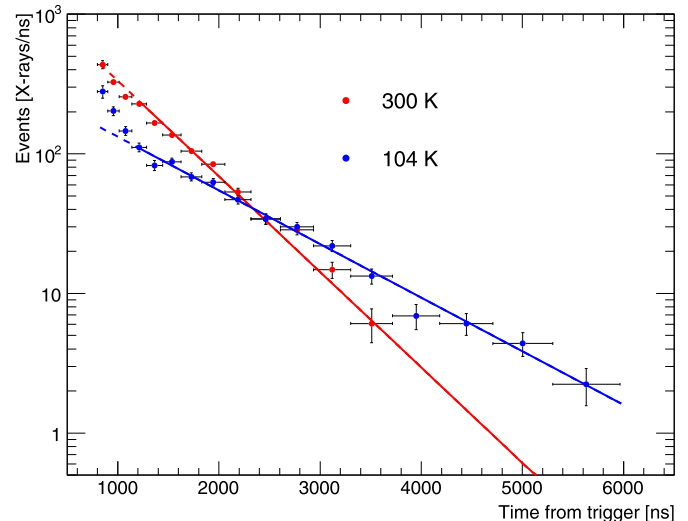


Fig. 2. Time dependence of oxygen line intensities at 300 K and 104 K. Two exponential functions (solid lines) correspond to the transfer rate fit. Dashed lines are an extrapolation of the fit at shorter times. Vertical error bars associated to the points include the statistical and background-systematic errors. Horizontal error bars represent the bin width.

Fig. 2 shows that the slopes of the two distributions differ significantly, corresponding to the different transfer rates as functions of temperature. Solid lines represent the exponential transfer rate obtained by the fit. The comparison between data points and curves has to be done considering the bin width (error bars on the x axis). Dashed lines are the extrapolation of the fit to shorter times. The first three data points were not included in the fit since they are too close to the muonic hydrogen production, hence the μp

Table 1

Time evolution of the oxygen line X-ray spectra measured at six different temperatures. Uncertainties associated to the measurements include statistical and background-systematic errors. Notice that the first three time bins cover a non-thermalized phase which coincide with the second muon beam spill arrival, low detection efficiency, and high pile-up effects. The fit was performed starting from the bin [1140, 1283] ns.

Time bin [ns]	Oxygen line time evolution [X-rays/ns]					
	300 K	272 K	240 K	201 K	153 K	104 K
800 – 900	435 ± 29	477 ± 28	451 ± 29	407 ± 28	365 ± 30	279 ± 28
900 – 1013	326 ± 15	333 ± 14	296 ± 15	266 ± 14	220 ± 15	202 ± 14
1013 – 1140	257 ± 11	252 ± 10	243 ± 11	224 ± 10	182 ± 11	146 ± 10
1140 – 1283	228 ± 8	216 ± 8	215 ± 8	192 ± 8	156 ± 8	111 ± 8
1283 – 1444	166 ± 7	165 ± 7	161 ± 7	133 ± 7	123 ± 7	83 ± 7
1444 – 1626	136 ± 5	124 ± 5	133 ± 5	119 ± 5	109 ± 6	88 ± 5
1626 – 1829	104 ± 5	104 ± 4	103 ± 5	96 ± 4	87 ± 5	68 ± 4
1829 – 2059	84 ± 4	85 ± 4	90 ± 4	83 ± 4	76 ± 4	63 ± 4
2059 – 2317	53 ± 3	60 ± 3	58 ± 3	57 ± 3	55 ± 3	47 ± 3
2317 – 2608	34 ± 3	36 ± 3	36 ± 3	40 ± 3	37 ± 3	34 ± 3
2608 – 2935	28 ± 2	26 ± 2	26 ± 2	30 ± 2	31 ± 2	30 ± 2
2935 – 3303	14.8±1.9	15.1±1.9	14 ± 2	17.1±1.9	20 ± 2	21.9±1.9
3303 – 3717	6.1±1.6	7.1±1.6	9.9 ±1.6	10.2±1.6	11.9±1.7	13.3±1.6
3717 – 4183	–	–	4.2 ±1.3	4.6 ±1.3	6.2 ±1.4	6.9 ±1.4
4183 – 4708	–	–	–	4.2 ±1.0	4.5 ±1.1	6.1 ±1.1
4708 – 5298	–	–	–	–	–	4.4 ±0.8
5298 – 5963	–	–	–	–	–	2.2 ±0.7

Table 2

Summary of transfer rates from muonic hydrogen to oxygen. The first error represents the statistical and background related systematic errors quadratically summed. The second error reports general systematic uncertainty. The third column reports the reduced χ^2 of the fit. The fourth and fifth columns show the results of the Kolmogorov-Smirnov (K-S) test and its derived maximum distance, respectively, performed on the pulls in comparison with a standard Gaussian distribution.

