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Laser Cooling Positronium with Broadband Laser Pulses

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Abstract. The first successful demonstration of broadband laser cooling of positronium (Ps) atoms, obtained within the AEgIS experiment at CERN, is presented here. By employing a custom-designed pulsed alexandrite laser system at 243 nm featuring long-duration pulses of 70 ns and an energy able to saturate the 1^3S–2^3P transition over the broad spectrum range of 360 GHz, the temperature of a room-temperature Ps cloud was reduced from 380 K to 170 K in 70 ns. This advancement opens new avenues for precision spectroscopy, antihydrogen production, and fundamental tests with antimatter.

1 Introduction

Positronium (Ps), the bound state of an electron (e^-) and its antiparticle, the positron (e^+), has been extensively used for testing fundamental symmetries and bound-state quantum electrodynamics (QED) [1], as well as an intermediate system to form antihydrogen in charge-exchange processes with antiprotons [2, 3]. Most of these experimental schemes rely on the availability of intense sources of cold Ps (room temperature or below). Laser cooling has been long discussed in the literature as a crucial step for enhancing both Ps precision spectroscopy measurements [4, 5] and antihydrogen production schemes [2, 6], as well as paving the way towards the first Bose-Einstein condensation of antimatter-containing species [7, 8].

In the specific case of antihydrogen production by charge-exchange with Rydberg Ps i.e., the main $\bar{\mathrm{H}}$ production mechanism chosen by the AEgIS (Antihydrogen Experiment: gravity, Interferometry, Spectroscopy) collaboration at CERN, aiming at gravitational studies with neutral antimatter systems [9] and in which context this work was conducted, successful Ps laser cooling allows accessing unprecedented cold Ps sources to enhance the $\bar{\mathrm{H}}$ production cross-section. Indeed, the cross-section σ not only scales as $\sigma \propto n^4$, where n is the principal quantum number of Ps, but also exhibits a $\sigma \propto {E_{Ps}}^{-1}$ dependency to the kinetic energy of Ps, E_{Ps}, in the low energy limit ($E_{Ps} \to 0$) [10]. The availability of laser cooled sources can lead to significant yield increases in $\bar{\mathrm{H}}$ production schemes, either in the low magnetic field or in the Paschen-Back regimes, whereas the Zeeman mixing regime (magnetic fields in the range $0.01 - 1.0\,\mathrm{T}$) is disfavoured by competing magnetic quenching loss mechanisms [11, 12].

Here the first successful implementation of broadband Doppler cooling on Ps is discussed, demonstrating the cooling of a significant fraction of the original Ps distribution in free-flight, by strongly saturating the $1^3S \to 2^3P$ transition using a broadband, long-pulsed 243 nm alexandrite laser [13]. The experiments were conducted in a magnetic and electric field-free environment to minimize perturbations to the Ps atoms.

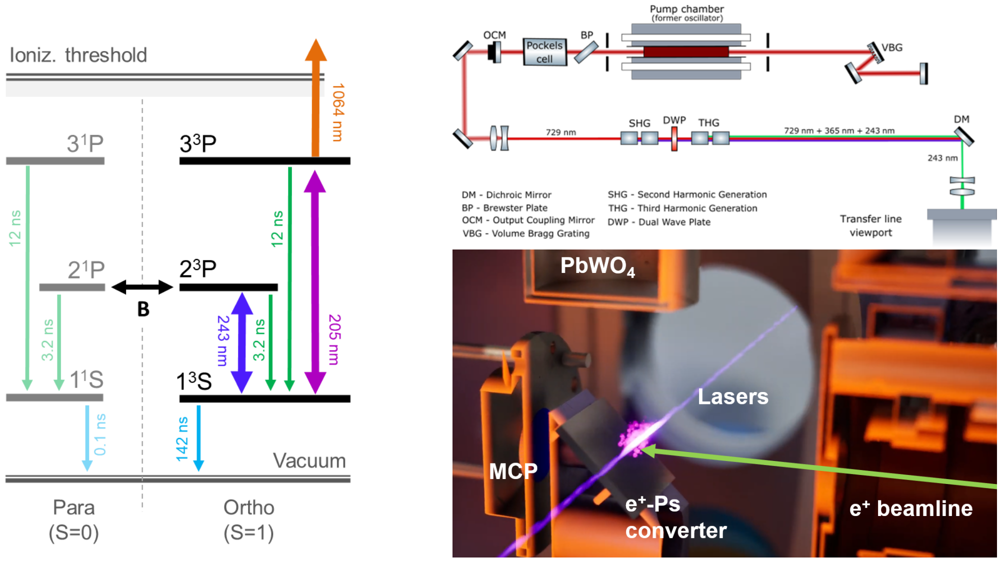

Figure 1: Left: Grotrian diagram of Positronium for the energy levels relevant for this work. Right: experimental scheme of the alexandrite laser used for laser cooling Ps (top, reproduced with permission from [18]) and of the interaction chamber (bottom).

