Primary Populations of Metastable Antiprotonic ⁴He and ³He Atoms

M. Hori,¹ J. Eades,¹ R. S. Hayano,² T. Ishikawa,² J. Sakaguchi,² T. Tasaki,² E. Widmann,² H. Yamaguchi,² H. A. Torii,³

B. Juhász,⁴ D. Horváth,⁵ and T. Yamazaki⁶

¹CERN, CH-1211 Geneva 23, Switzerland

²Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

³Institute of Physics, University of Tokyo, Komaba, Meguro-ku, Tokyo 153-8902, Japan

⁴Institute of Nuclear Research of the Hungarian Academy of Sciences, H-4001 Debrecen, Hungary

⁵KFKI Research Institute for Particle and Nuclear Physics, H-1525 Budapest, Hungary ⁶RI Beam Science Laboratory, RIKEN, Wako, Saitama 351-0198, Japan

(Received 8 December 2001; published 7 August 2002)

Initial distributions of metastable antiprotonic ⁴He and ³He atoms over principal (*n*) and angular momentum (ℓ) quantum numbers have been deduced using laser spectroscopy experiments. The regions

n = 37-40 and n = 35-38 in the two atoms account for almost all of the observed fractions $[(3.0 \pm 0.1)\%]$ and $(2.4 \pm 0.1)\%]$ of antiprotons captured into metastable states.

DOI: 10.1103/PhysRevLett.89.093401

PACS numbers: 36.10.-k, 25.43.+t, 34.90.+q

A negatively charged particle X^- (such as μ^- , π^- , or \overline{p}) approaching an atom at electron-volt energies can displace one of its electrons, thereby forming an exotic atom [1]. Little is known about the principal (*n*) and angular momentum (ℓ) quantum numbers of the atom's primordial (i.e., initially occupied) states. It has been widely believed since the 1950s [2–4] that in exotic hydrogen and helium atoms, *n* will be distributed around the value

$$n \sim n_0 = \sqrt{M^*/m_e} \tag{1}$$

(M^* being the reduced mass of the X^- -nucleus system, and m_e the electron mass), which corresponds to the X^- orbital with the same radius and binding energy as that of the displaced 1s electron. In this paper, we describe measurements of the (n, ℓ) distributions of antiprotons captured into atomic states in metastable antiprotonic helium [5–8], including the isotopes \overline{p} ⁴He⁺ for which $n_0 = 38.3$ and \overline{p} ³He⁺ for which $n_0 = 37.1$.

These \overline{p} He⁺ states (see Fig. 1) have microsecond-scale lifetimes against annihilation, because their large ℓ values $(\ell \sim n-1)$ do not permit them to deexcite by Auger emission of the electron. Instead, the atoms slowly undergo a series of constant $v = n - \ell - 1$ radiative transitions (i.e., $\Delta n = \Delta \ell = -1$). They finally reach states which proceed rapidly to \overline{p} He²⁺ ions, which are immediately destroyed by collisional Stark effects. So far, the numbers of antiprotons populating two metastable states in \overline{p} ⁴He⁺ [namely, $(n, \ell) = (39, 35)$ and (37, 34)] have been measured [9,10] using laser spectroscopic methods [11] at the former LEAR facility at CERN. The two states represent only a small fraction of the totality of metastable states shown in Fig. 1; moreover, since for technical reasons [9] the populations could be measured only after a minimum delay of $t = 1.8 \ \mu s$ following atomic formation, the primary populations (those at t = 0) could not be reliably estimated. We study here a total of 20 transitions in \overline{p} ⁴He⁺

and \overline{p} ³He⁺ atoms starting at t = 300 ns, and estimate the primary populations in nearly all the metastable states.