Mean temperature T [K]	$\Lambda_{\text{pO}}(T)$ [10^{10}s^{-1}]	Reduced χ^2	K-S test on pull	K-S max distance
104	3.07 ± 0.29 ± 0.07	1.21	1.00	0.14
153	5.20 ± 0.33 ± 0.10	0.81	0.85	0.25
201	6.48 ± 0.32 ± 0.13	1.95	1.00	0.17
240	8.03 ± 0.35 ± 0.16	1.64	0.81	0.27
272	8.18 ± 0.37 ± 0.17	1.69	0.76	0.30
300	8.79 ± 0.39 ± 0.18	1.80	0.76	0.30

are still energetic and in an epithermal state. The same break in the spectra was observed at PSI and was helpful in understanding the existence of a kinetic energy dependence of the transfer rate from muonic hydrogen to oxygen [5]. The error bars associated to the points include the statistical and background-systematic errors, as described previously.

4. Results

The experimental results are presented in Table 2. During the measurements the temperature was kept stable, with a variation around the mean value T less than 0.1% (column 1), to which the measured rate Λ_{pO} (column 2) is referred. For each fit, the reduced χ^2 is reported (third column). The quality of the fit was assessed also by studying the pull distribution, where the pull is defined as the ratio between the residual and the error associated to the point ($(X_{\text{measured}} - X_{\text{fitted}})/\sigma_X$). If the fit is good and the fluctuation purely statistical, then the pulls are distributed as a standard normal distribution (a Gaussian with zero mean and unit width). For each fit, the unbinned and ordered pulls data set was compared to a standard normal distribution using the Kolmogorov-Smirnov test. Results of probability and maximum distance are reported in columns four and five of Table 2. The results of the Kolmogorov-Smirnov test applied to the six measurements could not exclude normally distributed pulls.

The six data sets and corresponding fits are reported also in Fig. 3, left panel, where each one is artificially adjusted vertically for clearer visualization. It can be seen that the epithermal component becomes more evident at low temperatures when the thermal

transfer rate is smaller. The right panel of Fig. 3 shows the overall distribution of the pulls for the six temperature fits. The solid line represents a Gaussian fit to the distribution and it is in excellent agreement with a standard normal distribution.

Fig. 4 shows the results obtained in this work. This is the first measurement of the transfer rate from muonic hydrogen to oxygen as function of the temperature. The error bars represent the quadratic sum of statistical and systematic errors, as reported in Table 2. The analysis of systematic errors shows that the main source of uncertainty derives from the gas composition. The H_2/O_2 gas mixture was prepared by the supplier with a relative uncertainty of 3%. Other sources of systematic effects, including temperature and pressure measurements, timing, parameters error propagation were estimated to be smaller than 1% each and considered negligible. Results are in excellent agreement with the PSI measurement at 294 K [5]. The lines represent the theoretical results presented in [8,9] as function of kinetic energy, which we converted to temperature using Eq. (1). Experimental data are not in agreement with theoretical calculations, however. This is not surprising, since these rough calculations do not take into account in details the electron screening effects and also other aspects are simplified, e.g., oxygen is treated as a free atom and not as part of oxygen molecule.

5. Conclusions

In the first investigation of the temperature dependence of the muon-transfer process from the thermalized μp atoms to oxygen, we have observed a strong monotonic rise by a factor of ~ 3 of the rate $\Lambda_{\text{pO}}(T)$ in the temperature interval 104–300 K.

The measurements were performed in conditions of thermal equilibrium that allows us to use Eq. (1) and anticipate a much greater rise of the muon transfer rate $\lambda_{\text{pO}}(E)$ from energies $E \sim 0.01$ eV up to energies of the order of $E \sim 0.1$ eV. The work on extracting the explicit energy dependence and the uncertainty of $\lambda_{\text{pO}}(E)$ from the experimental data in Table 2 is currently in progress; the results will be presented elsewhere.

Such a strong change enables us to employ the muon transfer rate to oxygen as a signature of the kinetic-energy gain of the μp atom in the planned FAMU spectroscopy measurement of the hyperfine splitting of 1S state of this atom.

