Muon transfer from excited states of hydrogen and deuterium to nitrogen, neon, and argon

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Muon transfer from excited μp^* and μd^* atoms to the three gases nitrogen, neon, and argon is studied. Variations of the experimental conditions such as pressure and relative concentrations on the fractions α_{pZ} and α_{dZ} of excited state events in prompt muonic x rays are analyzed. The differences between excited-state transfer from μp^* and μd^* to N_2, Ne, and Ar are discussed. For these elements, capture ratios $A(H_2, Z)$ and $A(D_2, Z)$ are given, from which an indirect value $A(H_2, D_2)$ is deduced. [S1050-2947(98)01403-6]

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I. INTRODUCTION

In a previous work [1], we developed a method for studying the charge transfer of negative muons from excited muonic hydrogen atoms to elements with $Z > 2$. As a first illustration, two measurements performed in $H_2 + Ar$ mixtures were analyzed. About one-third of the prompt muonic x-ray intensity of the argon Lyman series was found to be the consequence of direct capture by argon atoms, and two-thirds to muon transfer from excited muonic hydrogen to argon. However, our analysis was restricted to the lower transitions in the Lyman series ($n < 6$), and experiments were performed at fixed pressure and at only two different relative concentrations.

In the present paper, we report on results about excited-state transfer obtained in gaseous mixtures of hydrogen ($H_2 + Z$) and of deuterium ($D_2 + Z$) containing small amounts of nitrogen, neon, or argon. We broach variations in pressure and consider a number of relative concentrations. Taking into account excited-state transfer, we determine the Coulomb capture ratios of negative muons $A(H_2, Z)$ and $A(D_2, Z)$ [1] in all investigated gas mixtures. Finally, we discuss the implications of the existence of a long-lived fraction of metastable muonic hydrogen in the $2s$ state [2] on our results.

II. METHOD

A negative muon stopped in a gas mixture $H_2 + Z$ is trapped either by a Z atom or by hydrogen. With a hydrogen nucleus, it forms a muonic hydrogen atom μp^* in an excited state. In collisions, the muon can be transferred to a Z atom before having reached the ground state [1]. Excited-state transfer, as well as ground-state transfer, populates excited states of the μZ^* atom. Hence the muonic x rays from the cascade of the muon toward the ground state of the μZ atom can originate from three different processes: from direct capture and excited-state transfer, for which the x rays are prompt with regard to the entrance of a muon into the target, and from ground-state transfer where the x rays are delayed due to the relatively long lifetime of the μp_{1s} atom [1]. From the measurement of the x-ray time distribution of any transition, one can determine the yields of the prompt and the delayed parts (see Fig. 1).

The direct capture and excited-state transfer processes are both faster than our time resolution. To determine the fraction of the prompt x rays which is due to excited-state transfer to an element Z, we make use of characteristic patterns of the muonic x-ray intensities in μZ [1]. Indeed, the relative intensities (normalized to the sum of the the whole Lyman series) are characteristic signatures of direct capture by the element Z [3–5] or of ground-state transfer to Z [6–8]. Since the prompt x rays are composed of events resulting from direct capture and from excited-state transfer, the prompt intensity pattern P_n is a superposition of the direct capture pattern C_n and excited-state transfer pattern T_n^*. P_n, C_n, and T_n^* are the normalized relative x-ray intensities of the n to 1 transition of the prompt events, of those of direct capture (which are known from measurements in a pure Z gas) and those of excited-state transfer, respectively. The last ones cannot be determined directly from a measurement.

By assuming that the ground-state transfer intensity pattern T_n is a good approximation for the pattern resulting from excited-state transfer [1], i.e., $T_n^* \cong T_n$, the prompt intensity pattern P_n can be expressed as a linear combination of the patterns of ground-state transfer and direct capture. For every transition n to 1, we may write
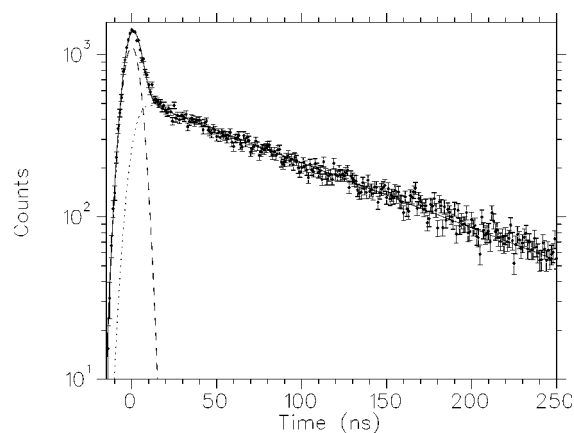

FIG. 1. Background-subtracted time spectrum of the $\mu N(2-1)$ transition measured in $D_2 + 1.07\%$ N_2 at 5 bar and room temperature. The solid line represents a fit made with the sum of a prompt Gaussian peak (dashed line) and a delayed exponential part (dotted line).

$$P_n = \alpha_n T_n + (1 - \alpha_n) C_n. \qquad (2.1)$$

For every transition of the Lyman series in μZ, we obtain the fraction α_n of prompt events which are due to transfer from excited states of the μp^* atoms. When hydrogen is replaced by deuterium in the mixtures, the same method can be applied without restriction, by using the pattern T_n following transfer from the μd_{1s} atoms [7,8]. For the element Z, the fractions α_{pZ} and α_{dZ} are then defined as the average of the respective α_n values over all transitions of the Lyman series in μZ. Hence α_{pZ} and α_{dZ} represent the fraction of prompt muonic x rays due to excited-state transfer from μp^* and μd^* atoms to Z.

