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**Observation of Double Resonant Laser Induced Transitions  
in the  $v = n - l - 1 = 2$  Metastable Cascade of Antiprotonic Helium-4 Atoms**

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**Abstract**

A new laser-induced resonant transition in the  $v = n - l - 1 = 2$  metastable cascade of antiprotonic  $^4\text{He}$  atoms has been found by using a double resonance technique. This was done by setting the first laser to the already known 470.724 nm resonance ( $(n, l) = (37, 34) \rightarrow (36, 33)$ ), while the  $(38, 35) \rightarrow (37, 34)$  transition was searched for with the second laser. The resonant transition was found at wavelength of  $529.622 \pm 0.003$  nm, showing excellent agreement with a recent prediction of Korobov.

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Stopped antiprotons live in matter usually only about  $10^{-12}$  s. An exceptional case is He where about 3% of the incoming stopped antiprotons live as long as about  $\sim 3 \mu\text{s}$  [1, 2, 3, 4]. The longevity is due to antiprotons caught in high Bohr orbits with high angular momentum where only slow radiative transitions are allowed. We report here the observation of a laser-induced resonant transition between two metastable states of antiprotonic helium atoms ( $\bar{p}^4\text{He}^+$ ) [1, 2, 3, 4]. These were formed by stopping antiprotons from the CERN Low Energy Antiproton Ring (LEAR) in a low temperature helium gas target ( $4 \sim 6$  K,  $\sim 0.5$  bar). The resonance was detected by a novel technique using two laser beams fired simultaneously. We set one of these at the 470.724 nm wavelength of the known  $(n, l) = (37, 34) \rightarrow (36, 33)$  transition with vibrational quantum number  $v = n - l - 1 = 2$  and searched with the other near the theoretically expected wavelength of the  $(38, 35) \rightarrow (37, 34)$  metastable-metastable transition immediately above it in the  $v = 2$  sequence.

The lower transition, being the last one in the  $v = 2$  cascade (see Fig.1), proceeds from a metastable to a nonmetastable state. The  $\sim 1 \mu\text{s}$  lifetime of the upper level thus ensured that it retained a large fraction of its initial population until the laser pulse arrived  $\sim 1.7 \mu\text{s}$  after the atom was formed. On the other hand the lower level was constantly being emptied by an Auger transition to  $\bar{p}^4\text{He}^{++}$  with lifetime  $\tau \leq 10$  ns. The laser light could therefore effect a substantial net transfer of  $\bar{p}$  to the  $(36, 33)$  state. As in our original observation of this transition [5], the extreme instability of the latter against annihilation via collisional Stark mixing [6, 7] then produced a sharp peak, synchronous with the laser pulse, in the  $\bar{p}$  annihilation rate. This peak served as a sensitive indicator of the resonance condition.

Clearly this single-laser technique will not work if the upper and lower levels are both metastable, since at any given instant their populations can be expected to be almost equal. In the present case however, as the second laser was tuned through the upper transition frequency, antiprotons in the  $(38, 35)$  state were transferred through the  $(37, 34)$  level, already being drained by the first laser, to the  $(36, 33)$  level. The upper resonant condition then revealed itself as an increase in the height of the abovementioned 470.724 nm annihilation peak.

Besides the 470.724 nm transition, we had previously observed another metastable-nonmetastable resonance at 597.259 nm. We were able to assign this to the  $v = 3$   $(39, 35) \rightarrow (38, 34)$  sequence ([8, 9]) with the help of theoretical calculations of wavelengths and Auger lifetimes. These imply that only five metastable-nonmetastable transitions (those with  $v = 0, 1, 2, 3, 4$ ) exist, and that only three of these ( $v = 2, 3, 4$ ) are within easy reach of our visible-light dye laser system. The work here described, carried out with an experimental arrangement identical to that described in [8] except for the addition of the second laser system, is evidently an important step in extending our laser spectroscopy technique beyond these few transitions.

