Investigation of charge transfer from excited muonic hydrogen atoms to argon

Y.-A. Thalmann, R. Jacot-Guillarmod, F. Mulhauser, L. A. Schaller, L. Schellenberg, H. Schneuwly, S. Tresch,

and A. Werthmüller

Institut de Physique de l'Université de Fribourg, CH-1700 Fribourg, Switzerland

(Received 18 November 1996)

A method is proposed for investigating muon transfer from excited muonic hydrogen atoms to an element Z>2, and is applied to argon measurements. It makes use of a comparison between the muonic x-ray intensity patterns of the Lyman series of this element measured in mixtures with and without hydrogen. The analysis of the data taken in two gas mixtures $H_2 + Ar$ at 15 bar with argon concentrations of 2% (0.3%), yields consistent results. In both mixtures, two thirds of the prompt argon x rays proceed from transfer from excited μp^* states. The Coulomb capture ratio determined in both mixtures yields a mean value of $A(H_2,Ar) = 0.21(2)$, and agrees with the corresponding pionic ratio. [S1050-2947(97)06207-0]

PACS number(s): 34.70.+e, 36.10.Dr, 82.30.Fi

I. INTRODUCTION

Charge transfer from the ground state of muonic protium and deuterium has already been extensively investigated [1–7]. To our knowledge, no data about muon transfer from excited states to elements with Z>2 have been reported as yet.

A muonic hydrogen atom in a highly excited circular state would have a lifetime of several tens of nanoseconds if it did not collide with ordinary atoms and molecules. In such collisions, it can deexcite by external Auger effect and by Stark mixing. In Coulomb deexcitations, the transition energy divides into the recoil of the heavy particle and to the muonic hydrogen atom, which can so gain tens of eV [8]. Hence, the μp systems in the ground state have a kinetic-energy distribution the further away from a thermal energy and the higher the hydrogen pressure [9].

From excited states of muonic hydrogen atoms, the muon can also be transferred to a collision partner [10]. These transfer rates to very light elements are comparable with the rates of competing deexcitation processes [11,12], whereas those to heavier elements such as neon are estimated to be much higher than the corresponding transfer rates from the ground state [13].

In the present paper, we develop a method to investigate transfer from excited states. The proposed method, which requires a knowledge of the muonic x-ray time structure of the investigated element Z, makes use of a comparison of the x-ray intensity pattern due to direct capture measured in pure Z, and the transfer pattern measured in mixtures of $H_2 + Z$. Two measurements performed in gaseous mixtures of hydrogen with argon serve as illustration. In addition, a value of the Coulomb capture ratio $A(H_2,Ar)$ more precise than a previous one [14] is given.

II. DESCRIPTION OF THE METHOD

Time spectra of muonic x rays measured in mixtures of hydrogen with a small admixture of element Z have been used for more than 20 years to investigate the transfer process from the ground state of muonic hydrogen atoms [1–7]. Figure 1 shows such a time distribution, typically composed

of a prompt peak and a delayed part. In a usual experimental setup, muons entering the target vessel cross a plastic scintillator which gives a start signal to a time-to-digital converter. The stop signal is given by the x-ray detection, usually by a Ge diode with a good energy resolution. Thus each event recorded by the data-acquisition system includes a time information besides the energy information. Background subtracted time spectra are then constructed off-line for the measured muonic transitions by setting windows in the energy spectrum (a more detailed description can be found in Ref. [6]).

Muons slowed down in the $H_2 + Z$ gas mixture are trapped either by hydrogen or by a Z atom. The muonic x rays resulting from direct capture in Z are prompt events. Muonic hydrogen atoms are originally formed in highly excited states. They deexcite very fast, in about 10^{-10} s in a gas of several bar [9]. If the muon does transfer from hydrogen in an excited state to a Z atom, the resulting x rays also appear as prompt events, the sum of cascade and transfer times being shorter than the time resolution of the detection system. Otherwise the muonic hydrogen atom reaches the



FIG. 1. Typical time spectrum after background subtraction. Here the μ Ar(2-1) transition measured in H₂ + 2% Ar at 15 bar. The solid line represents a fit made with the sum of a prompt Gaussian peak (dashed line) and a delayed exponential part (dotted line).

ground state. It may collide with a Z atom and transfer its muon. In this latter case, the resulting muonic x rays are delayed relative to those from direct capture and constitute the delayed part of the time distribution.