These experiments were made at the Antiproton Decelerator at CERN, the setup [12] being an extension of our former one. Metastable \overline{p} He⁺ atoms were produced



FIG. 1. Energy level diagrams of \overline{p}^{4} He⁺ and \overline{p}^{3} He⁺ atoms. Wavelengths of the studied transitions are indicated in nanometers. The one at $\lambda = 670.8$ nm was not observed.

by injecting a 200-ns-long pulse of 4×10^7 antiprotons into a cryogenic target, containing either ⁴He (of purity 99.9999%) or ³He (99.997%) gas, at temperatures T =5.5–6.2 K and pressures P = 0.2-0.4 bar [corresponding] to an atomic density $\rho = (2-5) \times 10^{20} \text{ cm}^{-3}$]. By detecting the charged pions produced in the annihilation with Cherenkov counters, a delayed annihilation time spectrum (the spectrum of intervals between an antiproton arrival in the target and its annihilation) could be obtained. The atoms were irradiated by a resonant laser pulse at time t_1 , which induced transitions between adjacent pairs of metastable and short-lived states, thereby forcing the annihilations of the antiprotons populating the metastable member of the two [11]. This superimposed a sharp spike on the delayed annihilation time spectrum [see Figs. 2(a)-2(c)]. A dye laser pumped by a Nd:YAG laser was used to produce laser pulses with wavelengths of $\lambda =$ 287.4-726.1 nm, bandwidths of 3 GHz, pulse lengths of 5 ns, and power densities of $4-40 \text{ mJ/cm}^2$. This was enough to saturate typical transitions with dipole moments of 0.03–0.3 D, and maximize the intensity in the annihilation spike at t_1 . By varying the timing of the laser pulse between $t_1 = 0.3$ and 16 μ s and measuring the spike intensity at each t_1 , the population $P_{(n,\ell)}(t_1)$ (i.e., the number of antiprotons populating the parent state at t_1 , normalized to the total number of antiprotons stopped in the target) could be obtained. The experimental background consisted mostly of pions stopping in the material around the target and undergoing $\pi \rightarrow \mu \rightarrow e$ decay [13], the contribution of which was estimated using Monte Carlo simulations.

Measurements were made for the 12 transitions in \overline{p} ⁴He⁺shown in Fig. 1, and eight transitions in \overline{p} ³He⁺ (excluding the one at $\lambda = 670.8$ nm which could not be detected). The 529.6-nm and 625.6-nm transitions in \overline{p} ⁴He⁺ are between metastable states and usually cannot be observed; here we resolved them at target densities $\rho = 2 \times 10^{21}$ cm⁻³, where the resonance daughter state (37, 34) common to both transitions became short-lived due to collisional effects [14]. Similarly, the 524.2-nm and 620.5-nm transitions in \overline{p} ³He⁺ were measured in ³He targets at $\rho = 1 \times 10^{21}$ cm⁻³, where the lifetime of the state (36, 33) was shortened to $\tau \sim 60$ ns [15]. The



FIG. 2. Three delayed annihilation time spectra of \overline{p} ⁴He⁺ with annihilation spikes of the transition $(n, \ell) = (33, 32) \rightarrow (32, 31)$ induced at $t_1 = 1.4 \ \mu s$ (a), 5.4 μs (b), and 10.4 μs (c).

population in the state (38, 36) in \overline{p}^{4} He⁺ was probed by irradiating the atoms with two successive laser pulses, the first inducing the transition $(38, 36) \rightarrow (37, 35)$ at $\lambda =$ 528.8 nm and the second (10 ns later) the transition $(37, 35) \rightarrow (38, 34)$ at 726.1 nm. The resulting annihilation spike thus contained antiprotons which populated both the (38, 36) and the (37, 35) states. We derived the primary populations $P_{(n,\ell)}(t=0)$ by extrapolating the measured population evolutions to t = 0. These were relatively insensitive to density between $\rho = 2 \times 10^{20}$ and $2 \times$ 10^{21} cm⁻³. The statistical and systematic experimental errors on $P_{(n,\ell)}(t=0)$ varied between (5–20)% and (15-30)%, respectively, depending on the transition. The estimated primary populations differ from those of previous experiments [9,10] by a factor of 3-5, primarily due to the new information at early ($t < 1.8 \ \mu s$) times.