III. COULOMB CAPTURE RATIO

Because a significant part of the prompt x rays proceeds from transfer from excited states of μp^* atoms, the formula proposed by Schneuwly [9] to evaluate the per-atom (-molecule) capture ratio in $H_2 + Z$ mixtures has to be modified [1]. Indeed, this formula considers the whole prompt intensity as due to direct capture by Z atoms. With the knowledge of the fraction α_{pZ}, we can subtract the part of the prompt intensity due to excited-state transfer. In order to have the number of muons captured by hydrogen, this part has to be added to the delayed intensity. One may write the per-atom (-molecule) Coulomb capture ratio [1]

$$A(H_2, Z) = \frac{c_Z}{(1 - c_Z)} \frac{N_D \dfrac{\lambda}{\phi c_Z \lambda_{pZ}} + \alpha_{pZ} N_P}{(1 - \alpha_{pZ}) N_P}, \qquad (3.1)$$

where N_P and N_D are the sum over the Lyman series of the prompt and delayed x-ray intensities of μZ (note that N_D was called N_T in Ref. [1]). The rate λ is the total disappearance rate of the μp_{1s} atoms in the mixture; λ_{pZ} is the transfer rate to Z from the ground state of the μp atoms, normalized to the atomic density of liquid hydrogen [7,8], ϕ the atomic gas density relative to the atomic density of liquid hydrogen, and, finally, c_Z the gas concentration relative to hydrogen, quoted as the ratio of partial pressure of the mixture.

TABLE I. Intensity patterns C_n following direct capture in neon and in nitrogen, measured in different Ne + Ar, N_2 + Ar, and N_2 + Ne mixtures between 5 and 15 bar. Results of cascade calculations (see text) are indicated for both elements. The intensity of the contaminated $\mu Ne(5-1)$ and $\mu Ne(6-1)$ transitions have been evaluated using calculations.

Transition	C_n in Ne	Calculations	C_n in N	Calculations
(2-1)	0.9209(93)	0.9211	0.840(33)	0.8416
(3-1)	0.0341(4)	0.0346	0.069(3)	0.0634
(4-1)	0.0103(2)	0.0096	0.030(1)	0.0302
(5-1)	0.0051(3)	0.0049	0.020(1)	0.0212
(6-1)	0.0043(3)	0.0042		
(n-1)	0.0253(6)	0.0256	0.041(2)	0.0436
ΣLyman	1.000	1.000	1.000	1.000

The product $N_D \lambda / \phi c_Z \lambda_{pZ}$ corresponds to the number of μp atoms which reach the ground state. With α_{pZ} being the part of the prompt events due to transfer from excited states, $\alpha_{pZ} N_P$ represents then the number of μp^* atoms which transfer their muon from an excited state to Z. The sum of these two terms consequently corresponds to the number of muons captured by hydrogen. $(1 - \alpha_{pZ}) N_P$ is the number of μZ atoms formed by direct capture. With the factor $c_Z / (1 - c_Z)$, which gives the ratio of the number of Z atoms to the number of hydrogen molecules per unit of volume in the $H_2 + Z$ gas mixture, one obtains the per-atom (-molecule) capture ratio.

Equation (3.1) can be applied without restriction to the corresponding data from deuterium mixtures to determine the capture ratios $A(D_2, Z)$.

IV. MEASUREMENTS AND DATA ANALYSIS

All measurements were performed at the Paul Scherrer Institute (Switzerland) in a similar way as for the two argon measurements already mentioned [1]. A total of three series of measurements were performed at room temperature, namely, binary mixtures of nitrogen, neon, and argon with hydrogen as well as with deuterium. Relative concentrations and total pressures of the mixtures were varied.

TABLE II. Prompt relative intensities P_n and α_n values for the transitions $\mu Ar(n \rightarrow 1)$ measured in hydrogen and in deuterium mixtures at 15 bar and room temperature. The intensities of the $\mu Ar(6-1)$ transition are based on estimations and are only indicative. $\mu Ar(n$-$1)$ represents the sum of the $\mu Ar(9-1)$ to the $\mu Ar(11-1)$ transitions.