By comparing the relative intensity structure of the prompt events belonging to the transitions of the Lyman series of argon measured in H_2 + Ar mixtures with the direct capture pattern measured in pure argon, a significant difference is noticed. Such a difference has already been observed for sulphur and oxygen in the SO₂ molecule in mixtures with and without hydrogen [7]. For the above-mentioned reasons, we assume that this difference is due to muon transfer from excited states. Hence, in H_2 + Ar mixtures, the prompt muonic argon x-ray intensity p_n is, for each $\mu Ar(n \rightarrow 1)$ transition, a superposition of direct muon capture events in argon, c_n , and events following muon transfer from excited μp^* states, t_n^* :

$$p_n = t_n^* + c_n$$
. (2.1)

If one could subtract those events due to direct muon capture from the prompt events, one would be left with events from excited-state transfer.

For the sum over the Lyman series, one can write

$$\sum p_n = \sum t_n^* + \sum c_n.$$
 (2.2)

When normalizing the intensities of the prompt, transfer, and direct capture events such that $\Sigma P_n = \Sigma T_n^* = \Sigma C_n = 1$, Eq. (2.2) writes

$$\sum P_n = \alpha \sum T_n^* + \beta \sum C_n.$$
 (2.3)

With this normalization, one has $\beta = 1 - \alpha$ for the relative intensities P_n , T_n^* , and C_n of the transitions of the Lyman series, and one can write

$$P_{n} = \alpha T_{n}^{*} + (1 - \alpha) C_{n}. \qquad (2.4)$$

Every transition can therefore be used to determine the fraction α of the prompt events which is due to muon transfer from excited μp^* states.

In Eq. (2.4), the relative prompt x-ray intensities, P_n , are those measured in $H_2 + Z$ gas mixtures. The relative intensities C_n are determined from a measurement of direct muon capture in element Z in pure Z or a gas mixture without hydrogen. The relative intensities T_n^* from transfer from excited states are unknown yet.

The muonic x-ray intensity pattern resulting from muon transfer from the ground state of muonic hydrogen to argon is well known [6,15]. The population of l states is predicted by theory [16] and explicit calculations show that low-lstates are favored [17]. The measured relative intensities are in satisfactory agreement with the calculated ones [6,15]. For energy reasons, transfer from excited μp^* atoms should occur to levels of the Z atom with higher principal quantum number n than from the ground state. The transfer mechanism being the same, one imagines that the population of low-l states is also favored. Cascade calculations [18] show



FIG. 2. Relative intensities of the lower transitions of the argon Lyman series. The prompt P_n and delayed T_n intensities are the result of a H₂ + 2% Ar measurement, the direct capture C_n ones are mean values of measurements performed in Ne + Ar mixtures.

by scaling the initial *l*-population that the relative intensities of the lower Lyman transitions of argon change by less than 6% if the transfer level is increased from n = 10 to 18. A similar observation is made in the case of neon between the levels n = 6 and 14.

Considering the above arguments and for lack of known relative intensities T_n^* , we consequently use the intensities of ground-state transfer T_n to replace the one from excited-state transfer T_n^* . The observation (Fig. 2) that the prompt relative intensities of the lower transitions of the Lyman series have values between the direct capture intensities and those following transfer from the ground state also strengthens this assumption.

The use of T_n instead of T_n^* leads to individual values of α , called α_n , for each $n \rightarrow 1$ transition. Thus Eq. (2.4) is replaced by

$$P_n = \alpha_n T_n + (1 - \alpha_n) C_n \,. \tag{2.5}$$

The meaning of these parameters α_n is the same as the one of α . If our approximation is justified, the values α_n should scatter around a mean value α for all transitions of the Lyman series.

