Two-photon laser spectroscopy of antiprotonic helium atoms, and the antiproton-to-electron mass ratio

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Abstract. Some two-photon transitions in antiprotonic helium atoms at the deep UV wavelengths $\lambda = 139.8\text{–}197.0$ nm were recently studied by laser spectroscopy. The thermal Doppler broadening of the observed antiprotonic resonances were reduced by exciting the atoms with two counterpropagating laser beams of wavelengths $\lambda = 265\text{–}417$ nm. The resulting narrow spectral lines allowed the measurement of three transition frequencies in antiprotonic helium-3 and helium-4 isotopes with fractional precisions of 2.3–5 parts in $10^9$. By comparing the results with three-body QED calculations, the antiproton-to-electron mass ratio was derived as 1836.1526736(23). We briefly review these experimental results that were presented in Ref. [1].

1. Introduction
Metastable antiprotonic helium ($\bar{p}\text{He}^+$) is a three-body atom [1, 2, 3, 4] consisting of a helium nucleus, an electron in the 1s state, and an antiproton occupying a Rydberg state with high principal and angular momentum quantum numbers $n \sim \ell + 1 \sim 38$. The atom retains microsecond-scale lifetimes against antiproton annihilation in the helium nucleus, due to the fact that the antiproton orbital has a negligible overlap with the nucleus; the 1s electron protects the antiproton against collisions with other helium atoms. This longevity makes $\bar{p}\text{He}^+$ especially amenable to precision laser spectroscopy [5, 6, 7]. The atomic transition frequencies of $\bar{p}\text{He}^+$ have been calculated by three-body QED calculations to fractional precisions of $1 \times 10^{-9}$ [8]. These calculations included relativistic and radiative recoil corrections up to order $m_e c^2 \alpha^6 / h$, and nuclear size effects. By comparing the measured and calculated transition frequencies, the antiproton-to-electron mass ratio can be determined to parts-per-billion scale precision.

The ASACUSA collaboration at CERN [9] has measured some $\bar{p}\text{He}^+$ transition frequencies in the optical region $0.3\text{–}1$ PHz with a fractional precision of $10^{-7} \text{–} 10^{-8}$, by single-photon laser spectroscopy [5, 6, 7]. The precision was limited by the Doppler broadening of the resonance lines which arose from the thermal motions of the $\bar{p}\text{He}^+$ in the target. Unlike the atomic hydrogen case [10], it is difficult to cancel the first-order Doppler broadening by irradiating the atom with two equal-frequency photons and inducing a two-photon transition; the probabilities involved in these nonlinear transitions of the massive antiproton are too small (a few a.u.). In fact, calculations indicate that gigawatt-scale laser powers would be needed to excite the antiproton within the atom’s microsecond-scale lifetime against annihilation [11].

In a recent experiment, two-photon transitions of the type $(n, \ell) = (n-2, \ell-2)$ [1] [Fig. 1(a)] were excited by utilizing the fact that the probability can be enhanced by factor $\sim 10^3$, to
Figure 1. Schematic energy level diagram of $^3\text{He}^+$ states involved in the two-photon transition $(n, \ell) = (36, 34) \rightarrow (34, 32)$ (a). The virtual intermediate state is tuned some $\Delta \omega_c \sim 10$ GHz from a real state $(35, 33)$. Cherenkov detector signals for two-photon transition (b). Experimental layout (c). See Ref. [1] for details.

around $\sim 10^4$ a.u., if the counterpropagating beams have non-equal frequencies $\nu_1$ and $\nu_2$, such that the virtual intermediate state of the two-photon transition lies within a few GHz of a real $^3\text{He}^+$ state $(n-1, \ell-1)$. The first-order Doppler width in the observed resonance lines can then be reduced by a factor $|\nu_1 - \nu_2|/(\nu_1 + \nu_2) \sim 1/20$.

