

Laser spectroscopy of metastable states in the $\nu=2$ cascade of antiprotonic ${}^3\text{He}$

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The pressure dependence of the metastable-state lifetimes in antiprotonic ${}^3\text{He}$ atoms $\bar{p}\text{-}{}^3\text{He}^+$ was studied using the recently observed laser resonance transition $(n, \ell) = (36, 33) \rightarrow (35, 32)$ in the $\nu = n - \ell - 1 = 2$ cascade. To this end antiprotons from the Low Energy Antiproton Ring (LEAR) at CERN were extracted in approximately 200-ns-long bunches with about 10^8 \bar{p} per bunch and stopped in a ${}^3\text{He}$ gas target. Time spectra of delayed annihilation products were taken with the help of a Čerenkov counter with a gated photomultiplier. Under our experimental conditions, $p = 136 - 690$ mbars and $T = 5.8$ K, the lowest metastable level (36,33) was found to be much shorter lived than the corresponding state (37,34) in the $\nu = 2$ cascade of $\bar{p}\text{-}{}^4\text{He}^+$. It was strongly quenched at pressures above 500 mbars, whereas the higher states in the cascade remained nearly unaffected. This enabled us to investigate the $(37,34) \rightarrow (36,33)$ transition by resonant laser deexcitation. Its wavelength was found to be $\lambda = 524.155 \pm 0.004$ nm, only 6 ppm lower than the theoretical value given by Korobov and Bakalov [Phys. Rev. Lett. **79**, 3379 1997]) after including relativistic corrections. [S1050-2947(98)06611-6]

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I. INTRODUCTION

During the past five years metastable antiprotonic helium atoms have developed into an attractive laboratory for the exotic-atom formation and for the three-body interaction in the helium nucleus He^{2+} plus antiproton \bar{p} plus electron $e^- \equiv \bar{p}\text{-}{}^3\text{He}^+$ system [1–9]. In particular, the observation of laser-induced transitions between metastable and short-lived states [5–9] has permitted the level scheme of metastable antiprotonic helium atoms to be studied with unprecedented accuracy. After a wealth of information had been collected for $\bar{p}\text{-}{}^4\text{He}^+$, some laser-stimulated transitions were also found in $\bar{p}\text{-}{}^3\text{He}^+$. The fact that the $\bar{p}\text{-}{}^3\text{He}^+$ data showed some unexpected peculiarities when compared with those

from $\bar{p}\text{-}{}^4\text{He}^+$ [4,9] suggested that it would be very worthwhile to study the $\bar{p}\text{-}{}^3\text{He}^+$ atom in more detail.

II. THE METASTABLE \bar{p} -HELIUM ATOM

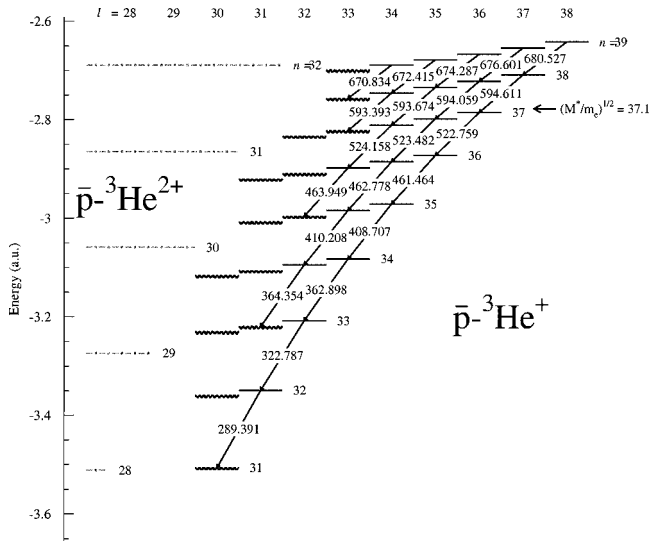
When antiprotons shot into helium are slowed down to kinetic energies lower than about the ionization potential of this element, they are captured by the Coulomb force to form $\bar{p}\text{-He}^+$ atoms [10,11], thereby ejecting one of the two electrons of the He atom. As has been shown by a number of experimental observations [5–9], states populated in the capture process have principal quantum numbers $n \approx \sqrt{M^*/m_e}$ (with M^* and m_e the reduced antiproton mass and the electron mass, respectively) that guarantee maximum overlap between the wave functions of the captured antiproton and of the ejected electron. This means, e.g., $n \approx 38$ for $\bar{p}\text{-}{}^4\text{He}^+$ and $n \approx 37$ for $\bar{p}\text{-}{}^3\text{He}^+$. Antiprotons captured into large-angular-momentum (ℓ) states form *metastable* $\bar{p}\text{-He}^+$ atoms with lifetimes of the order of microseconds [12] because the transitions to levels from which immediate annihilation is possible are suppressed.

