Laser Studies of the Decay Chain of Metastable Antiprotonic Helium Atoms

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Laser studies of metastable antiprotonic helium atoms, which we recently initiated by observing a sharp increase of the antiproton annihilation rate induced by laser-stimulated resonant transitions, have been extended. With a single laser tuned to the resonance already found at 597.26 nm, we have now established the time dependence of the upper state population. With two lasers ignited at variable time separation, we also studied the feeding of the upper state from higher atomic levels. The initial populations and level lifetimes of excited exotic atoms were determined.

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Recently, we observed laser-induced resonant transitions in antiprotonic helium atoms. These transitions occurred between metastable states and Auger dominated short lived states [1], and their observation unambiguously demonstrated that the anomalous longevity of antiprotons previously observed in helium media [2–5] results from the formation of high-n high angular momentum states of $\bar{p}\mathrm{He}^+$ [6,7]. The observed transition with vacuum wavelength 597.259 ± 0.002 nm was tentatively assigned to $(n,l) = (39,35) \rightarrow (38,34)$. The (n,l) assignment of [1] was based on the theoretical calculation that the $\Delta l \leq 3$ Auger transitions are fast [8], and has been used throughout this paper; the assumption does not, however, change the validity of our conclusions.

It is widely believed that, when an exotic atom is formed, one of the electrons in the target atom is replaced by the incoming particle, which occupies a state (n,l) of nearly the same energy (or nearly the same space) with the electron. This assumption leads to an estimate of the most probable principal quantum number of the initially formed state, $n \sim \sqrt{M^*/m_e}$ (in the case of $\bar{p}\mathrm{He}^+$, $n \sim 38$), where M^* is the reduced mass of the captured particle and m_e is the electron mass. So far, there is neither direct experimental information, nor realistic theoretical prediction, on the (n,l) distribution.

According to calculations [9,10], the main cascade sequence follows a propensity rule which keeps the radial node number (or the vibrational quantum number), $v = n - l - 1$, the same. The presently observed cascade is the "fourth circular" sequence, $v = 3$, namely, $(41,37) \rightarrow (40,36) \rightarrow (39,35) \rightarrow (38,34)$, of which the levels above $(39,35)$ are metastable; i.e., their Auger decay rates are much slower than the radiative rates. Hence, the antiprotons initially trapped into those states should cascade down mostly to $(39,35)$, and can contribute to the laser-resonance peak.

Such a chain-decay feature was already evident in the $\bar{p}$ delayed annihilation-time spectra. The time spectra $N(t)$ in low temperature helium gas are not a sum of single exponentials; when $\ln N(t)$ is plotted against t, it is observed to curve downwards at later times [3–5]. Functions based on a chain-decay model can reproduce such time spectra fairly well [11], providing indirect evidence for the chain decay.

In the present Letter we report results on the first attempt to determine the initial populations and level lifetimes of metastable $\bar{p}$ states, using two sequentially pulsed lasers. We set both laser wavelengths to the value 597.26 nm known to produce a sharp peak in the annihilation-time spectrum by stimulating transitions to a nonmetastable level.

Using a single laser and varying its trigger timing t_1, we could then measure the resonance peak intensity. This enables us to map out the time dependence of the $\bar{p}$

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population at the (39, 35) level, which we denote by $N_{39}(t)$. As mentioned above, $N_{39}(t)$ depends not only on the initial population and level lifetime of (39, 35), but also on those of higher-lying levels, which decay radiatively and feed the (39, 35) level.

The characteristics of these higher-lying levels could be determined by igniting the two lasers sequentially at times t_1 and $t_2(> t_1)$. The first laser depopulates the (39, 35) level at t_1. If the $\bar{p}$ occupies a higher-(n, l) level at $t = t_1$, it is unaffected by the first laser, but can cascade down to (39, 35) at later times, and can therefore contribute to the resonance peak at t_2 produced by firing the second laser. By varying the time difference between t_1 and t_2, the level populations and lifetimes of the states which feed (39, 35) can be determined.