2. Experiment

The two-photon transitions were induced between $^3\text{He}^+$ states with microsecond and nanosecond-scale lifetimes against Auger emission of the electron. After Auger decay, the remaining two-body $^3\text{He}^{2+}$ ion [12] was rapidly destroyed by Stark collisions with other helium atoms in the experimental target. The charged pions emerging from the resulting antiproton annihilations were detected by Cherenkov detectors [13] placed around the experimental target. The two-photon resonance condition between the counterpropagating laser beams and the $^3\text{He}^+$ was thus revealed as a sharp spike in the rate of antiproton annihilations [Fig. 1(b)].

Megawatt-scale laser pulses of high spectral purity are needed to excite these nonlinear two-photon transitions, that have amplitudes of $10^3$–$10^4$ a.u. We therefore developed two sets of Ti:Sapphire lasers [14] of pulse length 30-100 ns with a narrow spectral linewidth (~ 6 MHz). The laser system included continuous-wave (cw) lasers of wavelengths 728–940 nm, whose frequencies were measured to a precision of $< 1 \times 10^{-10}$ against a femtosecond optical frequency comb [15]. This beam was then used to seed a ring Ti:Sapphire oscillator and multipass amplifier which generated laser pulses of energy 50–100 mJ.

Nanosecond-scale changes in the refractive index of the Ti:Sapphire crystals during the amplification, as well as the so-called “mode-pulling” effects in the pulsed laser cavity [14], caused the laser linewidth to broaden and the frequency to shift by several tens MHz. This frequency chirp was measured using a heterodyne spectrometer, and corrected by intracavity electro-optic modulators located inside the Ti:Sapphire cavity. The spectral precision ($< 1.4 \times 10^{-9}$) of the
pulsed laser was verified by measuring some two-photon transition frequencies in Rb and Cs at wavelengths of 778 and 822 nm.

The Antiproton Decelerator (AD) facility of CERN provided 200-ns-long pulsed beams, which contained $\sim 10^7$ antiprotons of kinetic energy 5.3 MeV, at a repetition rate of 0.01 Hz [Fig. 1(c)]. The antiprotons were decelerated to $\sim 70$ keV, by allowing them to pass through a 3-m-long radiofrequency quadrupole decelerator [6]. Secondary electron emission detectors measured the spatial profiles of the beam [16]. The antiprotons were allowed to stop in a cryogenic target chamber filled with $^4$He or $^3$He gas at temperature $T \sim 15$ K and pressure $p = 0.8 - 3$ mbar. Two, horizontally-polarized laser beams of energy density $\sim 1$ mJ/cm$^2$ fired through the target in a perpendicular direction to the antiproton beam excited the two-photon transitions.

The Cherenkov signal corresponding to some $10^7$ $\bar{p}$He$^+$ atoms is shown in Fig. 1(b), as a function of time elapsed since the arrival of antiproton pulses at the experimental target. Lasers of wavelengths $c/\nu_1 = 417$ and $c/\nu_2 = 372$ nm were tuned to the two-photon transition $(n, \ell) = (36, 34) \rightarrow (34, 32)$, so that the virtual intermediate state lay $\Delta \nu_d \sim 6$ GHz away from the real state (35, 33). The annihilation spike which corresponds to the two-photon transition can be seen at $t = 2.4 \mu$s. The intensity of the spike reflects the number of antiprotons populating state $(36, 34)$ [17, 18, 19]. When the 417-nm laser was tuned some $0.5$ GHz off the two-photon resonance condition, the signal disappeared as indicated in the same figure.