Transition Ar	$H_2 + 1.98\%$ Ar P_n	α_n	$H_2 + 0.284\%$ Ar P_n	α_n	$D_2 + 0.302\%$ Ar P_n	α_n
(2-1)	0.591(21)	0.62(5)	0.570(26)	0.66(5)	0.721(25)	0.37(5)
(3-1)	0.090(4)	0.81(7)	0.084(12)	0.71(19)	0.083(4)	0.70(6)
(4-1)	0.043(3)	0.62(5)	0.049(9)	0.73(17)	0.030(4)	0.38(7)
(5-1)	0.063(4)	0.63(4)	0.063(7)	0.63(8)	0.035(4)	0.30(5)
(6-1)	0.076		0.076		0.042	
(7-1)	0.056(3)	0.60(4)	0.053(5)	0.57(6)	0.037(5)	0.37(6)
(8-1)	0.027(2)	0.51(7)	0.039(6)	0.82(17)	0.027(4)	0.50(11)
(n-1)	0.054(4)	0.45(5)	0.066(10)	0.60(14)	0.025(8)	0.24(7)
ΣLyman	1.000		1.000		1.000	

TABLE III. $\alpha_{p\mathrm{Ar}}$ and $\alpha_{d\mathrm{Ar}}$ fractions as well as Coulomb capture ratios $A(\mathrm{H}_2,\mathrm{Ar})$ and $A(\mathrm{D}_2,\mathrm{Ar})$ determined in hydrogen and deuterium mixtures at given pressure and room temperature.

Ar concentration (%)	Pressure (bar)	$\alpha_{p\mathrm{Ar}}$	$A(\mathrm{H}_2,\mathrm{Ar})$	$\alpha_{d\mathrm{Ar}}$	$A(\mathrm{D}_2,\mathrm{Ar})$
0.284	15	0.64(3)	0.19(2)		
1.98	15	0.60(3)	0.18(2)		
0.302	15			0.40(5)	0.17(2)
0.302	10			0.30(4)	0.16(2)
Mean		0.62(2)	0.19(1)	0.35(4)	0.17(1)

The target vessel and the electronic setup are described in Ref. [6]; supplementary details about the pile-up rejection system can be found in Ref. [10]. The gas mixtures were all prepared by the manufactoring plant Carbagas (Switzerland), which guaranteed the relative concentrations c_Z, quoted as ratios of partial pressures in the mixtures, to be accurate to $\pm 1\%$. The purity was better than 50 ppm for each Z component, about 5 ppm for natural hydrogen (with 150 ppm deuterium), and around 0.3% for deuterium. Three germanium detectors were employed. Their time resolutions were typically about 12 ns for the $\mu\mathrm{N}(2-1)$ transition at 101 keV, 9 ns for $\mu\mathrm{Ne}(2-1)$ at 207 keV, and 5 ns for $\mu\mathrm{Ar}(2-1)$ at 643 keV.

Time and energy spectra were constructed off line. The prompt intensities P_n of the $\mu\mathrm{N}$, $\mu\mathrm{Ne}$, and $\mu\mathrm{Ar}$ Lyman transitions were determined from background-subtracted time spectra [1]. The delayed intensities of $\mu\mathrm{N}$ were determined from delayed energy spectra, whereas the delayed intensity patterns of both $\mu\mathrm{Ne}$ and $\mu\mathrm{Ar}$ were taken from previous measurements [6–8]. Some muonic lines of interest, such as $\mu\mathrm{Ne}(5-1)$, $\mu\mathrm{Ne}(6-1)$, or $\mu\mathrm{Ar}(6-1)$, were contaminated by background events. Particular attention had to be paid to this contamination, as the relative intensity patterns P_n, T_n, and C_n are normalized to the sum of the respective intensities of all the Lyman transitions in the element studied.

To determine the intensity patterns of the Lyman transitions in direct capture in nitrogen, neon, and argon, we performed measurements in several $\mathrm{N}_2+\mathrm{Ar}$, $\mathrm{N}_2+\mathrm{Ne}$, and $\mathrm{Ne}+\mathrm{Ar}$ mixtures at pressures between 5 and 15 bar. The results for argon were already presented in Ref. [1]. The mean intensity patterns obtained for nitrogen and neon could be reproduced by cascade calculations using code written by Akylas and Vogel [11], where the K-shell refilling width can be adjusted. Table I shows the Lyman intensities measured in $\mu\mathrm{Ne}$ and $\mu\mathrm{N}$, and the corresponding values calculated with an initial statistical distribution of angular momenta at a level $n=14$ and a K-shell refilling width equal to 0.15% and 1.4% of the width of the corresponding neutral atom. Thanks to the good agreement between the first three $\mu\mathrm{Ne}$ Lyman transitions, the calculations have been used to estimate the intensities of the $\mu\mathrm{Ne}(5-1)$ and $\mu\mathrm{Ne}(6-1)$ transitions contaminated by muonic x rays from the iron of the target walls.