Three facts contribute to this effect.

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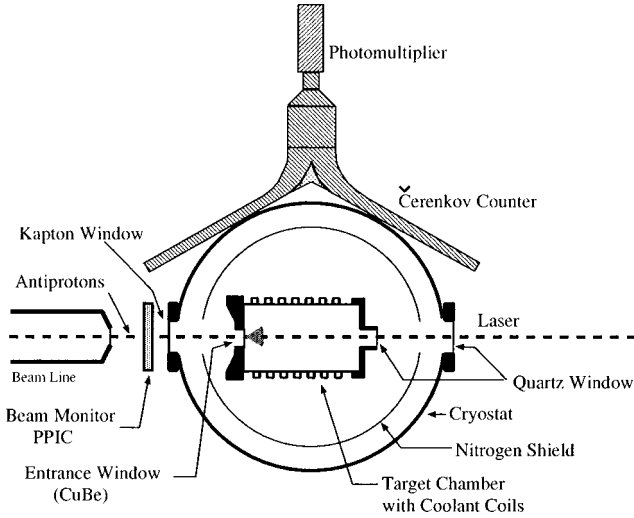


FIG. 2. Experimental arrangement for the fast-extraction studies. PPIC, the parallel plate ionization chamber.

root of the reduced mass M^* , $\Delta E \approx M^* c^2 (\alpha Z)^2 / n^3 \propto (M^*)^{-1/2}$, and (ii) the rate λ for radiative transitions depends on ΔE to the third power, $\lambda \propto (\Delta E)^3 \propto (M^*)^{-3/2}$. The total cascade time $\tau \propto [\Delta E \lambda]^{-1}$ hence is proportional to $(M^*)^2$. One expects a \bar{p} -He⁺ lifetime that is larger in \bar{p} -⁴He⁺ by a factor

$$\left[\frac{M^*(\bar{p}-^4\text{He}^{2+})}{M^*(\bar{p}-^3\text{He}^{2+})} \right]^2 = 1.14,$$

a value very close to the one found from experiment, 1.144 ± 0.009 [4]. A closer look reveals further differences between \bar{p} -³He⁺ and \bar{p} -⁴He⁺: (i) The fraction of trapped antiprotons is lower by 22.2(4)% in \bar{p} -³He⁺ than in \bar{p} -⁴He⁺ at 5.8 K and pressures between 400 and 580 mbar [4] and (ii) a short-lived component in \bar{p} -³He⁺ (lifetime $\tau = 154 \pm 7$ ns) exists at 5.8 K and 400 mbar, which is not visible in \bar{p} -⁴He⁺ [4]. The reason for all this was not clear and it seemed to be necessary to study the \bar{p} -³He⁺ cascade in more detail. Very recently it has been revealed that the lifetime of the last metastable state in the $\nu=2$ cascade of \bar{p} -⁴He⁺, (37,34), shows a much stronger density dependence than the levels with larger n [20]. It is interesting to investigate this density dependence for the corresponding state in \bar{p} -³He⁺, (36,33), which may be depopulated by laser irradiation with a wavelength of 463.947 ± 0.002 nm [9]. If the lifetime of this state can be shortened by increasing the density, the upper transition (37,34) \rightarrow (36,33) may also be investigated by laser resonance.