These results allow to test the available theoretical methods for the calculation of charge transfer processes in non-elastic atom

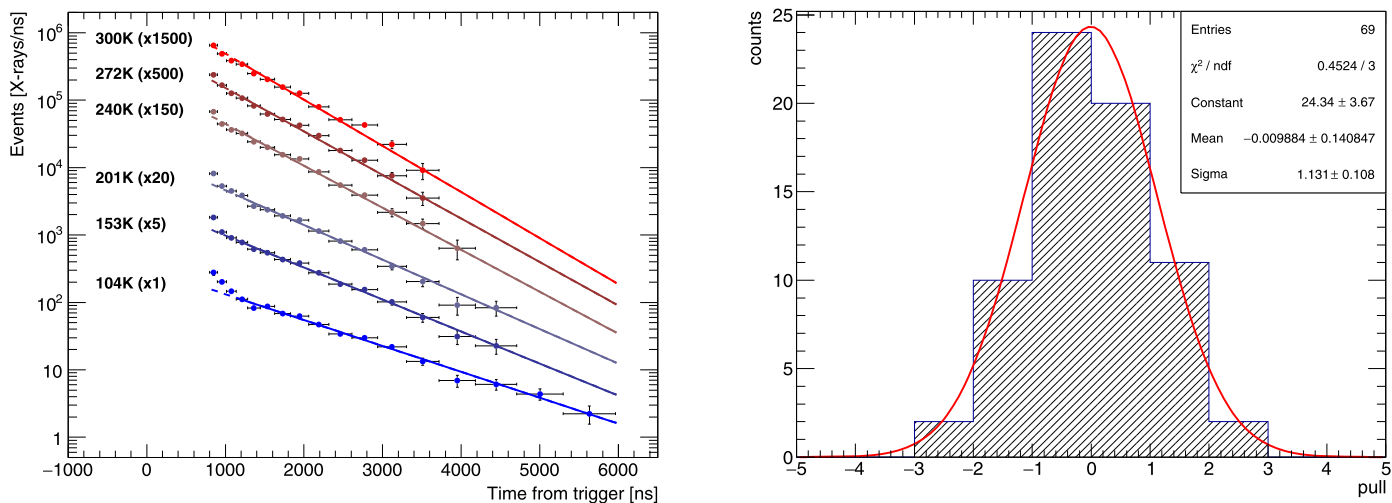


Fig. 3. Left panel: time dependence of the oxygen line intensity for the six temperature bins. Each set of data has been scaled by the factor reported in the figure. Error bars, solid and dashed lines have the same meaning as in Fig. 2. Right panel: overall pull distribution. Solid line is a Gaussian fit of the data. Mean and width are consistent with a standard normal distribution.

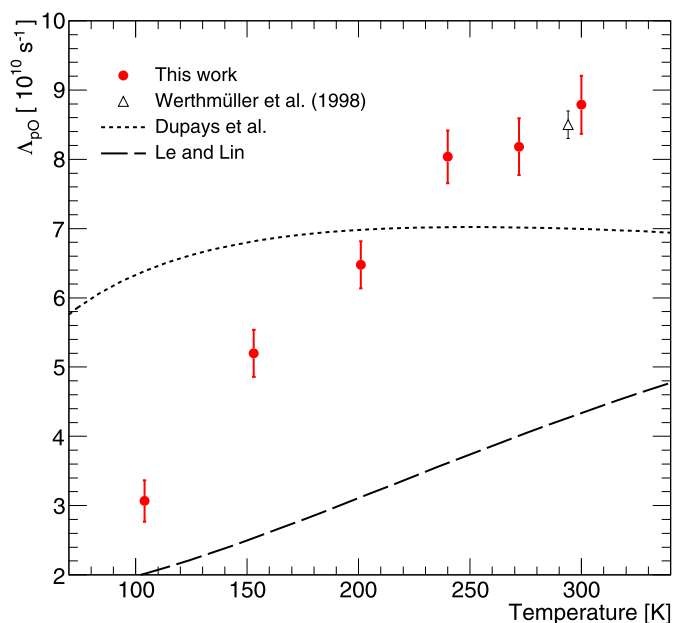


Fig. 4. Transfer rate from muonic hydrogen to oxygen: comparison of the present work with the experimental [5], and the theoretical results [8,9] converted in their temperature dependence using Maxwell–Boltzmann distributions.

scattering and will hopefully stimulate the development of new and more efficient computational approaches.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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