V. RESULTS

A. Argon

Two measurements performed in hydrogen with different argon concentrations at a pressure of 15 bar were analyzed, as well as one mixture of argon and deuterium at pressures of 10 and 15 bar. The prompt intensities of the Lyman transitions were determined individually up to the $\mu\mathrm{Ar}(8-1)$ transition, whereas the intensities of the higher transitions $\mu\mathrm{Ar}(9-1)$ to $\mu\mathrm{Ar}(11-1)$ were summed up. The intensity of the $\mu\mathrm{Ar}(6-1)$ peak contained background events due to γ rays from excited $^{56}\mathrm{Fe}$ nuclei in the walls of the target vessel. This nuclear reaction yielded delayed as well as prompt γ rays with regard to the entering muons. Hence one could not subtract their intensity to correct the prompt intensity of the $\mu\mathrm{Ar}(6-1)$ transition. To estimate this particular muonic intensity, we use an evaluation of the fractions $\alpha_{p\mathrm{Ar}}$ [1] and $\alpha_{d\mathrm{Ar}}$ based on the lower Lyman transitions ($n<6$) of argon. The knowledge of the two intensity patterns T_n [7] and C_n [1], together with $\alpha_{p\mathrm{Ar}}$ and $\alpha_{d\mathrm{Ar}}$, gives an estimate of the prompt intensity of the $\mu\mathrm{Ar}(6-1)$ transition by using Eq. (2.1).

Table II shows the prompt relative intensities P_n, normalized to the sum of the Lyman series, and the α_n values of the

TABLE IV. Prompt relative intensities P_n and α_n values for the transitions $\mu\mathrm{Ne}(n\rightarrow 1)$ measured in hydrogen and in deuterium mixtures at different pressures and room temperature.

Transition	$\mathrm{H}_2+1.51\%\,\mathrm{Ne}$				$\mathrm{D}_2+0.400\%\,\mathrm{Ne}$	
	15 bar		10 bar		7 bar	
Ne	P_n	α_n	P_n	α_n	P_n	α_n
(2-1)	0.700(17)	0.41(4)	0.690(19)	0.43(4)	0.823(25)	0.17(5)
(3-1)	0.102(5)	0.59(5)	0.102(6)	0.59(5)	0.059(7)	0.23(7)
(4-1)	0.081(5)	0.36(3)	0.091(6)	0.41(3)	0.050(6)	0.20(3)
(5-1)	0.084(8)	0.48(6)	0.084(8)	0.48(5)	0.042(9)	0.20(5)
(6-1)	0.033(7)	0.34(8)	0.033(7)	0.34(8)	0.022(8)	0.19(9)
ΣLyman	1.000		1.000		1.000	

TABLE V. $\alpha_{p\mathrm{Ne}}$ and $\alpha_{d\mathrm{Ne}}$ fractions as well as Coulomb capture ratios $A(\mathrm{H}_2,\mathrm{Ne})$ and $A(\mathrm{D}_2,\mathrm{Ne})$ determined in hydrogen and deuterium mixtures at given pressure and room temperature.

Ne concentration (%)	Pressure (bar)	$\alpha_{p\mathrm{Ne}}$	$A(\mathrm{H}_2,\mathrm{Ne})$	$\alpha_{d\mathrm{Ne}}$	$A(\mathrm{D}_2,\mathrm{Ne})$
0.687	15	0.47(4)	0.28(2)		
1.43	15	0.45(4)	0.25(2)		
1.51	15	0.42(4)	0.28(3)		
1.51	10	0.45(3)	0.27(3)		
1.98	15	0.42(3)	0.21(2)		
2.98	15	0.41(3)	0.25(2)		
0.400	7			0.19(2)	0.25(3)
0.434	15			0.14(4)	0.22(6)
Mean		0.43(1)	0.25(1)	0.18(2)	0.24(3)

Lyman transitions for the three mixtures measured at 15 bar. The P_n values in the hydrogen mixtures were slightly changed between the previous paper [1] and this final analysis, because of a more precise evaluation of the intensity of the $\mu\mathrm{Ar}(6\text{-}1)$ transition. The pattern of the relative intensities of direct capture in argon, C_n, used to determine the α_n values, is taken from the measurements of Ref. [1]. Earlier measurements [7] provided the ground-state transfer patterns T_n for the transfer from the μp and μd atoms to argon. Table III shows the $\alpha_{p\mathrm{Ar}}$ and $\alpha_{d\mathrm{Ar}}$ fractions for the four mixtures. For each measurement, $\alpha_{p\mathrm{Ar}}$ and $\alpha_{d\mathrm{Ar}}$ are mean values of the α_n from all the analyzed $\mu\mathrm{Ar}(n\to1)$ transitions except $\mu\mathrm{Ar}(6\text{-}1)$. The fractions $\alpha_{p\mathrm{Ar}}$ of x rays due to excited-state transfer in prompt events represent about two-thirds in hydrogen mixtures, and about one-third in the case of deuterium mixtures.