In reporting [5] the observation of the 470.724 nm transition we had, by varying the ignition times of two lasers at the same wavelength, already shown that the upper  $(37, 34)$  level population was being replenished from one or more still higher-lying states. This was deduced from the fact that when the earlier-firing laser was used to empty the  $(37, 34)$  state, the later-firing one produced a second annihilation peak, indicating that the  $(37, 34)$  population had partially recovered in the meantime. Our usual assumption is that the radiative transitions between metastable states occur preferentially along the constant- $v$  cascade chain. In this case, the  $(37, 34)$  state must have been replenished from the  $(38, 35)$ , possibly via still higher  $v = 2$  levels. A model based on this supposed chain

decay sequence [5] gave an initial population of the (38,35) state  $\sim 5\%$  of the total delayed fraction. We therefore expected that the (38,35) state would still be populated at the time of arrival of the laser pulse, and that this would produce a net enhancement of the lower resonance peak.

Until very recently, theoretically calculated wavelengths for the  $(n,l) \rightarrow (n-1,l-1)$  transition were all subject to uncertainties of order 1nm[10, 11]. Korobov[12] has now used a molecular-expansion variational calculation that gives transition energies for the two already observed transitions within 50 ppm of their measured values (*i.e.*  $< 0.03$ nm, only about 5 times the laser bandwidth). The high accuracy of Korobov's prediction already led us to a successful observation of laser-resonant transition in  $^3\text{He}$ [13]. We therefore fixed the wavelength of one laser to the 470 nm resonance (blue), and scanned the second one around Korobov's predicted value of 529.60 nm (green). The pulses that ignited the two lasers were synchronized to within  $\sim 5$  ns, although the unavoidable time jitter of the excimer lasers that pump the tunable dye lasers caused the actual light pulse arrival time to vary with a FWHM of  $\sim 15$  ns. However, this jitter was well within the width of the light pulse ( $\sim 30$ – $40$  ns), so that the two laser pulses almost always overlapped in time.

The expected enhancement of the lower (38,35)  $\rightarrow$  (37,34) transition was soon found at an upper wavelength of 529.622 nm. This can be seen in Fig. 2, which compares the annihilation time spectra with the second laser off- and on-resonance (upper and lower panels). The dependence of the annihilation peak counts (normalized to the total number of delayed events) on the second laser wavelength is shown in Fig. 3. The upper and lower panels show the scan result for helium at 1.0 bar and 1.3 bar respectively, the temperature being in both cases 5.3 K. The central wavelength of the resonant transition was found to be  $529.622 \pm 0.003$  nm, where the errors cover both statistical errors and calibration errors for the wavelength meter.

In addition to the discovery of a new resonant transition, a closer look at Fig. 3 reveals the following interesting facts.

1) The central wavelengths are slightly different for the two scans taken at different target pressures. It is 529.621 nm at 1 bar (upper panel) and 529.623 nm at 1.3 bar (lower panel). This is a small but statistically significant difference, although we cannot draw a definitive conclusion based on the measurements taken at only two pressure points. We later studied the pressure dependence of the resonance wavelengths both for the 470 nm and the 597 nm resonances, and found that the wavelengths became longer as the pressure was increased, and that the pressure shift was state dependent. This will be discussed in detail in our forthcoming publication[14].

2) The resonance peak heights also appear to have a pressure dependence. This is true for the 529 nm resonance, and also for the 470 nm resonance. The latter can be seen by comparing the off-resonance peak to total ratio, which is about 0.6 % at 1 bar and about 0.3% at 1.3 bar. These values are significantly smaller than the peak to total ratio of about 4% at 0.6 bar, reported in our previous publication[5]. We found this is due to the strong pressure dependence of the lifetime of the (37,34) level ( $\tau_{37}$ ). At low ( $< 0.2$  bar) pressure,  $\tau_{37}$  approached the calculated radiative-decay value of  $1.37 \mu\text{s}$ , while it became as short as  $\sim 0.3 \mu\text{s}$  at 1 bar. A detailed account of the pressure dependence of the level lifetimes will be given in a separate paper[15].

