# **Muon transfer from thermalized muonic hydrogen isotopes to argon**

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Recent experimental results on the muon exchange from muonic hydrogen to argon show that the reaction rate is energy dependent near 0.1 eV. A muonic hydrogen atom, formed by muon capture in  $H_2$  gas at 15 bars, is thermalized in a few hundreds of nanoseconds. If the muon transfer reaction occurs before that time, the rate is shown to be slightly lower compared to thermalized muonic atoms  $\left[\lambda_{pAr}=(1.63\pm0.09)\times10^{11} \text{ s}^{-1}\right]$ . As an indirect consequence, the muon transfer rate from  $\mu$ *p* to helium, determined by our group [R. Jacot-Guillarmod and co-workers, Phys. Rev. A 38, 6151 (1988)] is lowered by about 40%. The present value  $\lambda_{pHe}$ =(0.51±0.19)×10<sup>8</sup> s<sup>-1</sup> is in good agreement with other experiments. The transfer rate from muonic deuterium to argon shows also an energy dependence. The muon transfer rate to argon from the deuteron is  $\lambda_{dAr}$ =(0.86±0.04)×10<sup>11</sup> s<sup>-1</sup> at room temperature. The intensity patterns of the muonic Lyman series of argon obtained by muon transfer from both hydrogen isotopes are determined and compared with theoretical predictions. [S1050-2947(97)02705-4]

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## **I. MOTIVATION FOR NEW MEASUREMENTS**

Our first measurements of muon transfer to argon were performed almost ten years ago  $[1,2]$ . We needed a reliable and precise transfer rate to argon in order to deduce the transfer rate to helium from the time distributions of muonic argon x rays measured in triple gas mixtures of  $H_2$ +He+Ar. In mixtures of heavy hydrogen isotopes, the muon catalyzed fusion ( $\mu$ CF) cycle can be drastically shortened due to the muon transfer to helium nuclei. Besides the importance of this rate for the  $\mu$ CF research, the mechanism of muon transfer from hydrogen isotopes to  $Z \ge 2$  elements can be used to test the precision of three-body calculations. Hence this fundamental process is of genuine interest on its own.

The mean values of the normalized (to the atomic density of liquid hydrogen  $N_0$ =4.25×10<sup>22</sup> cm<sup>-3</sup>) transfer rates to argon, known at that time from nine different measurements, contradicted each other and were centered around three values:  $1.4 \times 10^{11} \text{ s}^{-1}$  [3,4],  $3.6 \times 10^{11} \text{ s}^{-1}$  [5,6], and  $9 \times 10^{11}$  s<sup>-1</sup> [7]. The lowest of these values was obtained from time spectra of the muon decay electrons, and one suspected  $[6]$  that the decay electron method yielded, for some yet unknown reason, smaller rates than the x-ray method. Our measurements used the muonic x-ray method. They have shown that in the range from 10 to 140 bars, the transfer rate is independent of the hydrogen pressure and that for relative argon concentrations between 60 ppm and 2% the normalized transfer rate is reproducible. Our four experimental rates, lying between  $1.42 \times 10^{11}$  s<sup>-1</sup> and  $1.48 \times 10^{11}$  s<sup>-1</sup> agreed with each other and with the lowest of the published values  $[1,2]$ . We planned to use the time spectra of the muonic argon x rays measured in  $H_2$ +Ar to check experimental results obtained from the triple mixtures.

There were two main motivations for performing new

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measurements of muon transfer from muonic hydrogen isotopes (protium and deuterium) to argon.

(i) Epithermal effects have been recently discovered in muon transfer to light elements such as carbon, oxygen, fluorine, and neon  $[8-10]$ . Until then, one believed that these were absent in transfer to argon. In this context, where most of the elements studied showed an energy-dependent transfer rate, the case of argon was surprising, because of the ''regu $larity'$  (i.e., single exponential time distribution) of its behavior.