B. Neon

The measurements of the $\mu\mathrm{Ne}$ x rays were performed in mixtures of hydrogen with five different neon concentrations, and in deuterium with two different neon concentrations. Pressures varied between 7 and 15 bar. The prompt Lyman intensities were determined up to the $\mu\mathrm{Ne}(6\text{-}1)$ transition in hydrogen mixtures, and up to $\mu\mathrm{Ne}(7\text{-}1)$ in deuterium mixtures. The higher Lyman transitions had prompt intensities too low to provide a significant value of α_n. Background lines of muonic x rays from the iron of the target walls contaminated the $\mu\mathrm{Ne}(5\text{-}1)$ and $\mu\mathrm{Ne}(6\text{-}1)$ transitions. Resulting from direct muon capture in iron, these x rays were prompt with regard to the entering muons. Their

intensities could be estimated from other measurements performed under similar experimental conditions. For instance, we used $\mathrm{H}_2+\mathrm{N}_2$ and $\mathrm{D}_2+\mathrm{N}_2$ mixtures to determine the $\mu\mathrm{Fe}(3\text{-}2)$ intensities around 265 keV. Taking the $\mu\mathrm{Cu}(3\text{-}2)$ transitions at 331 keV in the neon and nitrogen mixtures as reference lines, we could thus estimate the iron intensity in the neon mixtures and deduce the part due to contamination in the $\mu\mathrm{Ne}(5\text{-}1)$ and $\mu\mathrm{Ne}(6\text{-}1)$ prompt intensities.

The P_n and α_n values of measurements performed at three different pressures are shown in Table IV. Only the first five Lyman transitions are explicitly mentioned, although a $\mu\mathrm{Ne}(7\text{-}1)$ prompt intensity was determined in the deuterium mixture. The relative intensity patterns T_n following ground-state transfer from hydrogen and deuterium atoms to neon were taken from Ref. [8]. The measured pattern C_n of direct capture in neon shown in Table I was used to determine the α_n values. Table V shows the $\alpha_{p\mathrm{Ne}}$ and $\alpha_{d\mathrm{Ne}}$ fractions, which are the average values of the α_n's of the Lyman transitions up to the $\mu\mathrm{Ne}(6\text{-}1)$ in both cases. For the six $\mathrm{H}_2+\mathrm{Ne}$ mixtures, the mean values $\alpha_{p\mathrm{Ne}}$ amount to about 40%, whereas the corresponding $\alpha_{d\mathrm{Ne}}$ for the two $\mathrm{D}_2+\mathrm{Ne}$ mixtures are about half as small.

C. Nitrogen

Two measurements of muonic nitrogen x rays were performed in hydrogen mixtures, and one in a deuterium mixture. The relative intensity patterns T_n for transfer from μp and μd atoms to nitrogen were determined from delayed energy spectra. The T_n values for hydrogen and deuterium

TABLE VI. Relative intensities T_n for ground-state transfer from both μp and μd atoms to nitrogen, as well as prompt relative intensities P_n and α_n values for the transitions $\mu\mathrm{N}(n\to1)$ measured in hydrogen and in deuterium mixtures at 5 bar and room temperature.

Transition N	H$_2$+1.99%N$_2$			D$_2$+1.07%N$_2$		
	T_n	P_n	α_n	T_n	P_n	α_n
(2-1)	0.363(14)	0.727(59)	0.24(14)	0.370(9)	0.820(75)	0.05(17)
(3-1)	0.252(5)	0.105(7)	0.20(4)	0.252(5)	0.069(19)	0.03(11)
(4-1)	0.263(5)	0.075(5)	0.19(3)	0.251(5)	0.031(11)	0.00(5)
(5-1)	0.122(3)	0.065(5)	0.44(6)	0.127(3)	0.046(15)	0.23(19)
ΣLyman	1.000	1.000		1.000	1.000	

TABLE VII. α_{pN} and α_{dN} fractions as well as Coulomb capture ratios $A(H_2,N)$ and $A(D_2,N)$ determined in hydrogen and deuterium mixtures at given pressure and room temperature.

N_2 concentration (%)	Pressure	α_{pN}	$A(H_2,N)$	α_{dN}	$A(D_2,N)$
0.473	15	0.26(6)	0.26(6)		
1.99	5	0.23(5)	0.24(4)		
1.07	5			0.02(4)	0.22(2)
Mean		0.24(4)	0.25(3)	0.02(4)	0.22(2)

mixtures shown in Table VI were determined from measurements at 5 bar. The pattern T_n for transfer from μp to nitrogen is in good agreement with this of an older measurement [12]. In both hydrogen and deuterium mixtures, the highest Lyman transition clearly observed was the $\mu N(5-1)$ transition, which is in good agreement with the predictions of Holzwarth and Pfeiffer [13]. Hence we could consider the transitions up to $\mu N(5-1)$ in both cases to determine the α_n values. The pattern C_n of direct capture in nitrogen used to determine the α_n values is given in Table I.

The P_n patterns and the α_n values for the two mixtures are also presented in Table VI, and the α_{pN} and α_{dN} fractions are summarized in Table VII for all the measurements with nitrogen. Whereas the α_{pN} fractions represent about one-quarter of the prompt muonic intensity of μN, one notices that the α_{dN} value is zero within the limits of the uncertainties. Hence no transfer seems to occur from the excited states of the μd atom to nitrogen.