Fig. 2(b) shows the resonance profile which was measured by detuning the $\nu_1$ laser to $\Delta \nu_d = -6$ GHz, whereas the $\nu_2$ laser was scanned between -1 and 1 GHz around the two-photon resonance defined by $\nu_1 + \nu_2$. The linewidth ($\sim 200$ MHz) of this two-photon resonance is more than an order of magnitude smaller than the Doppler- and power-broadened profile of the single-photon one $(36, 34) \rightarrow (35, 33)$ [Fig. 2(a)]. The two-peak structure, with a frequency interval of 500 MHz, arises due to the interaction between the electron spin and the orbital angular momentum of the antiproton. We also detected the $(33, 32) \rightarrow (31, 30)$ resonance of $\bar{p}^3$He$^+$ at a wavelength of 139.8 nm, using lasers of $c/\nu_1 = 296$ nm and $c/\nu_2 = 265$ nm [Fig. 2(c)] detuned $\Delta \nu_d \sim 3$ GHz from state $(32, 31)$. All four hyperfine lines are much closer together ($\pm 100$ MHz).

We also measured the $\bar{p}^3$He$^+$ resonance $(35, 33) \rightarrow (33, 31)$ of $\lambda = 193.0$ nm [Fig. 2(d)] using lasers of 410 and 364 nm. This resonance profile contains eight partially-overlapping hyperfine lines.
lines, which arose from the spin-spin interactions of the three constituent particles.

The spin-independent transition frequencies \( \nu_{\text{exp}} \) were obtained by fitting these measured profiles with a theoretical lineshape (solid lines in Fig. 2) which was determined by numerically solving the rate equations of the two-photon process [11]. This included the transition rates, power and Doppler broadening effects, frequency modulation in the laser pulse, the experimentally-measured spatial and temporal profiles of the laser beam, and ac Stark effects. The positions of the hyperfine lines were fixed to the theoretical values calculated by Korobov [20], which have a precision of \(< 0.5\) MHz. For the transition \((36, 34) \rightarrow (34, 32)\) in \(p^3\)He\(^+\), the statistical uncertainty due to the finite number of atoms in the laser beam was estimated as 3 MHz. For the target densities studied here \(\rho = (1 - 3) \times 10^{18}\) cm\(^{-3}\), no significant collisional shift was observable within the experimental error. This agrees with quantum chemistry calculations [21], for which the predictions of 0.1–1-MHz collisional shifts in the single-photon lines agreed with experimental results [7] with a precision of \(\leq 20\%\). Theoretical calculations [11] also show that magnetic Zeeman shifts are also small \(< 0.5\) MHz. The ac Stark shift [11] was reduced to \(< 5\) MHz by adjusting the relative intensities of the two laser beams. Remaining ac Stark shifts were canceled to a level of 0.5 MHz by systematically comparing the resonance profiles measured at positive and negative detunings \(\pm \Delta \nu_{d} \) of the virtual intermediate state. The experimental uncertainty \(\sigma_{\text{exp}}\) was obtained as the quadratic sum of all these errors.

3. Results and conclusions

The experimental transition frequencies \( \nu_{\text{exp}} \) (filled circles with error bars in Fig. 3) agree with the QED calculated \( \nu_{\text{th}} \) values (squares) within a fractional precision of \((2 - 5) \times 10^{-9}\). The calculation uses the fundamental constants compiled in CODATA2002 [22], such as the \(^{3}\text{He}\) and \(^{4}\text{He}\)-to-electron mass ratios, the Bohr radius, and Rydberg constant. The charge radii of the \(^{3}\text{He}\) and \(^{4}\text{He}\) nuclei give relatively small corrections to \( \nu_{\text{th}} \) of 4 – 7 MHz, whereas the correction from the antiproton radius is less than 1 MHz. The small contributions are due to the fact that the wavefunctions of the antiprotonic states with large \( \ell \)-value have only a negligible

Figure 3. Fractional deviation between theoretical (squares) and experimental (circles) transition frequencies of \(p^3\)He\(^+\) isotopes measured by two-photon laser spectroscopy. From Ref. [1].