III. EXPERIMENTAL METHOD

Experimental setup

The experimental setup is shown in Fig. 2. Antiprotons of 200 MeV/ c momentum were extracted from the Low Energy Antiproton Ring (LEAR) at CERN in bunches of about 200 ns length containing around 10^8 \bar{p} per bunch (*fast extraction*). They were stopped in a target cooled down to

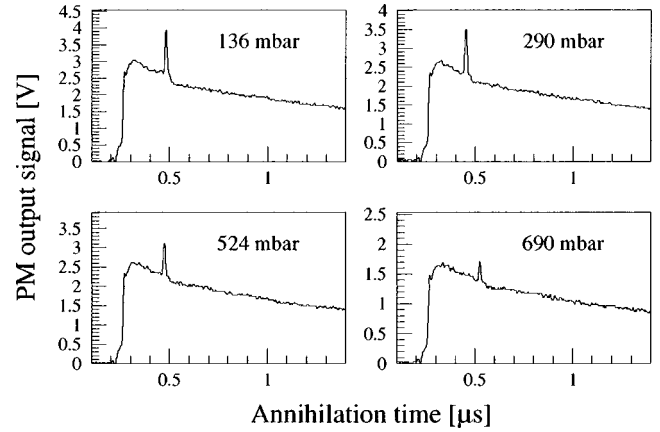


FIG. 3. Analog delayed annihilation time spectra for \bar{p} -³He⁺ at different pressures. The resonant deexcitation of the last metastable level manifests itself in the annihilation spikes seen. Their height decreases with increasing pressure due to the decreasing lifetime of the lowest level.

5.8 K and filled with ³He gas (purity 99.998%) at pressures between 136 and 690 mbar. The (charged) pions from \bar{p} annihilation were detected collectively (*analog method* [21]) with the help of a lucite Čerenkov counter of ≈ 2400 cm² area and 2 cm thickness, which was positioned close to the helium target and covered a solid angle for the annihilation pions of about 36% of 4π . The emitted Čerenkov light was fed into a photomultiplier via a light guide. One serious problem, however, existed: As 97% of the annihilations are prompt, the photomultiplier (PM) would be totally overloaded during the prompt annihilation period if one wanted to see the delayed annihilations with sufficient intensity. To remove this overloading, a gatable PM was developed by Hamamatsu Photonics Ltd. that could be switched on and off, respectively, within a few tens of nanoseconds. It had an on/off ratio of sensitivity of better than 1000:1 and very good integral linearity over the whole output-voltage operating range ($U \leq 20$ V into 50 Ω). The analog signal from this PM was recorded with a digital oscilloscope (Hewlett-Packard 54542A, with a bandwidth of 500 MHz, four channels at 2 G samples/s for each channel, and a resolution of eight bits). Finally the information was transferred from the digital oscilloscope to a computer and stored on disk and tape.

Four examples for an *analog delayed annihilation time spectrum* (ADATS) of this kind are shown in Fig. 3. Transitions from metastable to Auger-depopulated states were resonantly induced by a light pulse from an excimer-laser driven dye laser (≈ 4 mJ/pulse) shot into the target through a quartz window downstream along the beam. A detailed description of the laser system may be found in Ref. [22].

The *fast extraction* method has some advantages compared to the *slow extraction* method used in previous experiments.

(i) The laser is used much more efficiently as there are about 3×10^6 metastable \bar{p} -He⁺ atoms in the target when the laser fires and not just one as in slow extraction [22]; consequently, only one laser shot is necessary to accumulate a whole ADATS.