The experimental arrangement is the same as in our previous work [1]. The 200 MeV/c $\bar{p}$ beam from CERN Low Energy Antiproton Ring (LEAR) was stopped in low temperature (5–10 K) helium gas at a pressure of 0.7–1 bar. If no annihilation signal was detected within 100 ns of $\bar{p}$ arrival, we assumed that a metastable $\bar{p}\mathrm{He}^+$ atom was present in the gas, and generated a laser trigger. Our two pulsed dye lasers were pumped at 308 nm by XeCl excimer lasers. The minimum delay between the $\bar{p}$ arrival and laser ignition was 1.8 μs; additional delay was added to the trigger signal when studying the time dependence of the resonance peak intensity at later times.

Figure 1 shows $\bar{p}$ delayed annihilation-time spectra in low temperature helium gas with one laser tuned at 597.26 nm. We show spectra obtained by varying the laser trigger timing t_1 from 1.8 to 6.8 μs (top to bottom in the figure). The continuum is due to the delayed annihilation of metastable $\bar{p}\mathrm{He}^+$ unaffected by the laser resonance. Peaks due to laser resonance appear at times corresponding to laser triggers.

Figure 2 shows $\bar{p}$ annihilation-time spectra obtained by igniting the first laser at $t_1 = 1.8$ μs, and the second laser at $\Delta t = t_2 - t_1 = 0.2, 0.5, 1, 2, 4$ μs. Two resonance peaks appear at t_1 and t_2 as shown. When Δt is small, the peak intensity at t_2 is weak, indicating the first laser already nearly emptied the (39, 35) level. As Δt increases, however, the second peak intensity also increases. This provides direct evidence for the existence of metastable levels which feed (39, 35). At still later times, the second peak intensity decreases once again.

The time dependence of resonance peak intensities (integrated peak area divided by the total delayed component) is presented in Fig. 3. Figure 3(a) shows the time dependence of the resonance intensity, corresponding to the spectra shown in Fig. 1. Figure 3(b) shows the peak intensity of the second laser resonance and corresponds to the spectra shown in Fig. 2, where t_1 was fixed at 1.8 μs and the second laser timing t_2 was varied. Figures 3(c) and 3(d) are similar to Fig. 3(b), obtained by setting t_1 at a different timing [$t_1 = 2.8$ μs for Fig. 3(c) and $t_1 = 3.8$ μs for Fig. 3(d)].

The data shown in Fig. 3 were used to obtain the initial populations and level lifetimes of metastable states, with the aid of a simplified chain-decay model. The model assumes, according to the v = const propensity rule, that there exists a single ladder which feeds the (39, 35) level via (40, 36), (41, 37), etc. It is evident from Fig. 2 that there is at least one state which feeds the (39, 35) level.

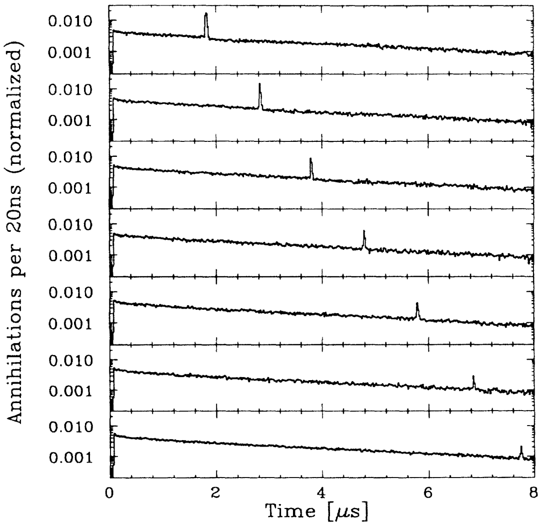

FIG. 1. Antiproton annihilation-time spectra in low temperature helium gas, obtained by varying the laser trigger times. The laser wavelength was fixed at the center of the resonance (597.26 nm) discovered in our previous work [1].

