First Observation of Laser-Induced Resonant Annihilation in Metastable Antiprotonic Helium Atoms

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We have observed the first laser-induced resonant transitions in antiprotonic helium atoms. These occur between metastable states and Auger dominated short lived states, and show that the anomalous longevity of antiprotons previously observed in helium media results from the formation of high-n high-l atomic states of \bar{p} He⁺. The observed transition with vacuum wavelength 597.259 \pm 0.002 nm is tentatively assigned to $(n, l) = (39, 35) \rightarrow (38, 34)$.

PACS numbers: 36.10.Gv

Unusual atoms with exotic constituents such as muons and positrons have always played an important role in the development of physics. One might wonder if exotic antiprotonic atoms would show similar promise. However, theory and experiment alike have generally implied that their lifetimes ought in all cases to be very short. The entire process from capture to annihilation from high-nS states should be over in less than a picosecond, placing antiprotonic atoms beyond the reach of many of the powerful modern techniques which have revolutionized atomic spectroscopy.

This conventional wisdom was called into question in 1991 when, in an experiment at KEK, Japan [1], a few percent of the antiprotons stopped in liquid helium were observed to have anomalously long lives, some of them annihilating up to 15 μ s after their entry into the liquid. This intriguing effect seems peculiar to helium, and has been further studied in the gaseous, liquid, and solid phases by some of the present authors using the Low Energy Antiproton Ring (LEAR) facility at CERN. Among the wealth of new results, we found a 14% reduction in the mean lifetime of these delayed annihilations when ³He was used to stop the antiprotons instead of 4 He [2,3]. This is in good agreement with an estimate [4] based on a model proposed by Condo [5] and theoretically studied by Russell [6], in which the antiprotons are trapped in large-n and large-l states of neutral antiprotonic helium \bar{p} He⁺. The initially formed states most likely have $n \sim n_0 = (M/m_e)^{1/2} \sim 38$, where M is the reduced mass of the \bar{p} He⁺ system. Here, Auger emission is strongly suppressed, because a large jump in n and l would be necessary to provide the energy ($\sim 24.6 \text{ eV}$, the ionization energy of a helium atom) required to liberate the remaining electron. The only competitive process is then a slow radiative cascade producing $\sim 2 \text{ eV}$ (visible region) photons. Furthermore, the presence of an electron in the \bar{p} He⁺ atom during this cascade strongly suppresses collisional quenching of the metastability by (a) ensuring the atom's neutrality, (b) removing the l degeneracy of the \bar{p} atomic levels, and (c) providing a Pauli-repulsion effect. The isotope effect alone could not, however, be unequivocally attributed to the formation of particular atomic structures. It was clear that a more informative and conclusive experiment would be to induce resonant transitions between internal energy levels of the exotic atom in question by laser irradiation.

In the present Letter we report results on the first laser spectroscopy experiment ever done with antiprotonic exotic atoms, and establish the large (n, l) states of neutral ${}^{-}p\text{He}^{+}$ as the source of the observed metastability. Our method, described in detail in [7], is to use a high-power dye laser pulse to stimulate resonant transitions between metastable-nonmetastable pairs of states differing by one unit in l. After Auger deexcitation of the newly populated short lived member to a $\bar{p}\text{He}^{++}$ ion, annihilation should follow within picoseconds via Stark mixing, and produce a sharp peak in the annihilation-time spectrum at the time the laser pulse is applied. Recent theoretical calculations by Ohtsuki [7,8] of \bar{p} He⁺ energy levels, their populations, and their Auger and radiative rates were used as a guide in searching for suitable transitions. In the present search we concentrated on the candidate transition $(n, l) = (39, 35) \rightarrow (38, 34)$ whose vacuum wavelength is expected to be around 598 nm (Fig. 1). This method is clearly superior to that of stimulating transitions between two metastable levels, which would change the annihilation-time spectrum only slightly.