(ii) Antiproton bunch and laser timing may be synchronized in such a way that the laser may be fired at any time

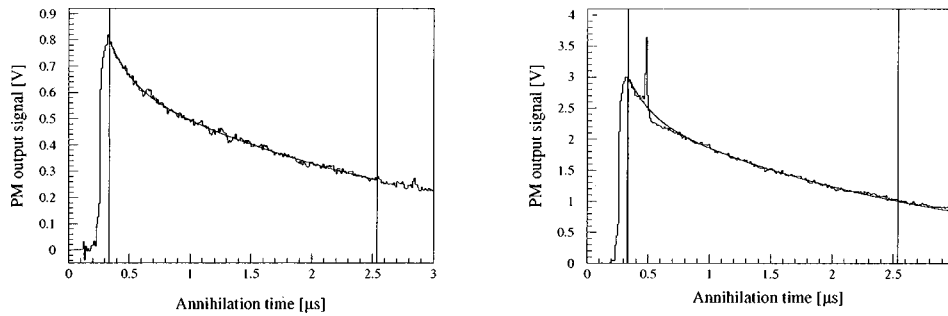


FIG. 4. (a) Fit to the *off-resonance* fast-extraction spectra. The double-exponential perfectly represents the data. (b) Typical depletion spectrum with the background from (a) overlaid. The vertical lines denote the borders of the fit region.

with respect to the antiproton bunch. With the conventional method [22] the laser could be triggered only after it became clear that *no* prompt annihilation had occurred. With a resulting minimum delay between prompt peak and laser ignition time of about $1.3 \mu\text{s}$, only rather late times could be investigated.

IV. EXPERIMENTAL RESULTS

In the ADATS of Fig. 3 the laser-induced annihilation peak at about 500 ns is displayed for various gas pressures at a constant temperature $T = 5.8 \text{ K}$. Two methods are available to derive level lifetimes and populations.

One is variation of the delay time t of the laser shot (t scan). The metastable level can be emptied at variable time t by a sufficiently powerful laser pulse. From the intensity of the annihilation peak the population of the state may be derived as a function of the laser timing and an overall lifetime $T_\nu = 1/\lambda_\nu$ of the antiprotons in the cascade $\nu = \text{const}$ may be extracted. It should be emphasized that T_ν is *not* the lifetime of the last metastable state.

The other is the depletion-recovery method. The metastable level is emptied at time t by laser irradiation. It is refilled from higher states afterwards. The annihilation-time spectrum after the laser shot approaches the time spectrum without laser shot with the lifetime of the emptied metastable state [20]. The reliability of this method in fast extraction was tested by comparing the values for the lifetime of the (37,34) state in $\bar{p}\text{-}^4\text{He}^+$ derived by the depletion-recovery method in fast and slow extraction [23]. Very good agreement was found between the fast-extraction value $\tau_{(37,34)} = 137 \pm 21 \text{ ns}$ at a density of $4.88 \pm 0.13 \text{ mol/l}$ and the slow-extraction value $\tau_{(37,34)} = 128 \pm 25 \text{ ns}$ at a density of $4.56 \pm 0.12 \text{ mol/l}$.

A. Data reduction

In order to extract level populations and lifetimes in $\bar{p}\text{-}^3\text{He}^+$ from the experimental data, peaks and decay parameters of the depletion have to be evaluated. In this context it is especially important to assign uncertainties to the measured voltages in the sampled analog spectra. Whereas in the normal delayed annihilation time spectrum (DATS) the error in intensity in each channel is given by the square root of the number of annihilations seen in the corresponding time bin Δt , in the ADATS the number of charged particles hitting the Cerenkov radiator and the number of photoelectrons

emitted from the photocathode of the PM for each charged particle determine the error for each time bin [24]. Furthermore, digitizing errors in the oscilloscope exist, which, however, may be minimized by carefully adjusting the oscilloscope sensitivity.