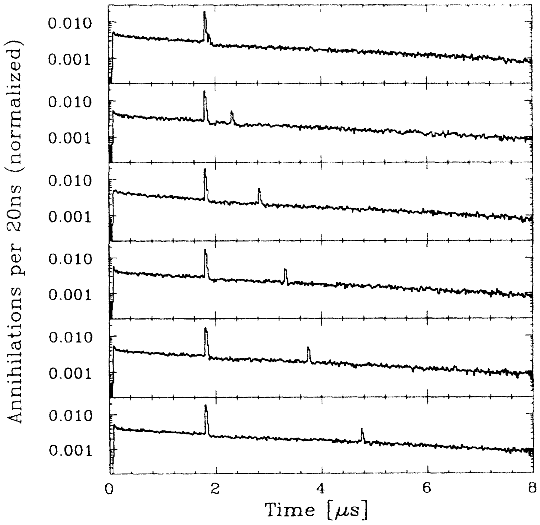

FIG. 2. Antiproton annihilation-time spectra in low temperature helium gas, obtained with two lasers both tuned at 597.26 nm. For all these spectra, the first laser was ignited at a fixed time $t_1 = 1.8$ μs. The second laser pulse was delayed with respect to the first by a range of values between $t_2 = t_1 + 50$ ns (top) and $t_1 + 3.0$ μs (bottom).

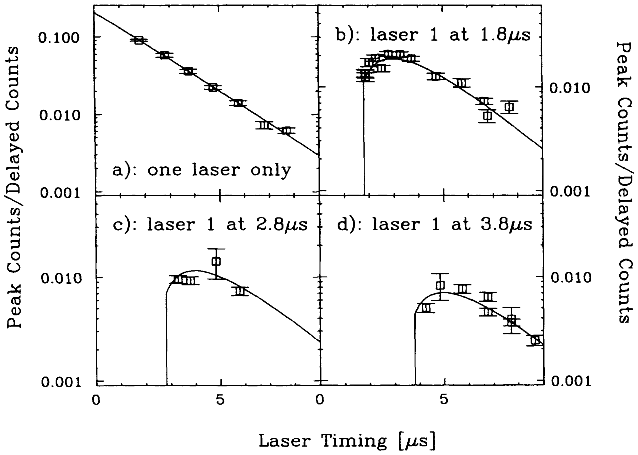

FIG. 3. Time variation of resonance intensity. (a) The intensity measured with one laser ignited at various times t_1. (b) The intensity of the peak produced by a second laser pulse at variable times t_2, when the first laser ignition was fixed at $t_1 = 1.8$ μs. (c), (d) Similar to (b), but with $t_1 = 2.8$ μs and $t_1 = 3.8$ μs.

Let us first try to fit the data shown in Fig. 3 with just one level [assumed to be (40, 36)] decaying to (39, 35). The fit parameters are the initial ($t = 0$) populations and lifetimes of these two levels, i.e., $N_{39}(0)$, $\tau_{39} = 1/\lambda_{39}$, $N_{40}(0)$, and $\tau_{40} = 1/\lambda_{40}$. We introduce another parameter ϵ which represents the efficiency of laser-induced depopulation; if the laser resonance completely empties the (39, 35) level, ϵ is 1.

In this model, the first resonance peak intensity at $t = t_1$ is then written as

$$\begin{aligned} I_1(t_1) &= \epsilon N_{39}(t_1), \\ N_{39}(t_1) &= \left[N_{39}(0) - \frac{\lambda_{40}}{(\lambda_{39} - \lambda_{40})} N_{40}(0) \right] e^{-\lambda_{39} t_1} \\ &\quad + \frac{\lambda_{40}}{(\lambda_{39} - \lambda_{40})} N_{40}(0) e^{-\lambda_{40} t_1}, \end{aligned}$$

which is used to fit the data in Fig. 3(a). The second resonance peak intensity at $t = t_2$ is

$$\begin{aligned} I_2(t_2) &= \epsilon N_{39}(t_2), \\ N_{39}(t_2) &= \Bigg[(1 - \epsilon) N_{39}(t_1) \\ &\quad - \frac{\lambda_{40}}{(\lambda_{39} - \lambda_{40})} N_{40}(t_1) \Bigg] e^{-\lambda_{39}(t_2 - t_1)} \\ &\quad + \frac{\lambda_{40}}{(\lambda_{39} - \lambda_{40})} N_{40}(t_1) e^{-\lambda_{40}(t_2 - t_1)}, \\ N_{40}(t_1) &= N_{40}(0) e^{-\lambda_{40} t_1}, \end{aligned}$$

which is used to fit the data in Figs. 3(b)-3(d). We performed simultaneous least squares fits to the data shown in Figs. 3(a)–3(d), using the functions given above, to deduce the initial populations and lifetimes of the (40, 36) and (39, 35) states. The resulting fits are shown by solid curves in Fig. 3, and best-fit parameters are summarized in Table I, from which it can be seen that the two-level model can fairly well represent the data. The efficiency ϵ was found to be 0.88.