III. ARGON MEASUREMENTS

To test the consistency of our method, we have analyzed measurements performed in two gas mixtures $H_2 + 1.98\%$ Ar and $H_2 + 0.284\%$ Ar at 15 bar. The quoted concentrations are given as ratios of partial pressure of the mixture. The gas mixtures have been prepared at 150 bar by an industrial factory (Carbagas, Switzerland). The purity of each component was better than 5 ppm, and the precision of the concentration $\pm 1\%$. For both measurements, the time resolution of the μ Ar(2-1) transition was about 6-ns full width at

TABLE I. Relative intensities C_n for direct capture in argon measured at 15 bar, and calculated.

Transition	Ar+Ne Average	10% Ar+10% Ne +80% He	Pure Ar [21]	Calculation [23]
Ar(2-1)	0.9081(49)	0.903(14)	0.9102(86)	0.9098
Ar(3-1)	0.0389(4)	0.0436(13)	0.0416(34)	0.0392
Ar(4-1)	0.0108(2)	0.0120(8)	0.0102(35)	0.0111
Ar(5-1)	0.0087(2)	0.0084(7)	0.0050(38)	0.0065
ΣLyman	1.0000	1.0000	1.0000	1.0000

half maximum, with a signal-to-background ratio better than 10. We have determined the prompt and the delayed intensities of the argon Lyman series transitions by fitting background-subtracted time spectra.

The precise shape of the prompt peak is determined from a measurement in a gas mixture containing no hydrogen, namely Ne + Ar. The prompt peak is well fitted by a Gaussian curve, which represents the time response of the detection system. The time t=0 is defined as the centroid of the curve. We use a single exponential function convoluted with the Gaussian to fit the delayed part in order to determine the disappearance rate λ of the μp atoms in the ground state. The number of μp_{1s} atoms remaining at time t, $N_{\mu p}(t)$, is given by

$$N_{\mu p}(t) = N_{\mu p}^{0} e^{-\lambda t}, \qquad (3.1)$$

where $N_{\mu p}^{0}$ is the number of μp systems in the 1s state at time t=0. The total disappearance rate λ can be written as

$$\lambda = \lambda_0 + \phi(c_p \lambda_{pp\mu} + c_{\rm Ar} \lambda_{\rm Ar}), \qquad (3.2)$$

where λ_0 is the free muon decay rate, $\lambda_{pp\mu}$ the mesomolecule formation rate [19,20], λ_{Ar} the transfer rate to argon, ϕ the atomic gas density normalized to the atomic density of liquid hydrogen, and c_x the relative atomic gas concentrations. The rates $\lambda_{pp\mu}$ (2.5 ×10⁶ s⁻¹) and λ_{Ar} [1.63(9) ×10¹¹ s⁻¹ for thermalized μp] are normalized to the atomic density of liquid hydrogen (see Ref. [15]).

By fitting a time spectrum, we determine the disappearance rate λ and also the intensities of the prompt peak and the delayed part. The background subtraction is made with an average of two background time spectra gated with events around the respective Lyman line. Detector efficiency and absorption in the target wall are accounted for each transition. The relative intensities P_n and T_n , to be inserted in formula (2.5), are obtained by normalizing the sum of the Lyman series transitions to unity for both prompt and delayed muonic x rays.

The relative intensities for direct capture, C_n , are obtained from measurements in argon gas mixtures containing no hydrogen. The measured intensity patterns show a weak density or pressure dependence [21–23]. Cascade calculations reproduce this dependence by assuming a faster refill-

TABLE II. Relative intensities T_n due to transfer from the ground state, measured in H₂ + Ar mixtures, compared with results from delayed energy spectra method and with calculation.

	This work		Ref. [6]	Calculation
Transition	$H_2+2\%$ Ar	$H_2 + 0.3\%$ Ar	Average	[15]
Ar(2-1)	0.398(10)	0.421(12)	0.396(6)	0.417
Ar(3-1)	0.099(3)	0.0095(3)	0.102(1)	0.093
Ar(4-1)	0.067(3)	0.061(3)	0.063(1)	0.064
Ar(5-1)	0.102(5)	0.089(3)	0.095(2)	0.095
Σ Lyman	1.000	1.000	1.000	1.000

ing of the electron holes in denser media during the muonic cascade. In H_2 + Ar mixtures with low argon concentrations, the refilling proceeds essentially through collisions with hydrogen molecules, which have larger velocities than argon atoms. At a given pressure, the collision frequency of muonic argon atoms with hydrogen in H_2 + Ar mixtures is higher than with argon atoms in pure argon. Even if the refilling is not only governed by the speed of the hydrogen molecules but also by the number and the binding energies of the electrons in the collision partners, one might expect that the electron refilling is faster (or different in some other respect) in our H_2 + Ar mixtures than, e.g., in Ne + Ar or pure Ar.