The state (40, 35) in \overline{p} ⁴He⁺ [see Fig. 3(a)] was found to have a primary population of (0.10 ± 0.02) %. This population decreased with a 1- μ s time constant, which indicates that very few metastable atoms occupy the region $n \ge 41$ in the v = 4 cascade. The population in (39, 35) decreased with a lifetime of 1.9 μ s; this value (which is longer than the $\tau_{(39,35)} = 1.4$ - μ s lifetime of that state [14]) results when the antiprotons initially occupying (40, 36) deexcite into (39, 35), and thus increase the population in the latter state. The corresponding lifetimes of (38, 35) and (38, 36)



FIG. 3. Time evolutions of the populations in $12 \overline{p}^{4}\text{He}^{+}$ [filled circles with error bars in (a)–(1)] and eight $\overline{p}^{3}\text{He}^{+}$ [filled triangles in (m)–(t)] states. The best fit of a cascade model is shown in solid lines.

were still longer ($\sim 2.2 \ \mu$ s), due to spontaneous deexcitation from (39, 36) and (39, 37). The populations in (37, 34) and (37, 35) decreased with 3- μ s-lifetimes, and had downward-bending profiles at early times caused by the large feeding from (38, 35) and (38, 36).

In contrast, very small primary populations were found in \overline{p} ⁴He⁺ states with $n \leq 36$. These populations increased [see Figs. 3(g)–3(l)] as antiprotons deexcited from higher, initially populated states, reaching a maximum at progressively later times t_{max} with decreasing *n*. The longestliving antiprotons were found in the lowest metastable state (33,32) in the v = 0 cascade; since antiprotons captured in the $n \sim 38$ regions made 6–7 radiative transitions before reaching this state, a significant population ($\sim 0.01\%$) could be observed even at $t_1 = 16 \ \mu$ s. The primary populations in the states (37, 36) and above in the v = 0cascade were estimated by fitting cascade models on the population evolution of the state (36, 35).

In \overline{p} ³He⁺ atoms [see Figs. 3(m)–3(t)], the population in the state (38, 34) decreased with a 1.0- μ s lifetime which (being similar to the theoretical state lifetime of $\tau_{(38,34)} =$ 1.0 μ s [16,17]) indicates that the populations in the region $n \ge 39$ in the v = 3 cascade are very small. The last transition (39, 34) \rightarrow (38, 33) in the v = 4 cascade at $\lambda =$ 670.8 nm was searched for, but could not be detected, which shows the populations in this cascade are also negligible. The largest populations $[P_{(n,\ell)}(t=0) \sim 0.3\%]$ were found in the states (36, 34), (36, 35), (37, 34), (37, 35), and (37, 36). In contrast to the \overline{p}^{4} He⁺ case, significant primary populations were detected even in low-n regions, with the lowest metastable state in the v = 1 cascade having a value of $P_{(34,32)}(t=0) = (0.06 \pm 0.02)\%$. The lowest state (32, 31) in the v = 0 cascade had, on the other hand, a negligible primary population. The populations in states which were not directly observed were estimated by fitting cascade models on the population evolutions of the measured states.

Information obtained from the delayed annihilation time spectrum of \overline{p}^{4} He⁺ (undisturbed by laser stimulation and measured at LEAR) [18] was also incorporated into the overall picture afforded by the laser studies. The spectrum (Fig. 4) showed that $f_{trap} = (3.0 \pm 0.1)\%$ of the antiprotons survive for more than 15 ns. In the \overline{p}^{3} He⁺ case, measurements gave a value of $(2.4 \pm 0.1)\%$ [19].