Unfortunately, it is rather difficult to get a reliable value for the number N_{ph} of photons reaching the photocathode, as the light collection in Cerenkov counter and light guide is hard to evaluate; also the attempt to determine N_{ph} from the anode-signal peak height, the gain of the PM, and the quantum efficiency of its photocathode suffers from uncertainties. Therefore, a statistical test was used to derive the error of the voltage measured in each bin from the voltage fluctuations. To this end a small region of the ADATS was fitted, which (i) could be perfectly described by an exponential and where (ii) digitizing errors played no role. With bin errors equal to \sqrt{V} the values of $\sqrt{\chi_\nu^2}$, the square root of the reduced χ^2 , was evaluated. $\sqrt{\chi_\nu^2}$ is just the factor each bin error has to be multiplied by to attain the correct voltage-height error. Digitizing errors were added afterwards in quadrature.

Another problem in the evaluation of the depletion-recovery spectra concerned the fit of the ADATS without laser shot. Fortunately, only the area of the laser peak (in the case of the t -scan method) or the slope in the trailing edge of the negative depletion peak (in the case of the depletion-recovery method) had to be evaluated. Nevertheless, a reliable fitting procedure was mandatory.

The off-resonance ADATS in the analog method stems from two sources: (i) the pions from delayed \bar{p} annihilation, which fade away with the overall lifetime of the metastable states and lead to a short-lived component with a lifetime τ of several hundred nanoseconds and a longer-lived one with $\tau \approx 3 \mu\text{s}$ in the DATS [4], and (ii) the positrons from the decay of μ^+ (lifetime $2.2 \mu\text{s}$) generated by the decay of prompt-annihilation π^+ stopped in the experimental environment. By fitting the ADATS with very late laser pulses by the sum of two exponentials,

$$I(t) = A \exp(-\lambda_{\text{short}}t) + B \exp(-\lambda_{\text{long}}t),$$

reliable off-resonance time spectra could be derived. A typical fit result is shown in Fig. 4.

B. Density dependence of the t -scan decay rate

The time development of the (36,33)-level population was determined with the t -scan method described above.

TABLE I. Decay rates $\lambda_{\nu=2}$ ($n > 36$) and $\lambda_{(36,33)}$ as functions of the ${}^3\text{He}$ density.

Density (mol/l)	Density (10^{20} cm^{-3})	$\lambda_{\nu=2}$ ($n > 36$) (μs^{-1})	$\lambda_{(36,33)}$ (μs^{-1})
0.29 ± 0.05	1.8 ± 0.3	0.80 ± 0.12	8.6 ± 1.1
0.62 ± 0.06	3.7 ± 0.3	0.93 ± 0.16	9.2 ± 0.9
1.13 ± 0.06	6.8 ± 0.4	1.16 ± 0.24	10.8 ± 1.5
1.52 ± 0.06	9.1 ± 0.4	1.05 ± 0.38	16.3 ± 2.4

Nine shots from one LEAR spill were used for each of the four target pressures investigated.

The results for $\lambda_{\nu=2}$ ($n > 36$) are shown in Table I, third column. Figure 5(a) depicts them as a function of density (full squares). Also shown in this figure (as a full line) is the result for $\lambda_{\nu=2}$ as derived from the fit of a straight line to the decay-rate data [cf. Eq. (1)].

C. Density dependence of the (36,33) lifetime

The decay rate $\lambda_{(36,33)}$ of the (36,33) level was directly determined by the depletion-recovery method. The results are given in Table I, fourth column. This rate is shown in Fig. 5(b) (full circles). The result of a linear fit to these λ values [cf. Eq. (1)] is depicted as a dashed line in the figure. Astonishing enough, the decay rate in the limit of zero density,