Figure 4. Antiproton-to-electron mass ratio determined in this work, compared with the proton-to-electron mass ratios measured in Penning trap experiments [22, 23, 24, 25] and the CODATA 2002 recommended value obtained by averaging them. From Ref. [1].
overlap with the helium nucleus. The theoretical precision of $\nu_{th}$ is now mainly limited by the uncalculated radiative corrections of order $m_e c^2 \alpha^5 / \hbar$ [8], but higher-order corrections are now being calculated. When the antiproton-to-electron mass ratio $M_p/m_e$ in these calculations was increased by a relative amount of $10^{-9}$, the $\nu_{th}$-value changed by 2.3–2.8 MHz. By minimizing the difference between $\nu_{th}$ and $\nu_{exp}$ and considering the systematic errors, we obtained the antiproton-to-electron mass ratio as,

$$M_p/m_e = 1836.1526736(23),$$  \hspace{1cm} (1)

which yielded the best agreement between theoretical and experimental frequencies. The uncertainty includes the statistical and systematic experimental, and theoretical contributions of $18 \times 10^{-7}$, $12 \times 10^{-7}$, and $10 \times 10^{-7}$. This is in good agreement with previous measurements[22, 23, 24, 25] of the proton-to-electron mass ratio (Fig. 4), which have a similar experimental precision. By assuming that CPT invariance is valid (i.e., $M_p = M_p$ = 1.00726746677(10) u), we further derived a value for the electron mass, $m_e$ = 0.005485799091(7) u, from this experimental result on pHe$^+$ [1].

Hughes and Deutch [26, 27] has constrained any difference between the antiproton and proton charges and masses $\delta Q = (Q_p + Q_\text{He})/Q_p$ and $\delta M = (M_p + M_\text{He})/M_p$ to better than $2 \times 10^{-5}$. To do this, they combined X-ray spectroscopic data of antiprotonic atoms (proportional to $Q_p^2 M_\text{He}$) and the cyclotron frequency ($\propto Q_p/M_p$) of antiprotons confined in magnetic Penning traps measured to a higher precision. We improved this limit by studying the linear dependence of $\delta M$ and $\delta Q$ on $\nu_{th}$ of pHe$^+$, i.e., $\delta M \propto M_p + \delta Q < |\nu_{exp} - \nu_{th}|/\nu_{exp}$ [2]. For the three pHe$^+$ transitions studied in this work, the constants were estimated as $\kappa_M = 2.3 - 2.8$ and $\kappa_Q = 2.7 - 3.4$, whereas the right side of this equation was evaluated by averaging over the three transitions as $< (8 \pm 15) \times 10^{-10}$. Meanwhile the constraint of $(Q_p/M_p)/(Q_p/M_p) + 1 = 1.6(9) \times 10^{-10}$ from the TRAP experiment [28, 29] implies that $\delta Q \approx \delta M$. We conclude from this that any deviation between the charges and masses of protons and antiprotons are less than $7 \times 10^{-10}$ at 90% confidence level [1].

We are currently attempting to further improve the experimental and theoretical uncertainties in these experiments, by e.g., cooling the atoms to lower temperature and improving the quality of the antiproton beam.

**Table 1.** Spin-averaged transition frequencies of pHe$^+$. Experimental values show respective total, statistical and systematic errors in parentheses. Theoretical values (From Ref. [8] and V. I. Korobov, private communication) show respective uncertainties from uncalculated QED terms and numerical errors in parentheses. From Ref. [1].

| Isotope   | Transition   | Transition frequency (MHz) | Experiment | Theory |
|-----------|--------------|-----------------------------|------------|--------|
|           | $(n, \ell) \rightarrow (n - 2, \ell - 2)$ |                           |            |        |
| p$^3$He$^+$ | (36, 34) → (34, 32) | 1,522,107,062(4)(3)(2)     | 1,522,107,058.9(2.1)(0.3) |
|           | (33, 32) → (31, 30) | 2,145,054,858(5)(5)(2)     | 2,145,054,857.9(1.6)(0.3) |
| p$^3$He$^+$ | (35, 33) → (33, 31) | 1,553,643,100(7)(7)(3)     | 1,553,643,100.7(2.2)(0.2) |

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