The 200 MeV/c \bar{p} beam from the LEAR storage ring used in these experiments was extracted in an 80 min long spill at an intensity of some 10^4 /s. The beam traversed a 0.5 mm thick plastic scintillation counter (Bin Fig. 2) before entering a target-gas chamber. We used low temperature helium gas at a pressure between 0.7 and 1 bar, since we have found that these conditions provide the best environment for studying the metastability [9]. The chamber and its contents were maintained at 5-10 K by cold helium gas (evaporated from a separate liquid helium reservoir) which flowed around the chamber walls. The small emittance $(10\pi \text{ mm mrad in both di-}$ rections) and momentum bite (0.1%) of the LEAR beam meant that we could achieve a stopping volume of about $1 \times 1 \times 5$ cm³ in helium at these temperatures and pressures. Light from the B counter edges was viewed by two phototubes operated in coincidence, while light emerging



FIG. 1. Relevant energy levels versus orbital angular momentum of metastable antiprotonic helium atom $\bar{p}\text{He}^+$ calculated by Ohtsuki [7,8]. The levels indicated by bold lines have long lifetimes, while those indicated by broken lines are Auger dominated with short lifetimes. The calculated Auger rates (upper number) and radiative rates (lower number) per second are attached to each level, the number in parentheses being the power of 10. Possible laser-induced transitions for forced annihilation are indicated by solid arrows, with the present search candidate shown by a bold arrow. Broken arrows are used for transitions between metastable states, and $\Delta l = 3$ Auger (i.e., nonradiative) transitions are indicated by wavy lines.

from its surface was seen by a charged-coupled-device (CCD) image-intensified camera with its axis at 45° to the beam axis. This provided us with a continuously visible image of the beam spot which was used to maintain the alignment of the antiproton beam with the laser beam. The latter entered the target chamber through a downstream quartz window, and left it after reflection from the highly polished inner surface of the antiproton beam entrance window.

The lasers were to be ignited by all events for which no prompt annihilation had been detected. These occurred with a random time distribution and at a rate ($\sim 400/s$) close to the maximum laser repetition rate. We therefore had to make sure that the much more numerous prompt annihilations were vetoed with very high efficiency, since even a small undetected prompt fraction would generate many spurious triggers. For this purpose the helium target was surrounded by stacks of interleaved lead plates and plastic scintillators ("shower counters," labeled S in Fig. 2) which were designed to detect both charged and neutral pions from \bar{p} annihilations with high efficiency. The S counter stacks had a thickness of about 5 radiation lengths and were estimated to have an overall efficiency of about 99.7% for \bar{p} annihilation events. In the laser experiments, any event in which an S signal was received within 100 ns of a B signal was discarded as a prompt annihilation. If no such "prompt" S signal was received, we assumed that a metastable \bar{p} He⁺ atom was present in the gas, and after a suitable delay allowed the B counter pulse to ignite the laser. Our method of obtaining undistorted and background-free \bar{p} annihilation time spectra is described in detail in [3]. In the present laser runs we recorded delayed annihilations occurring in



FIG. 2. Experimental setup. 200 MeV/ $c \ \bar{p}$'s, passing through a beam counter (B) and the hole in a ring counter (A), are stopped in a target chamber containing helium, which is cooled to 5–10 K at 0.7–1 bar. Seven \bar{p} annihilation counters (S, 6 sides and 1 bottom) are located around the target. Two laser beams enter the target chamber through quartz windows from the direction opposite to the \bar{p} beam.

the 10 μ s following the \bar{p} arrival time. Control events without laser irradiation were accumulated during the laser dead time of 2.5 ms. We searched the region of the expected resonance by scanning the wavelength step by step, the typical run time for one wavelength point being about 20 min.