In Fig. 5, the distributions of primary populations (characterized by the *n* and *v* values) are shown. Nearly all the metastable \overline{p} ⁴He⁺ atoms lie in the region n = 37–40, with the n = 38 states containing the largest population. This appears to support the estimate $n_0 \sim 38$ given by Eq. (1). The primary populations of \overline{p} ³He⁺ atoms were distributed over a lower range of *n* values between 35 and 38, which is compatible with the estimation $n_0 \sim 37$. The sums of the individual populations in all the measured \overline{p} ⁴He⁺ and \overline{p} ³He⁺ states were $(3.5 \pm 0.8)\%$ and $(2.5 \pm 0.8)\%$, respectively, which account for the observed fractions of delayed annihilations described in the previous paragraph.



FIG. 4. Measured delayed annihilation time spectra of \overline{p}^{4} He, compared with simulated spectra for cases where the states are populated up to $n_{\text{max}} = 39$ (dashed lines), 40 (solid lines), 41 (dot-dashed lines), and 50 (dotted lines).

By energy conservation, the binding energy B_n of the populated atom is equal to $I_0 - T_{\overline{p}}m_{\text{He}}/(m_{\text{He}} + m_{\overline{p}}) + T_e$, I_0 being the ionization potential of helium (~ 24.6 eV), $T_{\overline{p}}$ and T_e the laboratory energies of the incoming antiproton and ejected electron, and m_{He} and $m_{\overline{p}}$ the helium and antiproton masses. Several theoretical calculations [20–25] (two such results are shown in Fig. 5) predict that T_e is small with respect to I_0 . Only antiprotons $T_{\overline{p}} \ll I_0$ will be captured into the $n_0 \sim 37-38$ states having binding energies $B_n \sim I_0$, whereas the more energetic antiprotons ($T_{\overline{p}} \sim 25 \text{ eV}$) are captured into much higher ($B_n \sim 0$) regions with *n* values exceeding 50. The calculations predict that $f_{\text{trap}} = (12-25)\%$ of the antiprotons should be captured into \overline{p}^4 He⁺ states (see Table I), with most of them in states above $n \geq 41$.



FIG. 5. Measured distributions of primary populations in \overline{p} ⁴He⁺(filled circles with error bars) and \overline{p} ³He⁺ (filled triangles). Theoretical \overline{p} ⁴He⁺ populations derived from diabatic state [20] (solid lines) and semiclassical coupled-channel [21] (dashed lines) calculations are shown.

	Metastable population (%)	
	Total	$n \le 40$
Diabatic state [20]	11.5	4.8
Coupled-channel [21]	23.9	1.8
Semiclassical Monte Carlo [24]	24.7	11
Fermion molecular dynamics [25]	20	
Expt.	3.0 ± 0.1	3.5 ± 0.8

TABLE I. Total fractions of antiprotons captured into \overline{p}^{4} He⁺states, and the fractions of those captured into the $n \le 40$ region: theoretical and experimental values. Direct theory-experiment comparison may be difficult (see text).

As these theoretical populations do not take the effect of collisions into account, they may not be directly comparable with the values presented here, which refer to atoms thermalized by many collisions. Nevertheless, as shown in Fig. 5, the experimental data for \overline{p}^{4} He⁺ atoms agree with the results of diabatic state calculations [20,22] in the region $n \leq 40$. We detected very little or no metastable population in the region $n \ge 41$, and due to this nonobservation the total metastable fraction was found to be $f_{\text{trap}} =$ 3%. One theoretical study [26] suggests that the \overline{p}^{4} He⁺ created in the $n \ge 41$ states recoil with such large kinetic energies that they are rapidly destroyed by collisions. Another study [27] suggests that the quenching cross sections for the $n \ge 41$ states are large even at thermal energies due to the low activation barrier in the \overline{p}^{4} He⁺-He potential. A recent work [28] predicts that the rates of collisional Stark transitions are several times larger in \overline{p}^{3} He⁺ compared to \overline{p}^{4} He⁺.