(ii) The transfer rate to helium determined from our triple mixtures  $H_2$ +He+Ar [2] was in contradiction with another value, measured in  $H_2$ +He+Xe at comparable pressures, which was independent of the knowledge of the transfer rate to the third element  $[11]$ . The year after our result on the transfer rate to helium was published, another measurement appeared that yielded a rate more than an order of magnitude less than ours  $[12]$ . The measurement was performed in binary mixtures  $H_2$ +He and the method of analysis was based on the intensity of the 6.85-keV line that is emitted during the deexcitation of the  $(\mu p^4$ He)<sup>+</sup> complex in the  $2p\sigma$  state. It was found later that this rate was in agreement with the value of Ref.  $[11]$  by taking the particle decay channel of the deexcitation into account [13].

The time spectrum of the muonic x rays following the transfer mechanism is generally used to extract the rate of the process. From the intensity pattern of these x rays, especially those of the Lyman series, complementary informations can be obtained with the help of a cascade calculation program. Thus, at first sight, one would expect that the level *n* of an element *Z*, to which the muon is transferred, is the same from both hydrogen isotopes' ground state. More precisely, since the muon is slightly more bound in deuterium, the transfer level *n* should be at the very most lower from deuterium. In oxygen  $[9]$  and neon, however  $[8]$ , the observed

transfer level is by one unit higher from deuterium compared to that from protium. Hence new measurements of time spectra and intensity patterns of the muonic argon x rays in gas mixtures  $H_2$  + Ar and  $D_2$  + Ar should contribute to clarify the situation.

### **II. EXPERIMENT**

The measurements have been performed at the  $\mu$ E4 channel of the Paul Scherrer Institute in Switzerland. Negative muons were stopped in gas mixtures contained in a stainlesssteel vessel of about 9 l  $[2]$ . The mixtures have been prepared at about 150 bars by an industrial factory (Carbagas, Bern), which certified their homogeneity and the relative concentration of the components within  $\pm 2\%$  precision. A mixture of  $H_2$ +0.284% Ar was measured at room temperature at a pressure of 15.1 bars and another of  $D_2$ +0.302% Ar at pressures of 14.6 and 10.0 bars. Following this notation, the concentration corresponds to the ratio of partial argon pressure to the total pressure (for the sake of simplicity, both mixtures will be further labeled with a 0.3% argon concentration. The mixture of  $H_2$ +0.3% Ar contained some deuterium at the natural isotopic concentration level (about 150 ppm). All measurements have been performed during the same datataking period without any change, but the gas composition, in the experimental setup. The muon beam was tuned to 33.0 MeV/*c* for a gas pressure of 15 bars and to 32.0 MeV/*c* at 10 bars.

The performances of our experimental setup can be characterized as follows: energy and timing resolution 1.42 keV and  $6.3$  ns, respectively  $[both full width at half maximum]$ (FWHM) at 643 keV, the energy of the  $\mu$ Ar(2-1) transition. Gating the time spectra with this transition, signal to background ratios of 18.6 and 14.1 were achieved for time intervals from 15 ns to 200 ns and 1  $\mu$ s, respectively (time zero is defined by the prompt peak centroid). The time-to-digital converters had a time resolution of 1 ns per bin. An antipileup system rejected events caused by muons entering the gas target within a  $\pm 5$   $\mu$ s time gate. The fitted time spectra were formed by selecting the events in a 1.85-keV wide window centered on the line of interest, and on its nearby lowand high-energy sides in order to subtract the contribution of the background events from the raw spectrum.

The value of the time resolution was obtained from separate measurements performed in mixtures containing argon gas but no hydrogen. In this case the time spectrum of the argon lines followed a Gaussian distribution around time zero; no delayed events were observed then.

The energy-dependent efficiency of our detection system was determined using off-line calibration sources of  $^{133}$ Ba and 152Eu. Further details concerning the experimental setup, the x-ray detection, the data acquisition system, the method of analysis, and in particular the background subtraction in the time spectra were similar to earlier experiments  $[2,8,9]$ .