To test whether the refilling of the electron holes through hydrogen collisions changes the muonic argon x-ray patterns, we have measured the relative intensities of the Lyman series in a gas mixture of 80% He + 10% Ne + 10% Ar at 15 bar. The refilling width through collisions with helium should be comparable to the one with hydrogen. The comparison of the measured intensity pattern with the relative argon intensities observed in Ne + Ar mixtures at 15 bar shows no significant differences (cf. Table I). Thus we use the mean values of the measurements in Ne + Ar mixtures as relative intensities C_n for direct capture.

For both mixtures $H_2 + 2\%$ Ar and $H_2 + 0.3\%$ Ar, the measured relative intensities of the muonic argon Lyman series due to transfer from the ground state of μp atoms are given in Table II (see Ref. [15] for the details of $H_2 + 0.3\%$ Ar measurement). Their absolute intensities together with the prompt intensities have been determined from time distributions (see Fig. 1). Only the intensities of the first four transitions are given, the μ Ar(6-1) transition being contaminated by background γ rays due to muons stopped in the target walls. They are in good agreement with those of Refs. [6,15], which are more precise, because they are the mean over four

TABLE III. Relative intensities for prompt events P_n measured in Ar + H₂ mixtures and α_n values for muonic Ar(*n*-1) transitions.

Transition	$H_2+2\%$ Ar	α_n	$H_2+0.3\%$ Ar	α_n
Ar(2-1)	0.601(22)	0.60(5)	0.570(27)	0.69(6)
Ar(3-1)	0.091(5)	0.86(9)	0.088(12)	0.87(22)
Ar(4-1)	0.044(3)	0.59(6)	0.051(9)	0.80(18)
Ar(5-1)	0.065(3)	0.60(5)	0.066(7)	0.71(9)
Σ Lyman	1.000		1.000	

measurements and have been determined from delayed energy spectra. They are also in agreement with those predicted by theory.

For both measurements, the relative intensities of the prompt events and the calculated values α_n are presented in Table III. The normalized intensity structures of the prompt argon x rays are very similar although the argon concentrations differ by almost one order of magnitude between the two mixtures. For each measurement, the values α_n form two statistically coherent sets. From Table III, for both mixtures we obtain mean values for the parameter α :

H₂+2.0% Ar
$$\alpha$$
=0.62±0.05,
H₂+0.3% Ar α =0.71±0.05.

We conclude that in both mixtures about two-thirds of the prompt events are due to muon transfer from excited states, even if the argon concentrations differ by one order of magnitude. The agreement between both mixtures was not unexpected, since the collision probabilities with an argon atom of both the free muon as well as the excited μp^* atom are proportional to the argon concentration.

IV. EVALUATION OF THE CAPTURE RATIO A (H₂,Ar)

The ratio of delayed to prompt x-ray intensities of the Lyman series in argon, together with the parameter α , allows us to estimate the capture ratio $A(H_2,Ar)$ in gas mixtures, since the muonic Lyman series transitions are almost exclusively radiative and the muonic cascade ends up always in the ground state of the atom. Because an important part of the prompt x rays proceeds from transfer from excited μp^* states, the formula proposed by Schneuwly [24] to evaluate the Coulomb capture ratio of negative muons in $H_2 + Z$ mixtures has to be modified. The new formula is

$$A(H_2, Ar) = \frac{c_{Ar}}{(1 - c_{Ar})} \frac{N_T \frac{\lambda}{\lambda - \lambda_0 - \phi c_p \lambda_{pp\mu}} + \alpha N_P}{(1 - \alpha) N_P}$$
(4.1)