Finally, cascade calculations of \overline{p}^{4} He atoms were made using the measured primary populations, in an attempt to reproduce the delayed annihilation time spectrum shown in Fig. 4. We simulated the distributions of antiprotons among the metastable states in the region $n \le 40$ as shown in Fig. 5, and allowed them to cascade through individual states (using either the experimental state lifetimes [14,15] where available or the theoretical values [16,17]), and calculated the time elapsed until their annihilation. The simulated and measured spectra (Fig. 4) agree, including the 97% fraction of antiprotons that annihilate within 15 ns, and the 3% metastable atoms that decay with a slightly downward-bending structure having a mean lifetime of $\tau_{\rm mean} = 3 \ \mu s$. Simulated spectra for cases where the states are populated up to a maximum n value of 39, 41, and 50 are also shown, derived by appending the theoretical populations for the $n \ge 41$ states. The agreement with experiment was best for the case of $n_{\text{max}} = 40$, which supports the findings of the current study that the metastable populations in the $n \ge 41$ regions are very small.

We thank the CERN PS division for their help, and K. Ohtsuki, G. Ya. Korenman, and J. S. Cohen for many dis-

cussions. This work was supported by the Grant-in-Aid for Creative Basic Research (10NP0101) of Monbukagakusho of Japan, and the Hungarian Scientific Research Fund (OTKA T033079 and TeT-Jap-4/98).

- [1] E. Fermi and E. Teller, Phys. Rev. 72, 399 (1947).
- [2] G.A. Baker, Jr., Phys. Rev. 117, 1130 (1960).
- [3] T.B. Day, Nuovo Cimento 18, 381 (1960).
- [4] R. Landua and E. Klempt, Phys. Rev. Lett. 48, 1722 (1982).
- [5] G.T. Condo, Phys. Lett. 9, 65 (1964).
- [6] J.E. Russell, Phys. Rev. Lett. 23, 63 (1969).
- [7] M. Iwasaki et al., Phys. Rev. Lett. 67, 1246 (1991).
- [8] T. Yamazaki et al., Nature (London) 361, 238 (1993).
- [9] R. S. Hayano *et al.*, Phys. Rev. Lett. **73**, 1485 (1994); **73**, 3181(E) (1994).
- [10] F.E. Maas et al., Phys. Rev. A 52, 4266 (1995).
- [11] N. Morita et al., Phys. Rev. Lett. 72, 1180 (1994).
- [12] M. Hori et al., Phys. Rev. Lett. 87, 093401 (2001).
- [13] H. A. Torii *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **396**, 257 (1997).
- [14] M. Hori et al., Phys. Rev. A 58, 1612 (1998).
- [15] F.J. Hartmann et al., Phys. Rev. A 58, 3604 (1998).
- [16] V. I. Korobov and I. Shimamura, Phys. Rev. A 56, 4587 (1997).
- [17] I. Shimamura, Phys. Rev. A 46, 3776 (1992).
- [18] E. Widmann et al., Phys. Rev. A 51, 2870 (1995).
- [19] B. Ketzer et al., Phys. Rev. A 53, 2108 (1996).
- [20] K. Ohtsuki (private communication).
- [21] G. Ya. Korenman, Hyperfine Interact. 101-102, 81 (1996); Nucl. Phys. A692, 145c (2001).
- [22] J. S. Cohen, R. L. Martin, and W. R. Wadt, Phys. Rev. A 27, 1821 (1983).
- [23] V.K. Dolinov et al., Muon Cat. Fusion 4, 169 (1989).
- [24] W. A. Beck, L. Wilets, and M. A. Alberg, Phys. Rev. A 48, 2779 (1993); (private communication).
- [25] J.S. Cohen, Phys. Rev. A 62, 022512 (2000).
- [26] G. Ya. Korenman, Phys. At. Nucl. 59, 1665 (1996).
- [27] S. Sauge and P. Valiron, Chem. Phys. 265, 47 (2001).
- [28] J.E. Russell, Phys. Rev. A 65, 032509 (2002).