D. Capture ratios

The Coulomb capture ratios $A(H_2,Z)$ and $A(D_2,Z)$ were evaluated using Eq. (3.1) for all the mixtures measured. The number of prompt x-ray events of the Lyman series of μZ, N_P, was in every evaluated mixture higher than 25 000 counts. The correction of the delayed intensities due to the decay channels other than ground-state transfer to Z, $(\phi c_Z \lambda_{p(d)Z})/\lambda$, ranged from a few percent to 50% in the extreme case of the $H_2 + 0.69\%$ Ne mixture. By using one single or two exponential functions to fit the delayed part of the time spectra of the $H_2 + Ar$ and $D_2 + Ar$ mixtures (see Ref. [7]), the variation of the capture ratios did not exceed 2% for $A(H_2,Ar)$ and 4% for $A(D_2,Ar)$. This was taken into account by adding a systematic error to the measured intensities. The transfer rates used for the determination of the capture ratios were those of Ref. [7] for argon, and of Ref. [8] for neon. The transfer rate $\lambda_{pN} = 0.34(7)$ 10^{11} s^{-1} of Ref. [12] was used for the capture ratios $A(H_2,N)$, but $\lambda_{dN} = 1.00(30)$ 10^{11} s^{-1} from the same reference (measurement with poor statistics) was replaced by a more accurate value, $\lambda_{dN} = 1.45(2)$ 10^{11} s^{-1}, deduced from the present measurements.

Tables III, V, and VII show the results of the capture ratios $A(H_2,Z)$ and $A(D_2,Z)$ determined in $H_2 + Z$ and $D_2 + Z$ mixtures at different pressures and with different concentrations of argon, neon, and nitrogen. These per-atom (-molecule) capture ratios are fairly similar for a same element measured in hydrogen and in deuterium. Table VIII shows values for the capture ratio $A(H_2,D_2)$ deduced from the ratios $A(H_2,Z)/A(D_2,Z)$ for argon, neon, and nitrogen

measurements, with a mean value of $A(H_2,D_2) = 1.10(7)$.

VI. DISCUSSION

A. Excited-state transfer

The prompt intensity patterns P_n determined in hydrogen mixtures are observed to remain constant when increasing the concentration of the element Z up to a factor of 7 in the case of argon, and a factor of 4 in the cases of neon and nitrogen. In addition, they do not vary with pressures between 5 and 15 bar. Hence all our observations show that in our experimental range the patterns P_n are independent on both the relative concentration of the element Z and the total gas pressure. On the other hand, the patterns P_n depend on the element Z and on the hydrogen isotope used. Similar conclusions were already drawn for the intensity patterns T_n following ground-state transfer, e.g., to neon [6], argon [8], or oxygen [14].

For several mixtures, some transitions ($n \rightarrow 1$) yield individual values α_n which are systematically higher than the respective α_{pZ} or α_{dZ} mean values. In the two $D_2 + Ar$ mixtures, e.g., the α_3 value is about twice as high than the mean fraction α_{dAr}. The same is true for the α_5's in the two $H_2 + N_2$ mixtures. In addition, in the $H_2 + Ne$ mixtures the α_3's exceed the mean value α_{pNe} by more than two standard deviations in the six mixtures measured. These systematic discrepancies are interpreted as a limit to our hypothesis that the excited-state transfer pattern T_n^* is equal to the ground-state transfer pattern T_n. This assumption yields good results on average, but the above-mentioned particular Lyman transitions show that the excited-state transfer patterns T_n^* cannot be considered strictly the same as the ground-state transfer patterns T_n. Nevertheless, one concludes that excited-state transfer favors the population of low-l states of μZ.

From Tables III, V, and VII, one sees that most of the fractions α_{pZ} and α_{dZ} for argon, neon, and nitrogen, respec-

TABLE VIII. Mean capture ratios $A(H_2,Z)$ and $A(D_2,Z)$ for nitrogen, neon, and argon, as well as deduced capture ratios $A(H_2,D_2)$. For comparison, pionic capture ratios $A_\pi(H_2,Z)$ of Ref. [18] are given.

Element	$A(H_2,Z)$	$A_\pi(H_2,Z)$	$A(D_2,Z)$	$A(H_2,D_2)$
N	0.25(3)	0.303(14)	0.22(2)	1.14(17)
Ne	0.25(1)	0.261(12)	0.24(3)	1.04(14)
Ar	0.19(1)	0.172(6)	0.17(1)	1.12(9)
Mean				1.10(7)

tively, measured under different experimental conditions, scatter around mean values; no real dependence on concentration or pressure can be found within the limits of the uncertainties. The value $\alpha_{p\mathrm{Ar}}$ indicates that more than 60% of the prompt muonic intensity of $\mu\mathrm{Ar}$ is due to excited-state transfer, whereas the corresponding fractions amount to 43% in Ne and 24% in N_2. A significant difference is found between the fractions α_{pZ} measured in hydrogen and α_{dZ} measured in deuterium. In the cases of neon and argon, the quantities α_{dZ} are about half as small as α_{pZ}. For nitrogen, $\alpha_{d\mathrm{N}}$ is surprisingly compatible with zero, meaning that no transfer occurs from the excited states of the μd atom to nitrogen.

Because a higher concentration c_Z implies an increase of the probability of both direct capture in Z and transfer to Z, we expect the fractions α_{pZ} and α_{dZ} to be stable with regard to the concentration c_Z. In addition, C_n as well as T_n intensity patterns were shown to depend only weakly on parameters such as relative concentrations or pressure of the mixtures [3–8]. As the P_n patterns do not depend on our experimental conditions either, this also leads to constant α_{pZ} and α_{dZ} fractions.