## **III. DATA ANALYSIS**

## **A. Muon transfer rate from protium to argon**  $\lambda_{p\text{Ar}}$

The time spectrum of the  $\mu$ Ar(2-1) x-ray transition of the present measurement in  $H_2$ +0.3% Ar at 15 bars is shown in Fig. 1. A clear deviation from the single exponential shape is



FIG. 1. Top: the time spectrum of the muonic  $Ar(2-1)$  x-ray transition measured in  $H_2$ +0.3% Ar at 15 bars. The sharp peak around  $t=0$  is due to direct muon capture in argon and muon transfer from excited  $\mu p^*$  states. The dashed line corresponds to a fit done with a single exponential function after time  $t=200$  ns. Bottom: the corresponding relative deviation expressed in terms of  $(y_{\text{meas}} - y_{\text{fit}})/\sigma(y_{\text{meas}})$ . One observes a strong and negative deviation of the experimental data from the single exponential distribution before  $t=100$  ns. The bin size is 8 ns.

observed at times  $t$ <100 ns. The experimental intensity is systematically smaller by several standard deviations. The higher transitions of the muonic Lyman series show the same behavior. In our earlier measurements  $[2]$ , such deviations were not observed either in the mixture  $H_2 + 0.4\%$  Ar at 10 bars or in  $H_2 + 2\%$  Ar at 15 bars. In the latter mixture, the  $\mu p$ -atom lifetime was only 40 ns [2]. In the present measurement, the statistics is higher and the peak-to-background ratio improved by a factor of about 3–10, depending on the time interval. This improvement has been achieved mainly by reducing the muon beam size at the entrance of the vessel (causing less muon stops in the stainless-steel front flange) and narrowing the energy window set on the transition in order to lower the amount of background counts, without losing dramatically in the intensity of the transition.

By fitting the whole delayed time spectrum of the  $\mu$ Ar(2-1) transition with a single exponential function

$$
\frac{dN_{\mu \text{Ar}}}{dt}(t) \propto e^{-\lambda t},\tag{3.1}
$$

where  $\lambda$  is the disappearance rate of the muonic hydrogen atoms in the mixture, the transfer rate to argon  $\lambda_{pAr}$  (normalized to the atomic density of liquid hydrogen) can be determined by

$$
\lambda_{pAr} = \frac{1}{c_{Ar}} \left\{ \frac{\lambda - \lambda_0}{\varphi} - (c_p \lambda_{pp\mu} + c_d \lambda_{pd}) \right\},\tag{3.2}
$$

where  $\varphi$  is the atomic density of the mixture relative to  $N_0$ ,  $\lambda_0$  is the free muon decay rate  $(0.455 \times 10^6 \text{ s}^{-1})$ ,  $\lambda_{pp\mu} = 2.5 \times 10^6 \text{ s}^{-1}$  is the  $pp\mu$  formation rate,  $\lambda_{pd} = 1.68 \times 10^{10}$  s<sup>-1</sup> is the transfer rate to deuterium, and  $c_p$  and  $c_d$  are the atomic fractions of each hydrogen isotope  $(c_p + c_d + c_{Ar} = 1)$ . By fitting the whole delayed time spectrum  $(t>15 \text{ ns})$  with one single exponential function, one obtains a transfer rate to argon that is compatible with our preceding values, but at the expense of a poor fit  $(\chi^2 \approx 1.5)$ . By fitting only the late part (*t*>200 ns), which has a single exponential shape, the transfer rate to argon increases to  $\lambda_{pAr} = 1.63 \times 10^{11} \text{ s}^{-1}$ . This value is confirmed by fitting in the same manner the time spectra of the higher members of the Lyman series.

This value is interpreted as the transfer rate from thermalized muonic hydrogen atoms in the ground state to argon. At early times, due to collisional deexcitation, muonic hydrogen atoms may well have a nonthermal distribution when they reach the ground state. Indeed, simulations have shown  $[14]$ that at pressures around 10 bars, about half of the  $\mu p_{1s}$  atoms have high energies, i.e.,  $E > 10$  eV, at the time of formation. Thermal energy is only reached after further collisions. We have verified that the time distributions of our present and preceding measurements can be both reproduced by Monte Carlo (MC) simulations using the same set of parameters for the energy dependence of the transfer rate to argon. The code makes use of the energy dependent  $\mu p_{1s}$ +H<sub>2</sub> and  $\mu d_{1s}$ +D<sub>2</sub> scattering cross sections [15,16].

