Observation of double-resonant laser-induced transitions in the \(v = n - l - 1 = 2\) metastable cascade of antiprotonic \(^4\text{He}\) atoms

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Another 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 \((n, l) = (37, 34) \to (36, 33)\), while the \((38, 35) \to (37, 34)\) transition was searched for with the second laser. The resonant transition was found at a wavelength of 529.622 ± 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 \(\sim 3 \mu s\) [1–4]. 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 (\(\bar{p}\, ^{4}\text{He}^+\)) [1–4]. These were formed by stopping antiprotons from the CERN Low Energy Antiproton Ring (LEAR) in a low-temperature helium gas target (4–6 K, \(\sim 0.5\) bar). The resonance was detected by a technique using two laser beams fired simultaneously. We set one of these at the 470.724-nm wavelength of the known \((n, l) = (37, 34) \to (36, 33)\), while the \((38, 35) \to (37, 34)\) transition was searched for with the second laser. The resonant transition was found at a wavelength of 529.622 ± 0.003 nm, showing excellent agreement with a recent prediction of Korobov. [S1050-2947(97)0601-9]
stable state. The $\sim 1\,\mu s$ lifetime of the upper level thus ensured that it retained a large fraction of its initial population until the laser pulse arrived $\sim 1.7\,\mu s$ after the atom was formed. However, the lower level was constantly being emptied by an Auger transition to $\bar{p}\,^4\text{He}\,^+\!$ with lifetime $\tau \leq 10\,\text{ns}$. 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 above-mentioned 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 $\nu = 3\rightarrow 39$ sequence [8,9] with the help of theoretical calculations of wavelengths and Auger lifetimes. These imply that only five metastable-nonmetastable transitions (those with $\nu = 0,1,2,3,4$) exist, and that only three of these ($\nu = 2,3,4$) are within easy reach of our visible-light dye laser system. The work described here, 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-$\nu$ cascade chain. In this case, the (37,34) state must have been replenished from the (38,35), possibly via still higher $\nu = 2$ 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 1 nm [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 five 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); as in our previous measurements, the $\bar{p}$ annihilation time spectra were accumulated by detecting the annihilation pions. The pulses

![Image](image-url)

**FIG. 1.** 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 as wavy lines.

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**FIG. 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 double-resonant condition (lower panel: first laser at 470.724 nm, second laser at 529.622 nm).
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 full width at half maximum 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 ± 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.

(i) The central wavelengths are slightly different for the two scans taken at different target pressures. They are 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- 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 presentation [14].

(ii) 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 indicated by the dotted lines in Fig. 3, 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 that 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 $\mu$s, while it became as short as $\sim 0.3$ $\mu$s at 1 bar. A detailed account of the pressure dependence of the level lifetimes will be given elsewhere [15].

We note 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 the $(37,34)$ level ($\tau_{37}$). The peak to total ratio at 0.2 bar pressure, $\tau_{37}$ approached the calculated radiative-decay value of 1.37 $\mu$s, while it became as short as $\sim 0.3$ $\mu$s at 1 bar. A detailed account of the pressure dependence of the level lifetimes will be given elsewhere [15].

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