$$\lambda_{(36,33)}(\rho_{\text{atom}}=0) = 6.7 \pm 1.3 \mu\text{s}^{-1},$$

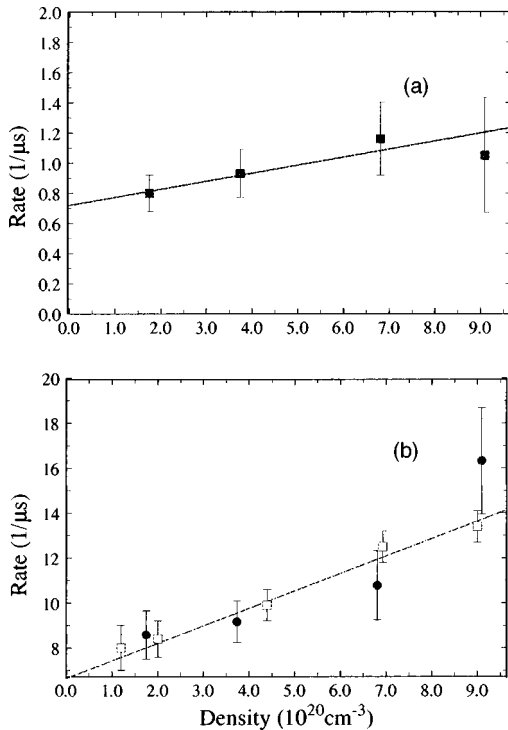


FIG. 5. Rates in the $\nu=2$ cascades of $\bar{p}^{-3}\text{He}^+$ and $\bar{p}^{-4}\text{He}^+$. (a) Data for $\lambda_{\nu=2}$ in $\bar{p}^{-3}\text{He}^+$ (full squares). The full line denotes the linear fit to the data. (b) Data for $\lambda_{(36,33)}$. Full circles, experimental $\bar{p}^{-3}\text{He}^+$ data; dashed line, fit to the experimental data; open squares, experimental data for $\lambda_{(37,34)}$ in $\bar{p}^{-4}\text{He}^+$, multiplied by a factor of 10.

TABLE II. Comparison of quenching cross sections in $\bar{p}^{-3}\text{He}^+$ and $\bar{p}^{-4}\text{He}^+$.

Level	$\sigma_{\text{Stark}} (10^{-19} \text{ cm}^2)$
$\bar{p}^{-3}\text{He}^+ (36,33)$	$3.0_{-1.0}^{+0.9}$
$\bar{p}^{-4}\text{He}^+ (37,34)$	0.52 ± 0.03

is about a factor of 4 larger than the sum of the calculated radiation and Auger rates, $\lambda = 1.7 \mu\text{s}^{-1}$ [25,26]. Assuming a quadratic density dependence, as it might be expected when three-body collisions play a role, makes the agreement even worse.

From the same fit we obtained

$$[\sigma_{\text{Stark}}]_{(36,33)} \langle \nu \rangle = 8.0_{-2.7}^{+2.3} \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}.$$

Since in the time range of the fit the $\bar{p}^{-3}\text{He}^+$ atoms are totally thermalized, $\langle \nu \rangle = 2.7 \times 10^4 \text{ cm}^3 \text{ s}^{-1}$, a cross section

$$\sigma_{\text{Stark}} = 3.0_{-1.0}^{+0.9} \times 10^{-19} \text{ cm}^2$$

follows. This cross section is much smaller than the geometrical cross section. Apparently only a small fraction of the collisions with He atoms lead to Stark mixing with subsequent fast Auger deexcitation and annihilation.

A comparison of the (36,33) decay rate with that of the (37,34) level in $\bar{p}^{-4}\text{He}^+$ [20], shown, multiplied by ten, as open squares in Fig. 5(b), reveals that the intrinsic decay rate of the last metastable state is about a factor of 10 smaller in $\bar{p}^{-4}\text{He}^+$ than in $\bar{p}^{-3}\text{He}^+$ and quenching is much less efficient in the case of $\bar{p}^{-4}\text{He}^+$. The reason for this phenomenon is not yet clear. Table II gives a comparison of the corresponding quenching cross sections.