For the energy spectrum of the  $\mu p_{1s}$  atoms at time  $t=0$ , we assumed a sum of two Maxwellian distributions of equal weights, one centered at  $E_1=20$  eV and the other at thermal energy  $E_2$ =0.04 eV. This choice has been triggered by the extensive simulations made for  $H_2+O_2$  mixtures, which are found to be very sensitive to the energy distribution of the  $\mu p_{1s}$  atoms at the time of transfer [17]. For the energy dependence of the transfer rate, we assumed for the sake of simplicity a step function  $\lambda_{pAr}^e$  for  $E>E_0$  and  $\lambda_{pAr}^t$ for  $E \le E_0$ , with  $\lambda_{pAr}^e < \lambda_{pAr}^t$ . The last inequality is suggested by the shape of the experimental time spectra.

In the MC simulations, only the three parameters  $\lambda_{pAr}^e$ ,  $\lambda_{pAr}^t$ , and  $E_0$  related to the energy dependence of the transfer rate were varied. The experimental and simulation histograms were compared using a  $\chi^2$ -minimization method. For the present measurement, i.e., the mixture  $H_2$ +0.3% Ar, the  $\mu$ Ar(2-1) time spectrum (cf. Fig. 1) was compared with simulated spectra in the time interval from 35 ns to 635 ns. With the parameters  $\lambda_{pAr}^e = 1.3 \times 10^{11} \text{ s}^{-1}$ ,  $\lambda_{pAr}^{t}$  = 1.8 × 10<sup>11</sup> s<sup>-1</sup>, and  $E_0$  = 0.06 eV, a  $\chi^2$  of 0.98 was obtained. The precision on  $\lambda_{pAr}^e$  and  $\lambda_{pAr}^t$  depends on the empirical function used for the transfer rate and is estimated to be about  $\pm 0.1 \times 10^{11}$  s<sup>-1</sup>. The comparison of the corresponding time distributions measured in the mixtures  $H_2$ +0.4% Ar and  $H_2$ +2% Ar [2] with the simulations using the same values for the three parameters yielded in both cases  $\chi^2$ = 1.07. A 20% variation of one of the parameters gave unacceptable  $\chi^2$  values, such that the given values can be considered as best values in the present approximation.

One observes that the value of  $\lambda_{pAr}^{t}$  is higher than the transfer rate  $\lambda_{pAr}$  from thermalized  $\mu p_{1s}$  atoms determined by the analytical fit. This is expected because one has to take into account that the energy cut  $E_0$  is so low that, even in a totally thermalized situation, as much as 21% of the  $\mu p_{1s}$ atoms are above this energy threshold (with an argon concentration of 0.3% at a total pressure of 15 bars). Hence one can determine from the two values  $\lambda_{pAr}^e$  and  $\lambda_{pAr}^t$  an approximate weighted mean value for the transfer rate to argon from

thermalized  $\mu p_{1s}$  atoms:  $\langle \lambda_{pAr} \rangle = 1.69 \times 10^{11} \text{ s}^{-1}$ , a value that is compatible with the transfer rate determined by the analytical fit for  $t > 200$  ns.

At kinetic energies  $E \ge 8/Z^2$  (eV), the transfer rate for the *S* wave should be of the form  $\lambda \propto 1/v$ , where *v* is the relative velocity of the collision partners  $[18]$ . In the case of argon, this energy  $(0.025 \text{ eV})$  has to be compared with the energy cut of  $E_0$ =0.06 eV. Our values for the two parameters  $\lambda_{pAr}^e$  and  $\lambda_{pAr}^t$  might point to such a velocity dependence. This result suggests that the transfer rate should increase by lowering the gas temperature.

From the analysis of the present measurement in the mixture  $H_2$ +0.3% Ar at 15 bars, we deduce a mean normalized transfer rate

$$
\lambda_{pAr} = (1.63 \pm 0.09) \times 10^{11} \text{ s}^{-1} \tag{3.3}
$$

from thermalized  $\mu p_{1s}$  atoms at room temperature ( $\approx 300$ K) to argon. Taking into account that the contribution of muon transfer to deuterium on the total disappearance rate of the  $\mu p_{1s}$  atoms is only 1%, the transfer reaction  $\mu$ *d*+Ar $\rightarrow \mu$ Ar+*d* introduces a negligible correction on the value of  $\lambda_{pAr}$ , well within the given uncertainty.