Our laser system consisted of two pulsed dye lasers (Lambda Physik LPD3002) pumped at 308 nm by XeCl excimer lasers (Lambda Physik LPX240i). In the wavelength range of the present experiment (594–600 nm) the dye used was Rhodamine 6G dissolved in methanol. The two lasers could be scanned in different wavelength regions simultaneously in order to search for the resonance more quickly. The laser beams were expanded to about 15 mm in diameter to cover the transverse \bar{p} stopping distribution and were merged before they entered the target chamber. Their wavelengths were measured with pulsemode wavemeters (Burleigh WA4500), the calibration of which was periodically verified against optogalvanic signals from an argon discharge cell, using an air gap etalon. The energy in each dye laser pulse was measured by *p-i-n* photodiodes, the values obtained being frequently calibrated against a calorimeter (Scientech AC2501). The excimer pulse energies were also recorded pulse by pulse using biplanar detectors and were calibrated against another calorimeter (Scientech AC50UV). Typical excimer pulse energies were in the region 80-140 mJ, and produced dye laser pulses of energy 3–5 mJ.

In Fig. 3 (left), we show a series of annihilation-time

spectra obtained at vacuum wavelengths near 597.2 nm. The delay in igniting the laser after receiving a "no prompt annihilation" B signal was in each case 1.8 μ s (the minimum we could achieve with the present setup). As the wavelength passes through the value 597.26 nm. a sharp peak appears in the spectrum, indicating forced annihilation via the mechanism described above. An enlarged view of the peak near the wavelength of maximum excitation is given in Fig. 3 (upper right). The peak profile showed an exponential decay with a time constant of 15 ± 1 ns, which reflects the lifetime of the Auger-dominated short lived state (although it also depends on the laser power and pulse profile). Even though this particular state contains only a fraction of the total metastable-state population, the laser-induced spike shows a striking peak/background ratio ($\sim 10:1$ on resonance). No other resonances were found over the ranges of vacuum wavelength scanned (from 594.152 to 594.370 and from 596.662 to 598.273 nm).

The resonance curve of Fig. 3 (lower right) displays the background subtracted number of delayed annihilations in the peak region normalized to their total number, plotted against wavelength. When the laser bandwidth of 0.007 nm (assumed Gaussian) has been deconvoluted, we find a mean vacuum wavelength of 597.259 ± 0.002 nm and a FWHM of 0.018 nm. Minor perturbations in the antiproton beam position during the scan could not be followed in the CCD image and could give rise to small systematic effects. Taking these as well as systematic



FIG. 3. Left: Observed time spectra of delayed annihilation of antiprotons with laser irradiation of various vacuum wavelengths near 597.2 nm, normalized to the total delayed component. Spikes due to forced annihilation through the resonance transitions are seen at 1.8 μ s. Upper right: Enlarged time profile of the resonance spike. A damping shape with a time constant of 15 ± 1 ns is observed. Lower right: Normalized peak count versus vacuum wavelength in the resonance region, showing a central wavelength 597.259 ± 0.002 and a FWHM 0.018 nm.

uncertainties in the wavelength calibration into account, we have assigned for the moment the conservative error given above to the resonance position. The width of the resonance line is larger than the laser bandwidth (0.007 nm), presumably because of collisional and power broadening. No measurement was made of the pressure shift.

The observed resonance wavelength can be compared with the theoretical calculations for the transition (n = $39, l = 35) \rightarrow (n = 38, l = 34)$ in \bar{p} He⁺. The calculation based on the molecular approach (Born-Oppenheimer approximation) by Shimamura [10] gives 598.01 nm, while Greenland and Thürwächter [11] predict 598.10 nm in a similar calculation. The configuration mixing calculation by Ohtsuki [8] yields 597.11 nm. Considering that none of these calculations aimed to equal the precision we achieved in our laser spectroscopy measurement, it is fair to say that the experimental value is in good agreement with them all. For comparison, the values for $(40, 35) \rightarrow (39, 34)$ and $(38, 35) \rightarrow (39, 34)$ expected from [10] are 673.7 nm and 841.1 nm, respectively. The n values 39 and 38 for the initial and final states follow immediately from this sensitive n dependence of the transition energy. On the other hand, the transition energies for different l values in $(39, l) \rightarrow (38, l-1)$ are nearly degenerate. The present assignment of the resonant transition to $(39, 35) \rightarrow (38, 34)$ is based on the theoretical estimate by Ohtsuki [7,8] of the boundary between the radiation-dominated long lived states and the Auger-dominated states, and thus is somewhat model dependent. From the observed 15 ± 1 ns decay time of the peak, the lifetime of the Auger-dominated state has been estimated to be 7 ns, which is in good agreement with theoretical values [7,8]. The integrated number of counts in the observed peak indicates that the partial population of the parent state at 1.8 μ s is at least 6% of the total metastable fraction, again consistent with expectations from [7,8].