We noted that the (37,34) level is already being constantly drained at high pressure, so that the 529 nm resonance must be observable without the aid of the first laser. This we verified by turning off the first laser, and setting the second laser on resonance. A typical result taken at a pressure of 1.2 bar is shown in Fig. 4. The laser-induced annihilation

peak is clearly visible, but unlike other cases the peak has a long tail, whose slope is determined by the lifetime of (37,34) level at this pressure.

In conclusion, we discovered a new laser induced resonant transition at 529.623 nm by introducing the double resonance method, and ascribed it to the  $(n,l) = (38,35) \rightarrow (37,34)$  transition in the  $v = n - l - 1 = 2$  metastable cascade.

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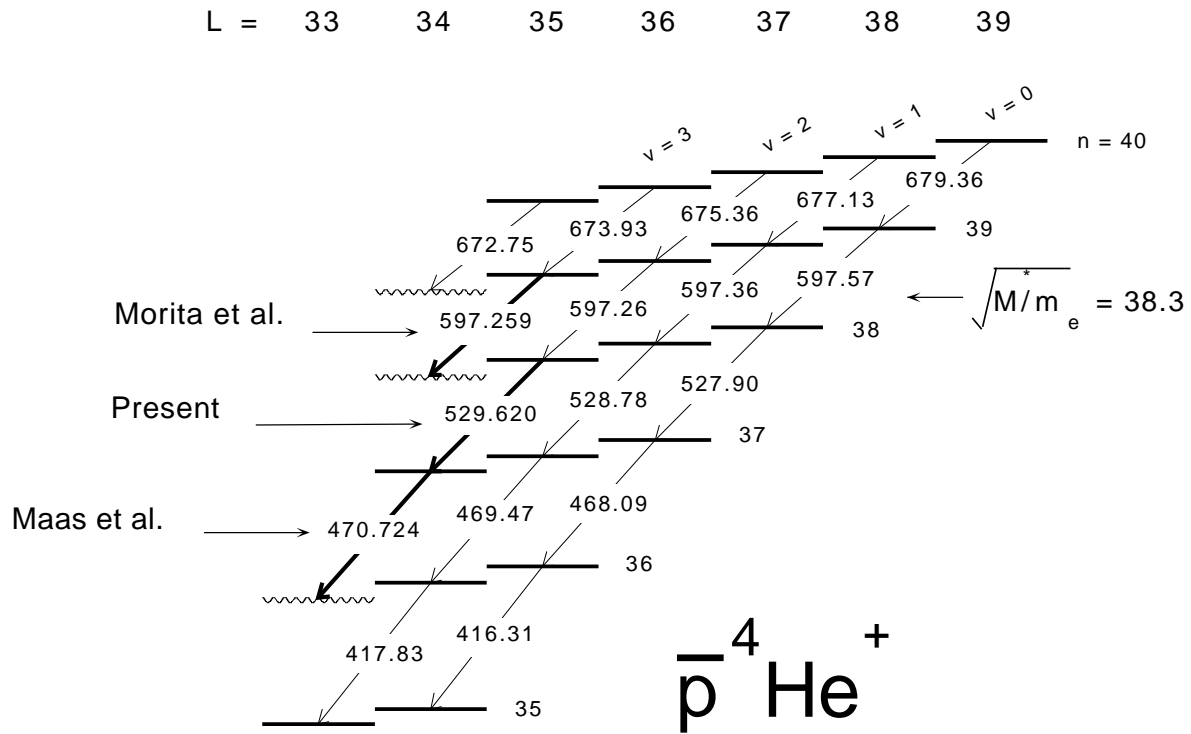


Figure 1: An energy level diagram of the  $\bar{p}^4\text{He}^+$  atom. The transitions found in our previous work and the one found in the present double resonance method are shown by thick arrows, with the observed wavelengths. All other transition wavelengths are taken from Korobov [12]. Metastable levels are indicated by solid lines and Auger-dominated short-lived states are drawn by wavy lines.