where N_T is the sum over the Lyman series of the muonic argon x-ray yields resulting from transfer from the ground state of the μp atoms, and N_P the sum over the Lyman series of the prompt muonic argon x-ray events. $N_T\lambda/(\lambda-\lambda_0-\phi c_p\lambda_{pp\mu})$ corresponds then to $N^0_{\mu p}$, the number of μp atoms formed which reach the ground state, and αN_P to those which transfer their muon from an excited state. $(1-\alpha)N_P$ is the number of muonic argon atoms formed by direct capture. With the factor $c_{\rm Ar}/(1-c_{\rm Ar})$, which gives the ratio of the number of argon atoms to the number of hydrogen molecules per unit volume in the H₂ + Ar gas mixture, one obtains a per-atom (molecule) capture ratio.

We have determined the capture ratios for both mixtures. The uncertainty on these ratios is mainly determined by the error on the mean α values. The per molecule ratios are $A(H_2,Ar) = 0.19(3)$ for the $H_2 + 2\%$ Ar mixture and 0.22(3) for $H_2 + 0.3\%$ Ar, with a mean value of 0.21(2). This mean ratio is in satisfactory agreement with both the corresponding pion capture ratio $A_{\pi}(H_2,Ar) = 0.172(5)$ [25] and the prediction of the mesomolecular model, $A(H_2,Ar) = 0.250$, [26]. An earlier estimate of the ratio $A(H_2,Ar)$ by our group [14], where transfer from excited states was neglected, was about twice smaller.

V. CONCLUSION

The analysis of the data taken in two gas mixtures H_2 + Ar with very different argon concentrations shows that the results are consistent. Hence the proposed method can be used to study muon transfer from excited states of muonic hydrogen atoms to other elements. In particular, the assumption that the muonic x-ray intensity pattern of the lower Lyman series transitions (n < 6) resulting from excited-state transfer is approximately equal to the one from the ground-state transfer seems to be confirmed. In this case one can conclude that the population of low-*l* states is favored by excited-state transfer.

With the present analysis of muonic x-ray data in mixtures $H_2 + Ar$, we have obtained quantitative informations for muon transfer from excited states of muonic hydrogen to argon. Specifically, this shows that about one third of the prompt muonic x-ray intensity is due to direct muon capture by argon atoms, and two-thirds to muon transfer from excited states of muonic hydrogen. The Coulomb capture probability of a negative muon by an argon atom is about ten times larger than by a hydrogen atom. Hence the probability that during its deexcitation an excited μp^* atom transfers in collisions its muon to argon in a mixture $H_2 + 1\%$ Ar is about 10%, i.e., much higher than the relative concentration. This high probability indicates that in collisions of an excited μp^* atom with an argon atom the transfer rate must be higher than the deexcitation rate.

This analysis of argon measurements shows that the proposed method can be generalized to obtain estimates of both muon transfer from excited states and muonic capture ratios $A(H_2,Z)$ in mixtures of hydrogen with a small admixture of element Z > 1. The analysis of other measurements, where argon has been replaced by nitrogen or neon, as well as measurements where natural hydrogen has been replaced by deuterium, is in progress. A comparison of the capture ratios $A(H_2,Z)$ and $A(D_2,Z)$, and of the transfer from the ground and excited states between the two hydrogen isotopes will be of particular interest. These results, as well as an exhaustive analysis of argon measurements including the higher Lyman transitions (n > 5), will be the subject of a forthcoming paper.

ACKNOWLEDGMENTS

The authors would like to thank C. Piller and O. Huot for their precious help during the experiments. This work was partially supported by the Swiss National Science Foundation.