In summary, we have carried out the first laser spectroscopy experiment on antiprotonic exotic atoms and demonstrated that the longevity against annihilation shown by some 3% of all antiprotons stopped in helium is due to the formation of \bar{p} He⁺ atoms with large n and l. The transition energy has been determined precisely, and the lifetime of the Auger-dominated state and the partial population of the parent metastable state have been deduced. All this "microscopic" information supports theoretical predictions of the characteristics of \bar{p} He⁺ atoms. We expect in the future to use our laser tools to study the formation, structure, and reactions of this new atomic species in more detail.

We are indebted to the LEAR and PS staff at CERN for their tireless dedication to providing us with our antiproton beam, to A.D. Donnachie and P. Darriulat for encouragement, to K. Ohtsuki for many valuable discussions and theoretical results, and to T. Morimoto for invaluable help in designing the experimental setup. The present work is supported by the Grants-in-Aid for Specially Promoted Research and for International Scientific Research of the Japanese Ministry of Education, Science and Culture, the Japan Society for the Promotion of Science (JSPS), and the Bundesministerium für Forschung und Technologie. E.W. and F.E.M. acknowledge the receipt of JSPS and INOUE fellowships, respectively.

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- M. Iwasaki, S. N. Nakamura, K. Shigaki, Y. Shimizu, H. Tamura, T. Ishikawa, R. S. Hayano, E. Takada, E. Widmann, H. Outa, M. Aoki, P. Kitching, and T. Yamazaki, Phys. Rev. Lett. 67, 1246 (1991).
- [2] T. Yamazaki, E. Widmann, R. S. Hayano, M. Iwasaki, S. N. Nakamura, K. Shigaki, F. J. Hartmann, H. Daniel, T. von Egidy, P. Hofmann, Y.-S. Kim, and J. Eades, Nature (London) 361, 238 (1993).
- [3] S. N. Nakamura, R. S. Hayano, M. Iwasaki, K. Shigaki, E. Widmann, T. Yamazaki, H. Daniel, T. von Egidy, F. J. Hartmann, P. Hofmann, Y.-S. Kim, and J. Eades (to be published).
- [4] T. Yamazaki and K. Ohtsuki, Phys. Rev. A 45, 7782 (1992).
- [5] G. T. Condo, Phys. Lett. 9, 65 (1964).
- [6] J. E. Russell, Phys. Rev. Lett. 23, 63 (1969); Phys. Rev. 188, 187 (1969); Phys. Rev. A 1, 721 (1970); 1, 735 (1970); 1, 742 (1970).
- [7] N. Morita, K. Ohtsuki, and T. Yamazaki, Nucl. Instrum. Methods Phys. Res., Sect. A 330, 439 (1993).
- [8] K. Ohtsuki (private communication).
- [9] E. Widmann, H. Daniel, J. Eades, T. von Egidy, F. J. Hartmann, R. S. Hayano, W. Higemoto, J. Hoffmann, T. M. Ito, Y. Ito, M. Iwasaki, A. Kawachi, N. Morita, S. N. Nakamura, N. Nishida, W. Schmid, I. Sugai, H. Tamura, and T. Yamazaki, Nucl Phys. A558, 679c (1993).
- [10] I. Shimamura, Phys. Rev. A 46, 3776 (1992); (private communication).
- [11] P. T. Greenland and R. Thürwächter, Hyperfine Interact. 76, 355 (1993); P. T. Greenland (private communication).