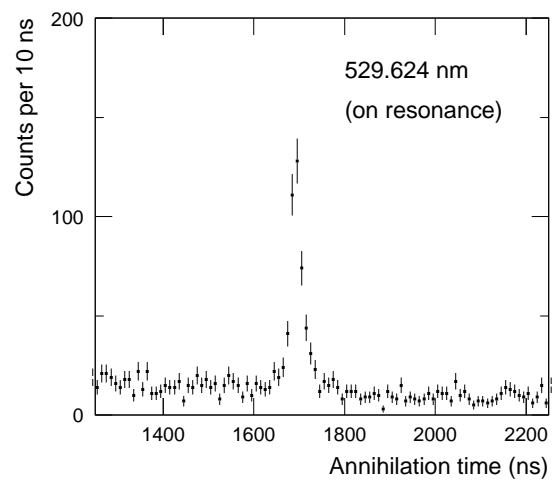
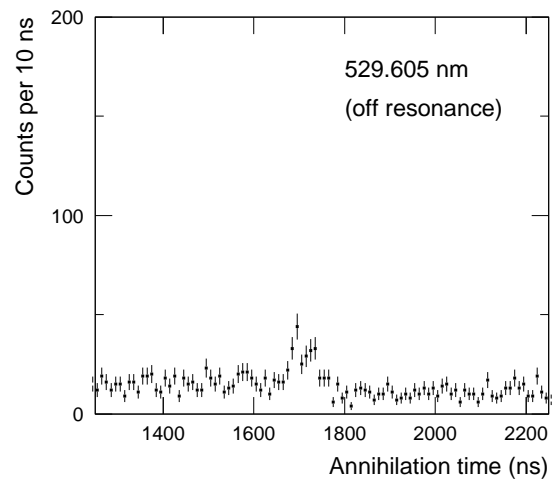


Figure 2: Delayed annihilation time spectra taken with the singly resonant condition (upper panel: first laser at 470.724 nm, second laser at 529.605 nm) and the doubly resonant condition (lower panel: first laser at 470.724 nm, second laser at 529.622 nm).

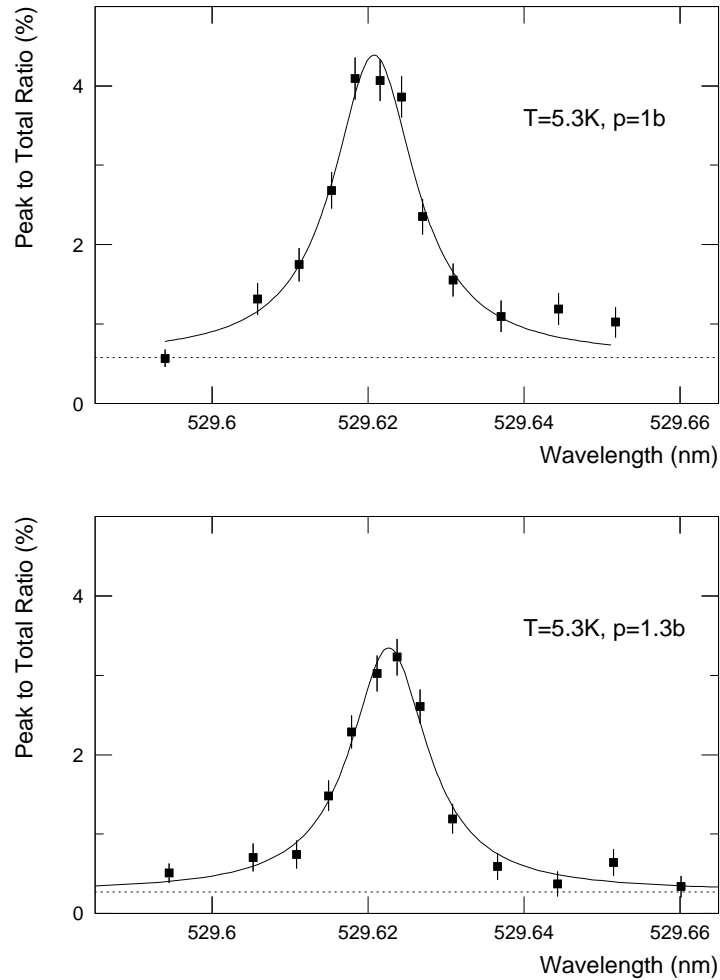


Figure 3: The dependence of the double resonance intensity on the wavelength of the second laser. The upper panel shows the resonance scan result taken with a helium gas target pressure of 1 bar and temperature of 5.3 K, while the lower panel is for a pressure of 1.3 bar and temperature of 5.3 K.