To our knowledge, no predictions for transfer rates from excited states of muonic hydrogen and deuterium, λ_{pZ}^* and λ_{dZ}^*, to elements with $Z>2$ are available (except from the $2s$ state). If explicit transfer rates cannot be deduced from the α_{pZ} or α_{dZ} fractions, qualitative comparisons of the transfer rates from excited μp^* and μd^* atoms to Z are nevertheless possible. Indeed, the fractions α_{pZ} for nitrogen, neon, and argon are systematically higher than the fractions α_{dZ}, meaning that more excited-state transfer with regard to direct capture has occurred to an element Z from μp^* atoms than from μd^* atoms. Because of the comparable values of the capture ratios $A(\mathrm{H}_2,Z)$ and $A(\mathrm{D}_2,Z)$, we conclude that the transfer rates λ_{pZ}^* are systematically higher than the transfer rates λ_{dZ}^* for nitrogen, neon, and argon. Regarding ground-state transfer rates, differences between μp and μd atoms have already been observed: in the case of oxygen [14] and argon [7], λ_{pZ} is higher than λ_{dZ}, contrary to nitrogen (see Sec. IV D), neon [8] and sulfur [14].

For elements Z which differ in their capture ratios $A(\mathrm{H}_2,Z)$, a comparison of excited-state transfer rates λ_{pZ}^* is made possible by using a normalization. The number of muons transferred to Z from excited μp^* atoms is given by the number of muons directly captured by Z multiplied by the factor $\alpha_{pZ}/(1-\alpha_{pZ})$ [see explanations for Eq. (3.1)]. Since our results do not depend on the experimental conditions, a concentration $c_Z=1\%$ is chosen as a reference. The proportion of muons transferred from excited states to the number of μp^* atoms initially formed can be interpreted as the probability that a muon is transferred to a Z atom from one of the excited states of the μp^* atom. In a reference mixture $\mathrm{H}_2+1\%$ Z, this probability is evaluated to be around 9% for argon, 3% for neon, and 1% for nitrogen. With the excited-state transfer rates λ_{pZ}^* being proportional to this probability, we conclude that $\lambda_{p\mathrm{Ar}}^*>\lambda_{p\mathrm{Ne}}^*>\lambda_{p\mathrm{N}}^*$. This is not true for the ground-state transfer rates. The value $\lambda_{p\mathrm{Ne}}$, e.g., is more than one order of magnitude smaller than $\lambda_{p\mathrm{Ar}}$ and $\lambda_{p\mathrm{N}}$.

Since the probabilities of excited-state transfer from the μd^* atoms are evaluated to be around 3% for argon, 1% for

neon, and 0% for nitrogen, corresponding arguments in the case of muonic deuterium lead to a similar progression for excited-state transfer rates.

In our analysis, every excited-state transfer event was considered as prompt with regard to a muon entering the target. Actually, a long-lived metastable μp_{2s} state in H_2 mixtures was predicted, with a lifetime of about 100 μs at 1 mbar [2]. Such a long lifetime may allow muons to be transferred from μp_{2s} to an element Z present in the mixture, thus yielding delayed x rays from μZ. The metastability of the μp_{2s} state is strongly dependent on the pressure of the mixture, because the μp_{2s} atoms quickly decay to the ground state via the $2p$ state after collisions with ordinary H_2 molecules. The μp_{2s} lifetime decreases, due to collisions, from 100 μs at 1 mbar to 20 ns at pressures of 5 bar, and to 7 ns at pressures of 15 bar. In the case of a mixture H_2+Z, the lifetime of μp_{2s} is further reduced by muon transfer to Z. The transfer rates from μp_{2s} to Z are predicted to exceed the ground-state transfer rates λ_Z by a large factor, i.e., by 1–2 orders of magnitude [15]. In our mixtures, the lifetime of the μp_{2s} state can be estimated to be about 14 ns at 5 bar and 6 ns at 15 bar. Hence, transfer from the μp_{2s} metastable state to an element Z should produce x rays which cannot be distinguished from the prompt x rays within our time resolution.

Besides excited-state transfer, one cannot *a priori* exclude that part of the prompt muonic x-ray result from ground-state transfer from hot (E>1eV) muonic hydrogen atoms. Monte Carlo simulations, using the program TRANSFER written by Adamczak [16], showed however, that such x rays would significantly contribute to the measured prompt x rays only in the case of a resonant process, i.e., if the transfer rate from the hot μp's were at least one order of magnitude larger than the rate from thermal muonic hydrogen atoms.

B. Capture ratios

For the Coulomb capture ratios determined in H_2+Z and D_2+Z mixtures (Tables III, V, and VII), only the intensities of the μZ Lyman series were needed, so that the uncertainties in efficiency and absorption remain small in comparison to methods using the x-ray intensities from two different elements (see, e.g., Ref. [17]). Our results for argon and nitrogen mixtures show that the per-atom (-molecule) capture ratios do not depend on the pressure or relative concentrations within our experimental range. In the case of neon, some variations are noticed, but a clear dependence on pressure or concentration within the given uncertainties cannot be deduced. Hence mean capture ratios $A(\mathrm{H}_2,Z)$ and $A(\mathrm{D}_2,Z)$ are meaningful for argon, neon, and nitrogen (Table VIII).