## **B. Consequence for the muon transfer rate from protium to helium**  $\lambda_{p\text{He}}$

The average value for the transfer rate to helium, which we deduced some years ago from measurements in triple gas mixtures H<sub>2</sub>+He+Ar, was  $\lambda_{pHe} = (0.88 \pm 0.09) \times 10^8 \text{ s}^{-1}$ [2]. This value is about twice as large as the transfer rates measured by Bystritsky et al. [11], von Arb et al. [12] (see also Kravtsov *et al.*  $|13|$ , and the theoretical calculations  $[19,20]$ .

To clarify this disagreement, we studied the transfer rate to helium using a triple gas mixture made  $H_2$ +He+CH<sub>4</sub>. The transfer rate to carbon was first determined in a binary mixture of  $H_2$ +0.18%CH<sub>4</sub> at pressures of 10, 15, and 40 bars. The time distributions of the muonic carbon x rays strongly deviated from the expected single exponential structure. The analysis revealed that the duration of the deviation was about proportional to the inverse of the total pressure, i.e., characteristic of a thermalization process. In the triple mixture  $H_2$ +50%He+0.18%CH<sub>4</sub> at 15 bars, the duration of the deviation was much shorter than in the binary mixture at the same pressure  $[10]$ . This indicated that the  $\mu p_{1s}$  atoms slow down much faster in collisions with helium atoms than with hydrogen molecules.

By using the value of the transfer rate from thermalized  $\mu p_{1s}$  atoms to carbon determined in the binary mixture [21], one deduced a transfer rate to helium from the total decay rate measured in the triple mixture. Unfortunately, this decay rate was due by more than 80% to muon transfer to carbon. As a consequence, the uncertainty in the transfer rate to carbon  $\left[\lambda_{pC} = (0.95 \pm 0.05) \times 10^{11} \text{ s}^{-1} \right]$  from the measurement with the binary mixture] was too large and the statistics accumulated insufficient to allow a precise determination of the transfer rate to helium. The upper limit that could be deduced

$$
\lambda_{p\text{He}} \le 0.6 \times 10^{11} \text{ s}^{-1} \text{ (carbon data)}
$$

was in contradiction with the average value deduced from our preceding measurements performed in the triple mixtures



FIG. 2. Top: the time spectra of the muonic  $Ar(2-1)$  x-ray transition measured in  $D_2$ +0.3% Ar at (a) 10 bars and (b) 15 bars. The sharp peak around  $t=0$  is due to direct muon capture in argon and muon transfer from excited  $\mu d^*$  states. The dashed lines correspond to fits with single exponential functions after time  $t = 250$  ns. Middle and bottom: the corresponding relative deviation expressed in terms of  $(y_{\text{meas}} - y_{\text{fit}})/\sigma(y_{\text{meas}})$  at (a) 10 bars and (b) 15 bars, respectively. One observes strong and positive deviations of the experimental data from the single exponential distribution before  $t=200$  ns. The bin size is 8 ns.

 $H_2$ +He+Ar [2]. It agreed, however, with the theoretical predictions and the other experimental values.

Since the deceleration of the epithermal  $\mu p_{1s}$  atoms in collisions with helium atoms is much faster than with hydrogen molecules, the  $\mu p_{1s}$  atoms reach thermal energies in mixtures  $H_2$ +He at 15 bars with a 35–50 % helium concentration in a few tens of nanoseconds  $[10]$ . The time spectra of the muonic argon x rays, measured in the triple mixtures  $H_2$ +He+Ar, extended up to about 1  $\mu$ s and their shapes followed single exponential distributions see Fig.  $4(b)$  of Ref.  $[2]$ . With the present value for the transfer rate to argon [see Eq. (3.3)] from thermalized  $\mu p_{1s}$  atoms, one deduces a different value for the transfer rate to helium, namely,

$$
\lambda_{p\text{He}} = (0.51 \pm 0.19) \times 10^8 \text{ s}^{-1} \text{ (present argon data)}.
$$

This rate agrees with the upper limit deduced from the triple mixture  $H_2$  + He + CH<sub>4</sub>.