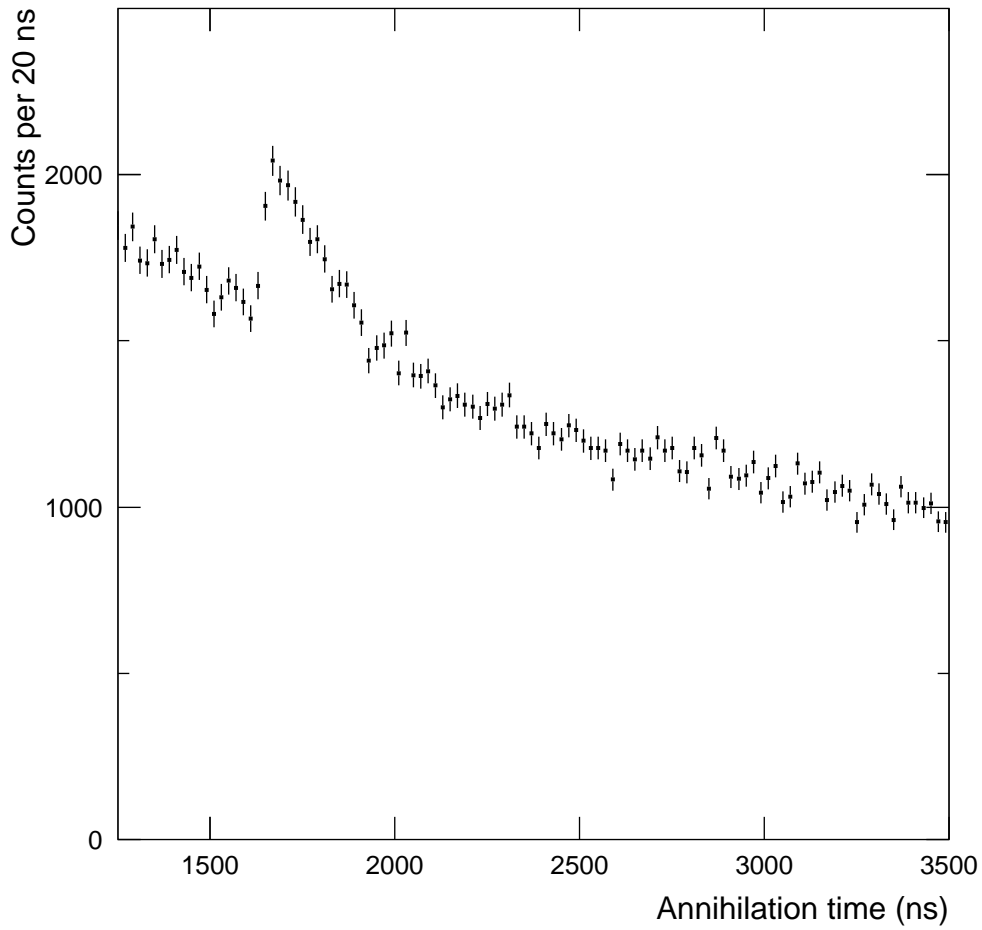


Figure 4: A delayed annihilation time spectrum taken without the first laser and with the second laser set at the 529 nm resonance. The helium target was at a temperature of 5.3 K and pressure of 1.2 bar. At this pressure, the lifetime of (37,34) level was about  $\sim 0.2\mu\text{s}$ , so that the 529 nm resonance was visible without the aid of the first laser.