2 Theoretical background

The scheme discussed here is based on Doppler cooling, targeting the Ps 1^3S–2^3P transition at 243 nm (see Fig. 1, left). In brief, Ps atoms in the 1^3S ground state absorbing photons with momentum opposing the atoms' movement from the red-detuned laser field are first excited to the 2^3P state. Subsequently they can either decay back to 1^3S by stimulated emission, also a directional process causing no net transfer of momentum, or by spontaneously emission, which is non-directional and on average induces a net transfer of momentum. Repeated iterations of these so-called cooling cycles, lasting 6.28 ns each, lead to the cumulative cooling effect referred to as Doppler cooling.

The 142 ns short lifetime of the ortho-Ps 1^3S ground state (the only one relevant for experimentation, as para-Ps 1^1S lifetime is only of 0.125 ns) and its 1/925 lighter mass than hydrogen, pose significant challenges to implementing standard Doppler cooling techniques. Previous theoretical studies have proposed various laser cooling schemes for Ps, including broadband Ps Doppler cooling [14] and chirped-pulse Ps Doppler cooling, which was also very recently reported for the first time [15]. Experimental efforts have been limited by the necessity to develop custom laser systems specifically designed to this purpose, featuring long pulse durations in the order of 100 ns (i.e., comparable to the 1^3S annihilation lifetime) and either 100 GHz-broad linewidths, for the case of broadband Doppler cooling, or the ability to reproducibly scan the laser frequency over 49 GHz in 100 ns, for the case of chirped-pulse Doppler cooling. These laser systems have become available only very recently [16, 17].

The use of a laser pulse able to address a significant portion of the initial Ps velocity distribution is crucial for efficient Doppler cooling. Doppler broadening of Ps transitions at room temperature is approximately 500 GHz, significantly larger than the 1^3S–2^3P transition natural linewidth of 0.3 GHz, and of typical Doppler cooling experiments on ordinary atoms. Hence, in order to cool a substantial fraction of the Ps velocity distribution, a broadband laser with an effective cooling linewidth in the hundreds GHz range is necessary.

3 Experimental

3.1 Cooling laser system

The third harmonic of broadband Q-switched alexandrite laser (schematized in Fig. 1, top-right panel), operating at 729 nm, was employed to drive the 1^3S–2^3P transition in Ps in a double-pass scheme. Its 1 m Q-switched oscillator was realized in a plane-parallel cavity with a flashlamp-pumped alexandrite rod, a Pockels-cell and a Brewster plate polarizer. The oscillator was forced to lase at 729 nm by introducing a volume Bragg grating in the cavity, which could be adjusted in angle to obtain fine wavelength tuning. The alexandrite rod was held at 70 °C to maximize the overall gain at this wavelength. Second and third harmonic generation stages were realized with lithium borate and barium borate crystals, respectively. This laser was able to produce 70 ns, 243 nm light pulses with an energy per pulse up to 2.3 mJ. Its spectrum is characterized by a comb structure with 450 MHz-distant modes, enveloped by a Gaussian amplitude profile of 101 GHz sigma. The high laser fluence obtained with typical beam spots of $\approx 10\,\mathrm{mm}^2$ strongly saturated the Ps transition, resulting in the spectral gaps between the modes be filled up over a large 360 GHz spectral interval. For further details about this laser, see [16].

3.2 Positronium Production

A pulsed e^+ beam from the AEgIS positron system was directed onto a porous silica target to produce Ps atoms with a thermal velocity distribution (see [19]). The e^+ transport beamline was operated in a fully electrostatic mode (as described in [20]), to prevent external magnetic fields in the measurement region and quenching between ortho- and para- states (Fig. 1, left). The target was maintained at room temperature, and the positrons implanted into the silica formed ortho-Ps atoms with a $\approx 30\%$ conversion efficiency, which subsequently diffused out into the vacuum. A micro-channel plate (MCP) detector was used to steer the e^+ beam onto the target (see Fig. 1, right). Although the MCP could in principle be used to image photo-e^+ from Ps photoionization events (as developed in [21]), the absence of a guiding magnetic field for photo-e^+ and the presence of strong laser backgrounds limited its usability.