The large difference at low density between the $\nu=2$ cascade lifetime $T_{\nu=2} = 1/\lambda_{\nu=2}$ and $\tau_{(36,33)} = 1/\lambda_{(36,33)}$ indicates that there are other metastable states above the (36,33) level in this cascade. However, not all of them are quenched as efficiently as the lowest one: $\lambda_{\nu=2}$ increases with density only to $\lambda_{\nu=2} \approx 1 \mu\text{s}^{-1}$ at 1.5 mol/l [cf. Table I]. Evidently, the metastable states above the lowest one retain their longevity. This opened up the very interesting possibility to directly observe transitions $(37,34) \rightarrow (36,33)$ resonantly induced by laser irradiation, as in the case of $\bar{p}^{-4}\text{He}^+$ [20]: The still long-lived upper state (37,34) is emptied by laser irradiation to the lower state, which has been made short-lived by density quenching, and a \bar{p} annihilation peak may be observed. The shape of this annihilation peak gives direct information on the (36,33)-level lifetime.

D. Resonance scan on the transition (37,34) \rightarrow (36,33)

For the resonance scan, performed at 690 mbar pressure, the peak intensity of the resonance spike was determined as a function of wavelength, with the background subtraction similar to that for the depletion analysis. The scan was considerably facilitated by the apparently reliable predictions for the transition energies in $\bar{p}^{-\text{He}}^+$ [27,28]. Figure 6 shows a typical resonance spike with its slow decay and Fig. 7 depicts the result of the scan together with a fit by a Gaussian

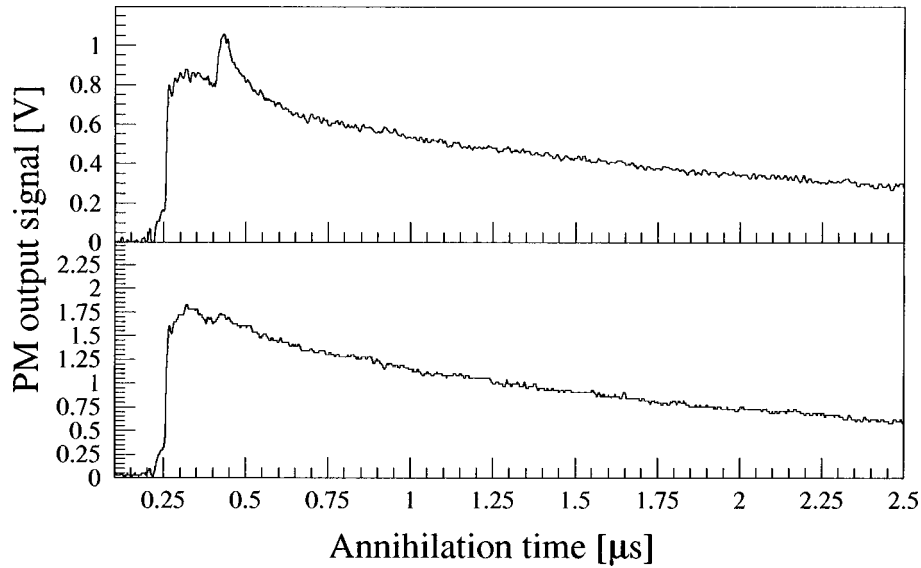


FIG. 6. (a) Resonance spike in the ADATS generated by the laser-induced transition $(37,34) \rightarrow (36,33)$ in $\bar{p}\text{-}^3\text{He}^+$, taken at $\lambda = 524.155$ nm. (b) Off-resonance data, taken at a wavelength smaller by 26 pm.