An invariance of these capture ratios with regard to the concentration c_Z is expected, because all mixtures contained at least 97% of hydrogen or deuterium, and the capture ratios are known to vary only with significant changes in relative concentration. For instance, the capture ratios $A(\mathrm{Ar},\mathrm{Ne})$ measured in mixtures of neon and argon show variations only when the proportion of one gas to the other changes drastically, from 1:1 to 4:1, e.g., Ref. [17].

Our result for $A(\mathrm{H}_2,\mathrm{Ar})=0.19(1)$ is in better agreement with the corresponding pion capture ratio $A_\pi(\mathrm{H}_2,\mathrm{Ar})$
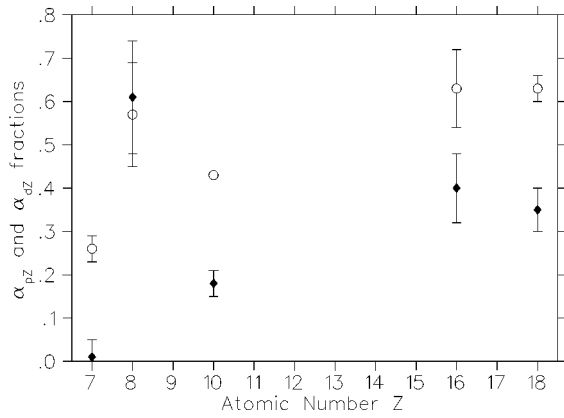
FIG. 2. α_{pZ} (circles) and α_{dZ} (diamonds) fractions in function of the atomic number Z. For $Z=7$, 10, and 18, these fractions are the mean values from our measurements, and for $Z=8$ and 16, they were determined from the results of Ref. [14].

$=0.172(5)$ [18] than an earlier estimate [19], where excited-state transfer was neglected. In addition, our capture ratios $A(H_2,Ne)$ and $A(H_2,N)$ are also in satisfactory agreement with the corresponding pion capture ratios, where excited-state transfer is included (see Table VIII).

For each mixture with deuterium, the per-atom (-molecule) capture ratio $A(D_2,Z)$ is also provided in Table VIII. As expected, because of the electromagnetic nature of the capture process, these ratios are observed to be very similar when measured in hydrogen or deuterium.

Finally, one can extract capture ratios $A_Z(H_2,D_2)$ between H_2 and D_2 molecules using the capture in Z as a reference. The results are shown in Table VIII for nitrogen, neon, and argon. All three ratios are somewhat higher than unity, with a mean value of $A(H_2,D_2)=1.10(7)$. Hence, an experimental value for the Coulomb capture ratio of negative muons between hydrogen and deuterium can be provided.

VII. CONCLUSION

In measuring muonic x rays in hydrogen mixtures with a small admixture of an element Z where both energy and time information are provided, our study of excited-state charge transfer of negative muons from μp^* and μd^* atoms provide consistent results. For nitrogen, neon, and argon, the fractions α_{pZ} and α_{dZ} of prompt x rays due to excited-state transfer (or transfer from hot μp's and μd's, respectively) do not depend on the relative concentrations or on the total gas pressure in our range of experimental conditions ($0.3\% \leq c_Z \leq 3\%$, 5 bar $\leq P \leq 15$ bar).

Figure 2 summarizes the mean values obtained for the fractions α_{pZ} and α_{dZ}, including those for oxygen and sulfur. Although the fractions α_{pZ} and α_{dZ} generally increase with the charge number Z, our results, together with the data of Ref. [14] in H_2+SO_2 and D_2+SO_2 mixtures, show that they are not directly proportional to the charge number Z. For a given element, the fractions are systematically higher (or at most equal) in hydrogen mixtures than in deuterium mixtures.

The fractions α_{pZ} and α_{dZ} are used to evaluate the Coulomb capture ratios $A(H_2,Z)$ and $A(D_2,Z)$, respectively. The per-atom (-molecule) capture ratios for nitrogen, neon, and argon are independent of relative concentration and pressure in our experimental range, and also in agreement with the corresponding pion capture ratios. In addition, an indirect value for the capture ratio of muons between hydrogen and deuterium molecules could be deduced, which is not incompatible with unity.

No quantitative estimate of excited-state transfer rates λ_{pZ}^* or λ_{dZ}^* can be given with our analysis. However, with the knowledge of the capture ratios $A(H_2,Z)$, a comparison of these transfer rates is possible. From our data, we conclude that the excited-state transfer rate from protium to nitrogen is smaller than the one to neon, and the excited-state transfer rate from protium to neon is smaller than the one to argon. The same conclusion is drawn for excited-state transfer rates from deuterium. In addition, for a given element, the excited-state transfer rate is found to be systematically higher (or at least equal) from μp^* than from μd^* atoms.

ACKNOWLEDGMENTS

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