#### **C.** Muon transfer rate from deuterium to argon  $\lambda_{\text{dAr}}$

Time spectra of the  $\mu$ Ar(2-1) x-ray transition measured in the mixture  $D_2$  + 0.3% Ar at 10 and 15 bars are shown in Fig. 2. Clear deviations from the shape of a single exponential are observed in the time interval  $t$  < 200 ns. The fits show that the deviation extends further at 10 bars compared to 15 bars, due to the longer time needed by the  $\mu d_{1s}$  atoms to thermalize at lower pressure.

In comparison with the time spectrum measured in  $H_2$ +0.3% Ar, the deviation goes in the opposite direction and extends to about twice longer times. The positive amplitude of the deviation suggests that the transfer rate from epithermal  $\mu d_{1s}$  atoms is higher than from thermalized ones. In the case of  $H_2$ +0.3% Ar, the deviation could be explained by the behavior of the *S*-wave transfer rate  $\lambda_{pAr} \propto 1/v$ , i.e., its decrease with energy. Obviously, the positive deviation in  $D_2$ + Ar cannot be explained by the same argument. It might, however, be due to  $P$ -wave transfer  $[22]$  or higher waves with a different energy dependence. The fact that the deviation extends to longer times than in  $H_2 + Ar$  indicates that either the energy cut  $E_0$  has a lower value or that the thermalization of  $\mu d_{1s}$  atoms in  $D_2$  takes longer than the one of  $\mu p_{1s}$  atoms in H<sub>2</sub>.

Following Eq.  $(3.1)$ , the normalized muon transfer rate from deuterium to argon can be calculated as

$$
\lambda_{dAr} = \frac{1}{c_{Ar}} \left\{ \frac{\lambda - \lambda_0}{\varphi} - c_d \omega_s^{dd} \lambda_{dd\mu} \right\},\tag{3.4}
$$

where  $\lambda_{dd\mu}$  is the formation rate of the  $dd\mu$  molecule and  $\omega_s^{dd}$  is the effective muon sticking coefficient after the fusion reaction.

By fitting only the late part ( $t > 250$  ns) of the  $\mu$ Ar(2-1) time spectra with an exponential distribution, one deduces transfer rates to argon of  $\lambda_{dAr}$ =[0.88±0.04 (statistical)]  $\times 10^{11}$  s<sup>-1</sup> for the mixture D<sub>2</sub>+0.3% Ar at 10 bars and  $\lambda_{dAr}$ =[0.84±0.04 (statistical)] $\times 10^{11}$  s<sup>-1</sup> at 15 bars. The weighted mean value of

$$
\lambda_{dAr} = (0.86 \pm 0.04) \times 10^{11} \text{ s}^{-1}
$$

is then the transfer rate from thermalized  $\mu d_{1s}$  atoms to argon at room temperature (the specified uncertainty includes both statistical and systematic ones). This value is slightly smaller than the rate  $\lambda_{dAr} = (0.94 \pm 0.08) \times 10^{11} \text{ s}^{-1}$  deduced from the time distribution of the muon decay electrons in a mixture of  $D_2$ +Ar at 6 bars [23].

#### **D. Muon cascade in argon following a transfer reaction**

By using the same approximations as those made by Holzwarth and Pfeiffer  $[24]$  for the case of muon transfer to fluorine, the atomic level to which both  $\mu p_{1s}$  and  $\mu d_{1s}$  atoms transfer their muon in collisions with argon should be  $n=12$ , the transfer probability to levels  $n \neq 12$  being practically negligible. The population of each angular momentum state *l* can be approximated by the formula

$$
P_l = (2l+1)\frac{[(n-1)!]^2}{(n+l)!(n-l-1)!},
$$
\n(3.5)

which favors low angular momenta  $[24]$ . From this angular momentum distribution, one can determine the intensity pattern of the muonic Lyman series of argon following muon transfer by a cascade calculation.

For the mixture  $H_2$ +0.3% Ar, the intensities of the delayed muonic x rays of the Lyman series resulting from muon transfer have been determined separately for thermalized  $\mu p_{1s}$  atoms (time window 340 ns  $\lt t \lt 1140$  ns; cf. Fig. 1) and for epithermal ones (time window 40 ns  $\lt t \lt 340$  ns).