We can proceed one step further by considering the contribution from (41, 37), which adds two more free parameters, $N_{41}(0)$ and $\tau_{41} = 1/\lambda_{41}$, to the model. The extension of the fit function to the three-level case is straightforward. As shown in Table I, the best-fit parameters of the three-level model are identical to those of the two-level model, with $N_{41}(0)$ being consistent with zero. This does not, however, mean that (41, 37) is not populated at all, but rather that N_{41} is small, and that our data do not have sufficient sensitivity for a precise determination of N_{41}. In any case, N_{40} may represent contributions from all the higher-lying levels which feed (40, 36).

The fit parameters show that $N_{39}(0)$ is (20 ± 2)% of the total delayed events and $N_{40}(0)$ is (12 ± 1)% of the total delayed events. Note, however, that the present models do not take account of side feeding and decay (for instance, $\Delta n = 2$ transitions). Theoretical estimations show that the sum of such branching ratios is of the order of 10% of the main stream, so that the N_{39} and N_{40} values should have additional uncertainty of (1–2)%. The overall fraction of the $v = 3$ metastable sequence is found to be 32% of total delayed events of which the majority is concentrated on (39, 35), a significantly large fraction in view of the likely presence of a number of possible metastable states.

The level decay rate of the (39, 35) state has been determined to be $\lambda_{39} = (7.23 \pm 0.24) \times 10^5$ s^{-1}. The decay rate of the (40, 36) state has also been determined to be $\lambda_{40} = (4.95 \pm 0.02) \times 10^5$ s^{-1}, but this may include cumulative contributions from upper states as discussed above. The observed level decay rates show fair agreement with the results of theoretical calculations, as shown in Table I. The agreement of the observed and calculated decay rates proves the theoretical results that the level lifetimes of (39, 35) and (40, 36) are dominated by radiative decays (the Auger rates are much smaller). The experiment also demonstrates the reduction of the dipole strength due to the $\bar{p}$-e^- correlation, as shown by the recent calculations based on the configuration mixing model [9] and on the molecular approach [10]; without the correlation taken into account [7], the calculated rates are 3 times larger.

In summary, we carried out a two-laser resonance experiment on antiprotonic helium atoms and demonstrated that there indeed exists a decay chain feeding the metastable (39, 35) state. By fitting the observed time dependence of the $\bar{p}$ population to a chain-decay model, we deduced the initial populations and decay rates of these metastable levels. The decay sequence $n - l - 1 = 3$ we

TABLE I. Results of the two-level and three-level fitting. The last column shows calculated level lifetimes which include side feeding [9,10].

	Two-level fit	Three-level fit	Calculated values
$N_{39}(0)$	(20 ± 2)%	(20 ± 2)%	
$N_{40}(0)$	(12 ± 1)%	(12 ± 1)%	
$N_{41}(0)$		$< 2\%$	
λ_{39}	$0.72 \pm 0.02\ \mu s^{-1}$	$0.72 \pm 0.02\ \mu s^{-1}$	$0.61\ \mu s^{-1}$
λ_{40}	$0.49 \pm 0.02\ \mu s^{-1}$	$0.49 \pm 0.02\ \mu s^{-1}$	$0.54\ \mu s^{-1}$
λ_{41}		...	
Reduced χ^2	2.4	2.4	

observed via laser resonance constitutes a substantial part of the total metastable fraction. Thus we confirm for the first time by direct observation the long-held belief that exotic atoms are initially formed at $n \sim \sqrt{M^*/m}$, at least for the metastable states. Furthermore, the observed strong concentration of the initial population on the $n = 39$ state appears surprising in comparison with the expected broad distribution of initial n [12] and calls for theoretical investigation.

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