3.3 Detection Methodology

In order to detect laser cooling of Ps, two techniques were combined in the same experimental setup (see Fig. 1, bottom-right panel): Single-Shot Positron Annihilation Lifetime Spectroscopy (SSPALS, see [22]), used to measure the total amount of Ps in the 1^3S ground state, and 1^3S–3^3P photoionization spectroscopy by means of two additional probing lasers, to measure the Ps cloud root-mean-square velocity along the laser propagation axis by fitting its Doppler-broadened lineshape (see [23] for the details).

A $25 \times 25 \times 20$ mm $PbWO_4$ scintillation detector coupled to a R11265-100 photomultiplier tube and a 2.5 Gs 12-bit digitizer were used for measuring SSPALS spectra in the different configurations of the cooling and probing lasers, necessary for each of the measurement in the campaign. SSPALS spectra acquired alternating lasers on and off, in each of the lasers configurations, were reduced to the single parameter $S = (N_{nolaser} - N_{laser})/N_{nolaser}$ i.e., the normalized difference in the amount of detected 1^3S Ps, N, with and without lasers (see [23]). The probing lasers included a 1.5 ns, 205 nm pulse driving the 1^3S–3^3P transition, and a strong 1064 nm, 4 ns pulse efficiently photo-ionizing the atoms on the 3^3P state. The Ps cloud root-mean-square velocity along the laser propagation axis is obtained from a Gaussian fit of the $S_{205+1064}(\lambda_{205})$ parameter obtained as a function of the 205 nm laser wavelength λ_{205} (see Fig. 2, top left, for an example).

4 Results and Discussion

4.1 Laser-Induced Positronium Lifetime Extension

Upon irradiation with the 243 nm laser alone, an increase in the number of ground-state Ps atoms was observed (see Fig. 2, top left), as a consequence of the atoms having spent time $\approx 50\%$ of the laser interaction time in 2^3P states. These have negligible annihilation rates in the time scales relevant for this experiment. As a result, the expected annihilation rate of the cycling 1^3S–2^3P atoms halves, and the expected lifetime doubles to 284 ns, resulting in more atoms are in the 1^3S state after spontaneous decay to the ground state. An increase in the 1^3S population up to 9 % was observed, with a characteristic Lamb-dip spectral structure due to the double-pass laser interaction scheme (see, for instance, [24]). This structure is not present in the $S_{205+1064}(\lambda_{205})$ spectrum of Fig. 2, bottom left, due to the absence of a retro-reflected 205 nm beam.

An important consequence of this effect is that, in order to detect unambiguously Doppler cooling, a correction to the standard SSPALS analysis methodology based on S parameters had to be applied to subtract the contribution from lifetime extension. An S_{cool} parameter was defined as $S_{cool} := S_{243+205+1064} - S_{243}$, where $S_{243+205+1064}$ is constructed from SSPALS spectra with all lasers active, and S_{243} is constructed from SSPALS spectra with only the cooling laser present, isolating the lifetime extension effect to subtract it (as in Fig. 2, top left).

4.2 Doppler Cooling

The Doppler cooling effect was investigated by two methods.

The first method consisted in scanning the probing laser wavelength λ_{205} while the cooling laser detuning was set to -200 GHz, and the modifications due to cooling to the Doppler broadened 1^3S–3^3P lineshape were observed with and without the presence of the cooling laser (see Fig. 2, top right panel).