with fixed width ($\Gamma_{\text{Gauss}} = 7$ pm, to account for the finite bandwidth of the laser) convoluted with a Lorentzian (width free). From this fit an experimental value

$$\lambda_{\text{expt}}[(37,34) \rightarrow (36,33)] = 524.155 \pm 0.004 \text{ nm}$$

for the resonance wavelength was found at $\rho_{\text{atom}} = 9.1 \times 10^{20} \text{ cm}^{-3}$. The error includes a 3-pm uncertainty of the wavelength calibration. The λ value differs by less than 6 ppm from the theoretical prediction for an isolated atom, $\lambda_{\text{th}} = 524.158$ nm [28], which includes the corrections for the relativistic motion of the e^- . A small positive difference $\Delta\lambda = \lambda_{\text{th}} - \lambda_{\text{expt}}$, observed here at low statistical significance, was already seen for other transitions; it may be attributed to missing additional corrections in the calculation.

The decay of the laser-induced annihilation peak could be adequately fitted by an exponential. The resulting lifetime

$\tau_{(36,33)} = 42.2 \pm 2.6$ ns is in reasonable agreement with the result derived from the depletion spectrum at 690 mbar, $\tau_{(36,33)} = 52.6 \pm 7.7$ ns.

V. SUMMARY AND CONCLUSIONS

The $\nu=2$ cascade in antiprotonic ^3He has been investigated. The last metastable level in this cascade ($n=36, \ell=33$) has an intrinsic decay rate of $6.7 \pm 1.3 \mu\text{s}^{-1}$, which is by a factor of 8 larger than the radiative rate and about three times larger than the value calculated for the sum of Auger and radiative rates. It is quenched with a cross section of $\sigma_{\text{Stark}} = 3.0_{-1.0}^{+0.9} \times 10^{-19} \text{ cm}^2$. At higher pressure values only the lowest metastable level (36,33) is quenched, this, however, to an extent that enabled the feeding transition $(37,34) \rightarrow (36,33)$ to be investigated by laser-resonance spectroscopy. The wavelength of this transition was found to be $\lambda_{\text{expt}}[(37,34) \rightarrow (36,33)] = 524.155 \pm 0.004$ nm, only

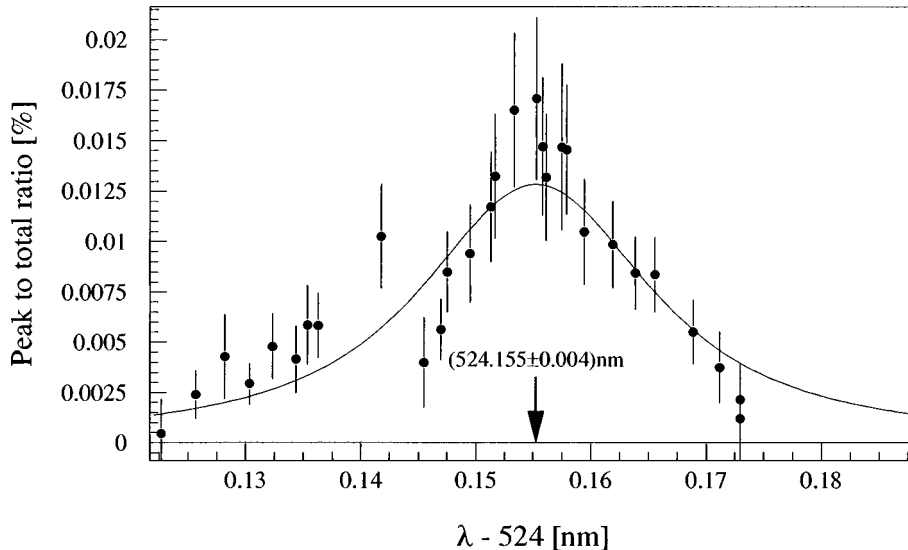


FIG. 7. Resonance curve for the 524-nm transition $(37,34) \rightarrow (36,33)$ in $\bar{p}\text{-}^3\text{He}^+$.

6 ppm smaller than the theoretical prediction of Korobov, which includes the relativistic corrections.

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