FIG. 3. Energy spectra of the delayed muonic x rays  $(5-1)$  to 11-1) of the argon Lyman series measured in the mixtures  $H_2$ +0.3% Ar and D<sub>2</sub>+0.3% Ar, both at 15 bars. The yields have been normalized to  $10<sup>6</sup>$  incident muons, in order to help the comparison between the spectra (the zero line for  $H_2$ +Ar has been raised by 1.5 to ease the comparison). The characteristic energy of each transition is indicated on the horizontal axes.

No significative differences have been observed in the relative intensities  $I(n-1)/I(2-1)$ . In both cases, the highest transition is the  $(11-1)$  with an intensity comparable to the  $(10-1)$ . The x-ray spectrum in this energy range is shown in Fig. 3. The background line at  $846.8 \text{ keV}$  (due to nuclear reaction with  $56Fe$ ), which coincides in energy with the transition  $\mu$ Ar(6-1), has been strongly suppressed compared to earlier measurements (see, e.g., Fig. 5 of Ref.  $[2]$ ) from more than 50% down to 20% of the peak intensity. The precise contribution of this background line has been determined from measurements performed in mixtures of hydrogen containing no argon. In Ref. [2], the intensity of the  $\mu$ Ar(6-1) transition could only be estimated using a cascade calculation.

The intensity of the muonic argon transitions have been evaluated by correlating their position and FWHM in order to reduce as much as possible the effects due to statistical fluctuations on the precision of the fit. By normalizing the sum of the measured muonic x-ray intensities of the Lyman series to unity, one can compare them with cascade calculations. Such calculations were performed by assuming that the refilling rate of the electronic  $K$  shell is as fast as in a neutral atom  $[25]$ . Table I shows that the agreement between measured and calculated intensities is very satisfactory if one assumes that the muon transfer occurs to a level  $n=11$ . If muon transfer proceeded to the level  $n=12$ , one should have observed a  $(12-1)$  transition with a relative intensity of 0.0270, which is well within the experimental level of sensitivity.

For the mixture  $D_2$  + 0.3% Ar at 10 and 15 bars, the intensities of the delayed muonic x rays of the Lyman series have been determined in the same manner. No pressure dependence has been observed. The relative x-ray intensities are the same for thermal and epithermal  $\mu d_{1s}$  atoms. For the highest *K*-series transitions  $[(9-1), (10-1),$  and  $(11-1)]$ , they are different from those in  $H_2$  + 0.3% Ar (see Table I and Fig. 3). In particular, the  $(11-1)$  transition is almost completely missing. The relative intensities deduced from cascade calculations done with  $n=11$  or 12 disagree with those measured in the mixture  $D_2$  + 0.3% Ar, but these are fairly reproduced by assuming that the muon from the deuteron is transferred to the argon level  $n=10$ . Calculations performed

TABLE I. Intensity patterns of the muonic Lyman series of argon after muon transfer from protium and deuterium  $(15 \text{ bars}).$ The calculated intensities (last two columns) were obtained by a cascade program  $[25]$ , using formula  $(3.5)$  for the initial angular momentum distribution.

|            | Experiment       |                  | Calculation |        |
|------------|------------------|------------------|-------------|--------|
| Transition | $H_2 + 0.3\%$ Ar | $D_2 + 0.3\%$ Ar | $n=10$      | $n=11$ |
| $Ar(2-1)$  | 0.418(11)        | 0.407(11)        | 0.4183      | 0.4172 |
| $Ar(3-1)$  | 0.0956(27)       | 0.0934(25)       | 0.0930      | 0.0925 |
| $Ar(4-1)$  | 0.0618(19)       | 0.0610(18)       | 0.0646      | 0.0642 |
| $Ar(5-1)$  | 0.0899(26)       | 0.0934(26)       | 0.0962      | 0.0953 |
| $Ar(6-1)$  | 0.112(14)        | 0.117(14)        | 0.1176      | 0.1167 |
| $Ar(7-1)$  | 0.0837(24)       | 0.0918(25)       | 0.0934      | 0.0930 |
| $Ar(8-1)$  | 0.0484(20)       | 0.0564(20)       | 0.0510      | 0.0525 |
| $Ar(9-1)$  | 0.0333(11)       | 0.0194(8)        | 0.0097      | 0.0188 |
| $Ar(10-1)$ | 0.0304(11)       | 0.0578(16)       | 0.0561      | 0.0205 |
| $Ar(11-1)$ | 0.0265(11)       | 0.0024(5)        |             | 0.0292 |