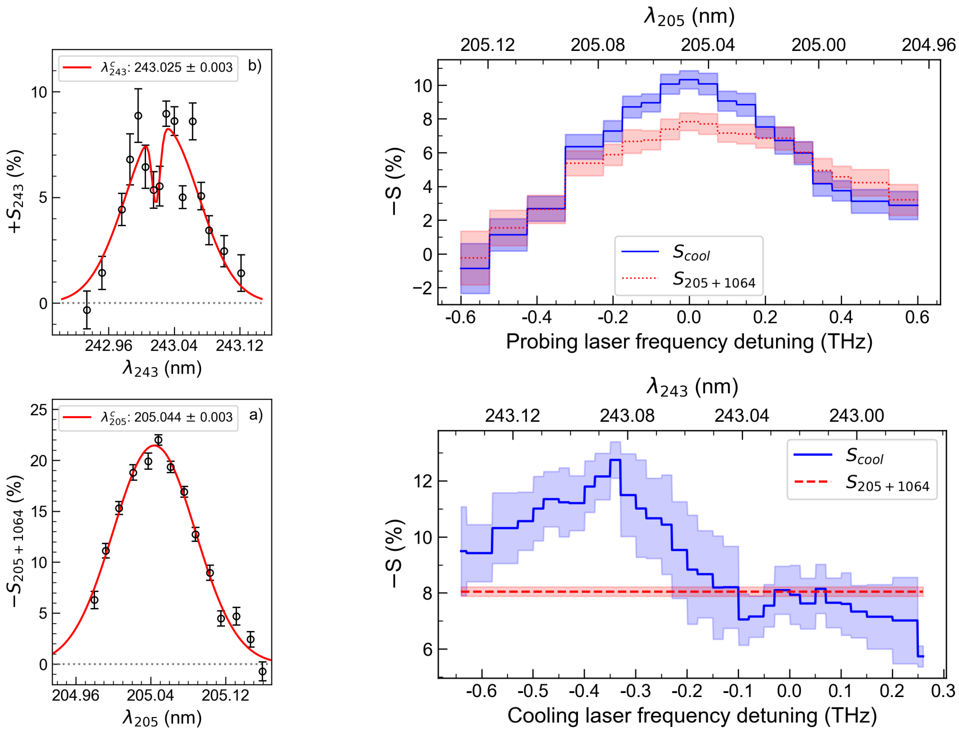

Figure 2: Left: Observation of the Ps lifetime extension effect due to the cycling along the 1^3S–2^3P transition by the long-pulsed 243 nm cooling laser (top), compared to a reference spectroscopic scan along the 1^3S–3^3P transition with the probing 205 nm+1064 nm lasers (bottom). Right: Observation of the laser cooling effect, by (top) the comparison of the 1^3S–3^3P transition linewidth, measured by the scanning the probe laser detuning, with (blue) and without (red) the presence of the cooling laser, and (bottom) the increment of the Ps atoms falling into the probe laser bandwidth, set on resonance of the 1^3S–3^3P transition, compared to the reference case without cooling laser, while scanning the cooling laser detuning. Reproduced from [13].

The initial 1D temperature of the Ps ensemble was measured in the dataset without cooling laser to be 380(20) K (Fig. 2, top right, red). After applying the cooling laser, the 1D temperature was reduced to 170(20) K (Fig. 2, top right, blue), indicating that cooling of a significant fraction of the velocity distribution had occurred. This effect is compatible with 11 cooling cycles, in good agreement with out expectations with a 70 ns cooling pulse. This is also a strong indication that our laser is efficient in cooling over a broad frequency range.

The second method consisted in scanning the cooling laser detuning while leaving the probing laser at the 1^3S–3^3P transition resonance at 205.047 nm. In this scheme, the probing laser acts as a collector of atoms falling into its bandwidth, whereas the cooling laser acts by pushing the atoms towards the probing laser bandwidth. As one would expect, the more the cooling laser is red-detuned from the transition resonance, the more atoms are detected as a result of Doppler cooling (Fig. 2, bottom right). In contrast, blue detuning causes a reduction of the detected atoms. The maximum effect is obtained at −350 GHz, in good agreement with the expected cooling saturation interval of 360 GHz. In this setting, an excess of 58(9) % due to cooling only is observed i.e., after correcting by the lifetime extension.

5 Conclusion and Future Outlook

Broadband Doppler cooling of Ps was demonstrated by developing and employing a broadband, long-pulsed, alexandrite-based 243 nm laser to saturate its $1^3S \rightarrow 2^3P$ transition. Ps atoms were produced by a nanoporous silica converter from a ns e^+ bunch in a field-free environment, and efficient cooling of a significant fraction of the Ps distribution was observed, with the 1D temperature of the ensemble reduced from 380(20) K to 170(20) K, on the top of an extension of the lifetime of the Ps atoms. It is worth noticing that this lifetime extension effect is advantageous for future experimental schemes employing

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laser cooled Ps, as it can enhance quite significantly the number of Ps atoms in the final state after the cooling has taken place. A future improved scheme of the experiment is being currently designed, aiming at cooling the entire Ps cloud to a final temperature of 10 K or better, by employing a cryogenic Ps converter and a longer cooling pulse.

The successful demonstration of Ps laser cooling of positronium opens new avenues for a variety of future experiments. One immediate application is the enhancement of precision spectroscopy measurements of Ps. The reduction in thermal velocity spread directly translates into narrower spectral lines, which will improve the accuracy of measurements of transition frequencies and thereby provide more stringent tests of QED. In the future, Ps laser cooling may lead to Ps Bose-Einstein condensation. The temperature reduction achieved in this work is several orders of magnitude away from condensation, assuming typical free space Ps cloud densities of $\approx 10^5\,\mathrm{mm}^{-3}$. Achieving BEC in a leptonic system such as Ps would enable studies of matter-antimatter interactions under conditions where quantum statistical effects are significant. Finally, and most importantly for the AEgIS collaboration, the techniques discussed here can be applied to the preparation of cold antihydrogen atoms. The ability to cool Ps efficiently is a critical step towards developing a new generation of intense pulsed source of antihydrogen to test fundamental symmetries such as the CPT theorem and the weak equivalence principle.

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