- A. Alberigi Quaranta, A. Bertin, G. Matone, F. Palmonari, A. Placci, P. Dalpiaz, G. Torelli, and E. Zavattini, Nuovo Cimento B 47, 92 (1967).
- [2] A. Placci, E. Zavattini, A. Bertin, and A. Vitale, Nuovo Cimento A 52, 1274 (1967).
- [3] H. Daniel, H. J. Pfeiffer, P. Stoekel, T. von Egidy, and H. P. Povel, Nucl. Phys. A 345, 409 (1980).
- [4] E. Iacopini, G. Carboni, G. Torelli, and V. Trobbiani, Nuovo Cimento A 67, 201 (1982).
- [5] V. M. Bystritskii, V. P. Dzhelepov, V. I. Petrukhin, A. I. Rudenko, V. M. Suvorov, V. V. Filchenkov, N. N. Khovanskii, and B. A. Khomenko, Zh. Eksp. Teor. Fiz. 84, 7257 (1983) [Sov. Phys. JETP 57, 728 (1983)].
- [6] R. Jacot-Guillarmod, F. Bienz, M. Boschung, C. Piller, L. A. Schaller, L. Schellenberg, H. Schneuwly, W. Reichart, and G. Torelli, Phys. Rev. A 38, 6151 (1988).
- [7] F. Mulhauser, H. Schneuwly, R. Jacot-Guillarmod, C. Piller, L. A. Schaller, and L. Schellenberg, Muon Catal. Fusion 4, 365 (1989).
- [8] L. Bracci and G. Fiorentini, Nuovo Cimento A 43, 9 (1978).
- [9] V. E. Markushin, Phys. Rev. A 50, 1137 (1994).
- [10] L. Bracci and G. Fiorentini, Nuovo Cimento A 50, 373 (1979).
- [11] W. Czaplinski, A. Gula, A. Kravtsov, A. Mikhailov, and N. Popov, Phys. Rev. A 50, 525 (1994).
- [12] A. V. Kravtsov and A. I. Mikhailovi, Phys. Rev. A 49, 3566 (1994).
- [13] G. Fiorentini and G. Torelli, Nuovo Cimento A 36, 317 (1976).
- [14] R. Jacot-Guillarmod, F. Bienz, M. Boschung, C. Piller, L. A.

Schaller, L. Schellenberg, H. Schneuwly, and W. Reichart, Phys. Rev. A **39**, 387 (1989).

- [15] R. Jacot-Guillarmod, F. Mulhauser, C. Piller, L. A. Schaller, L. Schellenberg, H. Schneuwly, Y.-A. Thalmann, S. Tresch, A. Werthmüller, and A. Adamczak, Phys. Rev. A 55, 3447 (1997).
- [16] S. S. Gershtein, Zh. Eksp. Teor. Fiz. 43, 706 (1962) [Sov. Phys. JETP 16, 501 (1963)].
- [17] G. Holzwarth and H. J. Pfeiffer, Z. Phys. A 272, 311 (1978).
- [18] V. R. Akylas and P. Vogel, Comput. Phys. Commun. 15, 291 (1978).
- [19] G. Conforto, C. Rubbia, E. Zavattini, and S. Focardi, Nuovo Cimento 33, 1001 (1964).
- [20] V. M. Bystritskii, V. P. Dzhelepov, V. I. Petrukhin, A. I. Rudenko, V. M. Suvorov, V. V. Filchenkov, G. Khemnits, N. N. Khovanskii, and B. A. Khomenko, Zh. Eksp. Teor. Fiz. **70**, 1167 (1976) [Sov. Phys. JETP **43**, 606 (1976)].
- [21] P. Ehrhart, F. J. Hartmann, E. Koehler, and H. Daniel, Z. Phys. A 311, 259 (1983).
- [22] J. D. Knight, C. J. Orth, M. E. Schillaci, R. A Naumann, F. J. Hartmann, and H. Schneuwly, Phys. Rev. A 27, 2936 (1983).
- [23] R. Jacot-Guillarmod, F. Bienz, M. Boschung, C. Piller, L. A. Schaller, L. Schellenberg, H. Schneuwly, and D. Siradovic, Phys. Rev. A 37, 3795 (1988).
- [24] H. Schneuwly, Nucl. Instrum. Methods Phys. Res. Sect. B 9, 97 (1985).
- [25] V. I. Petrukhin and V. M. Suvorov, Zh. Eksp. Teor. Fiz. 70, 1145 (1976) [Sov. Phys. JETP 43, 595 (1976)].
- [26] H. Schneuwly, V. N. Pokrovsky, and L. I. Ponomarev, Nucl. Phys. A **312**, 419 (1978).