by lowering or increasing the refilling rate of the electronic *K* shell do not improve the overall agreement.

## **IV. CONCLUSION**

The time spectra of the muonic argon x rays, measured in gas mixtures of H<sub>2</sub>+0.3% Ar and D<sub>2</sub>+0.3% Ar, show deviations from a single exponential distribution. These deviations can be explained as due to an energy dependence of the muon transfer rates from muonic protium and deuterium, respectively, to argon. Our previous measurements of muon transfer from protium to argon  $[2]$  are in agreement with such a dependence. By approximating the energy dependence of the transfer rate with a step function, MC simulations show a decrease of the rate by about 40% for energies above 0.06 eV. At room temperature, about 20% of the  $(\mu p)_{1s}$  atoms in thermal equilibrium have such energies. From the present analysis, one gets a value for the transfer rate from protium to argon that is a mean over the thermal energy distribution at room temperature:  $\lambda_{pAr}$  $=$  (1.63  $\pm$  0.09)  $\times$  10<sup>11</sup> s<sup>-1</sup>. The discrepancy with the results of other authors remains.

As a consequence of the updated value of  $\lambda_{pAr}$ , the transfer rate from protium to helium, measured in triple gas mixtures of  $H_2 + He + Ar$  [2], is lowered. One obtains  $\lambda_{p\text{He}} = (0.51 \pm 0.19) \times 10^8 \text{ s}^{-1}$ . This value is in good agreement with other experimental results and theoretical calculations. The relatively high uncertainty on this result is mainly due to the uncertainty in  $\lambda_{pAr}$ .

A comparison between the time distributions from Figs. 1 and 2 indicates different energy dependences for the transfer rate from the two hydrogen isotopes. If the first is basically in agreement with the explanation from Gershtein, who only studied the *S*-wave transfer, it appears that for the muon transfer from deuterium one may have to take into account higher angular momenta to explain the increase of the transfer rate at epithermal energies compared to thermal ones. A detailed study of the energy dependence of the muon transfer from muonic deuterium would require complementary measurements performed at different concentrations and pressures. This is, however, beyond the scope of this study.

The ratio of the transfer rates from both thermalized muonic hydrogen isotopes is  $\lambda_{pAr}/\lambda_{dAr} = 1.91 \pm 0.13$ , which is compatible with the reduced masses ratio of the colliding partners. Therefore, this result tends to support a dependence of the transfer rate going with the inverse of the mass, as also measured by other experiments  $[4]$ . This observation is not valid in the cases of oxygen  $[17]$ , neon  $[8]$ , and sulfur  $[9]$ .

The intensity patterns of the argon Lyman series due to transfer from both hydrogen isotopes are found to be identical at short (strong deviation from the exponential distribution) and long (from 200–250 ns to about 1  $\mu$ s) times. This indicates that the muonic argon atom is always formed with the same characteristic distribution over  $(n,l)$  states, independently of the collision energy (and the corresponding transfer rate).

The intensity patterns of the Lyman series resulting from muon transfer (cf. Table I) show good agreement between both hydrogen isotopes up to the  $(8-1)$  transition. The higher *K*-series transitions are not reproduced by cascade calculations assuming an initial distribution over angular momentum states given by formula  $(3.5)$ . The intensity pattern of the transitions  $\mu Ar(9-1)$  to  $\mu Ar(11-1)$  in D<sub>2</sub>+Ar compared to the pattern in  $H_2$ +Ar might indicate that the muon transfer from  $\mu d_{1s}$  atoms proceeds predominantly to a lower level  $(n=10)$  than from  $\mu p_{1s}$  atoms  $(n=11)$ .

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