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Positronium laser cooling: results and perspectives

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The experimental demonstration of positronium laser cooling with stationary broadband laser pulses with negative detuning is briefly described. Considerations on the limits of the current experiment and possible future developments follow. In particular, the benefit of positron remoderation, use of a magnetic field, positronium polarization, pulse shaping, coherent laser cooling and deceleration are shortly discussed.

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Introduction

In 1988, following the rapid development of laser cooling of neutral atoms, E. P. Liang and C. D. Dermer studied numerically the possibility to apply laser cooling to positronium (Ps) [1], in view of forming a Bose Einstein Condensate (BEC) of this bound state of an electron (e^-) and a positron (e^+). Among the many potential applications motivating their simulations, they identified testing quantum electrodynamics (QED) using Doppler free spectroscopy and the possibility to create a GRASAR (gamma ray amplification by stimulated annihilation radiation) at 511 keV based on the annihilation of para-Ps in the BEC phase. In the 1993 seminal work of M. S. Fee et al. [2], laser cooling is pointed out as a way forward to perform a measurement of the 1^3S_1-2^3S_1 interval with a precision significantly better than the 1.3 MHz natural line width of the transition. In addition to these motivations, the pulsed production of antihydrogen based on the charge exchange between a plasma of antiproton and a cloud of Ps excited in a high principal quantum number Rydberg state as demonstrated by the AE$\bar{\text{g}}$IS collaboration [3] could be increased by orders of magnitude using cold clouds of Ps [4] by minimizing the relative velocity between Rydberg Ps atoms and antiprotons.

In [1], two different laser cooling approaches are proposed to reach the BEC critical temperature within the ortho-Ps annihilation lifetime (142 ns). The first approach mentioned consists in rapidly sweeping the laser frequency “so that all Ps in the Doppler wing are effectively “snowploughed” to lower velocities”. This option is rapidly discarded based on the difficulty of achieving the required sweeping rates. The second approach, considered technically more feasible, consists in using “a stationary broadband laser with negative detuning”. The paper ends on these words: “We urge the experimental community to attempt such an experiment.” In spite of this call and the development of innovative laser systems [5], the challenge of Ps laser cooling remained unresolved until 2024.

Indeed, Ps laser cooling presents a unique set of difficulties. First, the laser cooling timescale is limited by the Ps annihilation lifetime. In practice, only the ortho-Ps annihilation lifetime allows for long enough interaction time with a cooling laser. Second, the velocity distribution of pulsed Ps clouds produced by implantation of e^+ nanosecond bunches into nano-channeled Si targets are typically 10^5 m/s large [6]. Third, the wavelength of the 1^3S-2^3P transition (243 nm), which is best suited for Ps laser cooling [1], lies in the ultraviolet range where no laser gain medium is available. Finally, Ps is produced in scarce quantities and as many Ps atoms as possible should be preserved through the cooling process.

Results

In 2024, Ps laser cooling was demonstrated by two different experiments. The group at the University of Tokyo developed a chirped pulse-train generator based laser [7] reaching the impressive chirping rate of 490 GHz/μs to laser cool Ps [8] with an approach reminiscent of the “snowploughing” mentioned in [1]. Independently, the AE$\bar{\text{g}}$IS collaboration demonstrated Ps laser cooling [9] using a red-detuned stationary broadband ~100 ns long laser pulse [10]. In the latter, the temperature of a Ps cloud was reduced from 380(20) down to 170(20) K in approximately 70 ns of interaction with the cooling laser in a magnetic and electric field free environment. This reduction in temperature corresponds to 11 cooling cycles composed of absorption of one photon at 243 nm followed by spontaneous emission, each cycle lasting for 6.38 ns when half of the

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population is excited in the 2^3P manifold. The Doppler broadening of the 1^3S-2^3P transition is thereby reduced at the rate of 970 GHz/μs corresponding to the shift of the resonance frequency caused by the absorption of one 243 nm photon (6.17 GHz) every other spontaneous emission lifetime (2×3.19 ns).

To reach the limit of traditional Doppler laser cooling, we developed an alexandrite-based laser system with a root-mean-square (rms) spectral bandwidth of 101(3) GHz and capable of reaching 100 kW/cm^2 on an area of 10 mm^2 for 70 ns. Given the 20 MHz rms spectral width of the transition, the power on the resonance line amounts to 20 W/cm^2 which is much higher than the 0.45 W/cm^2 saturation intensity of the transition. Considering that the third harmonic of our multi-longitudinal mode pulsed laser presents large gaps of 450 MHz between individual laser gain modes, we estimate that the 1^3S-2^3P transition is saturated for all Ps atoms in an equivalent spectral range of 360 GHz (see [10] for more details). This is well suited to efficiently cool the broad Doppler distribution of the Ps clouds produced in our experiment. The velocity distribution was measured by Doppler sensitive two-photon resonant ionization on the 1^3S-3^3P transition using two synchronized few nanosecond long pulses at 205 and 1064 nm. The single-shot positron annihilation lifetime spectroscopy (SSPALS) technique [11] was used to diagnose the interaction of the cooling and probing lasers with Ps. This allowed for the observation of Ps lifetime extension due to the cooling laser induced continuous excitation of a large fraction of the Ps cloud in the 2^3P manifold for which the annihilation lifetime is orders of magnitude larger compared to the ground state. This attribute of the stationary broadband laser with negative detuning approach is instrumental in the case of Ps since it allows to preserve the atoms during the cooling process. These results are presented and discussed in details in [9].

Perspectives

One limitation of the experiment presented in [9] is the ability to probe the velocity distribution at longer delays due to the motion of the Ps atoms in the directions which are not laser cooled. Indeed, the Ps cloud propagates away from the nano-channeled conversion target and expands in the dimensions parallel to its plane. Since the dimensions of the 205 and 1064 nm laser probe beam profiles are finite, the Ps cloud fraction interacting with the probe lasers decreases with time. Two-dimensional laser cooling in the plane parallel to the target's surface with the identical stationary broadband laser with negative detuning would allow to partially improve the situation and extend the probe time to longer delays. For large frequency detuning of the cooling laser, 2- and 3-D standard Doppler cooling do not take significantly longer time than 1-D cooling [12]. As for the direction normal to the target plane, the same pulse cannot be directly used since the mean velocity is of the order of 10^5 m/s in this direction. It is in principle possible to reduce the rms of the velocity distribution in this direction using the stationary broadband laser with negative detuning approach with different central frequencies accounting for the mean velocity and direction of propagation along this axis. The mean velocity can then be brought down to zero by deceleration. The fastest and most efficient deceleration technique consists in applying a train of short laser pulses with alternating direction of propagation and detuning (negative detuning for pulses propagating against the mean velocity and positive detuning for pulses propagating along the mean velocity). Ideally, the Ps cloud is fully polarized [13] so that Ps atoms individually reduce to a two-level system, in

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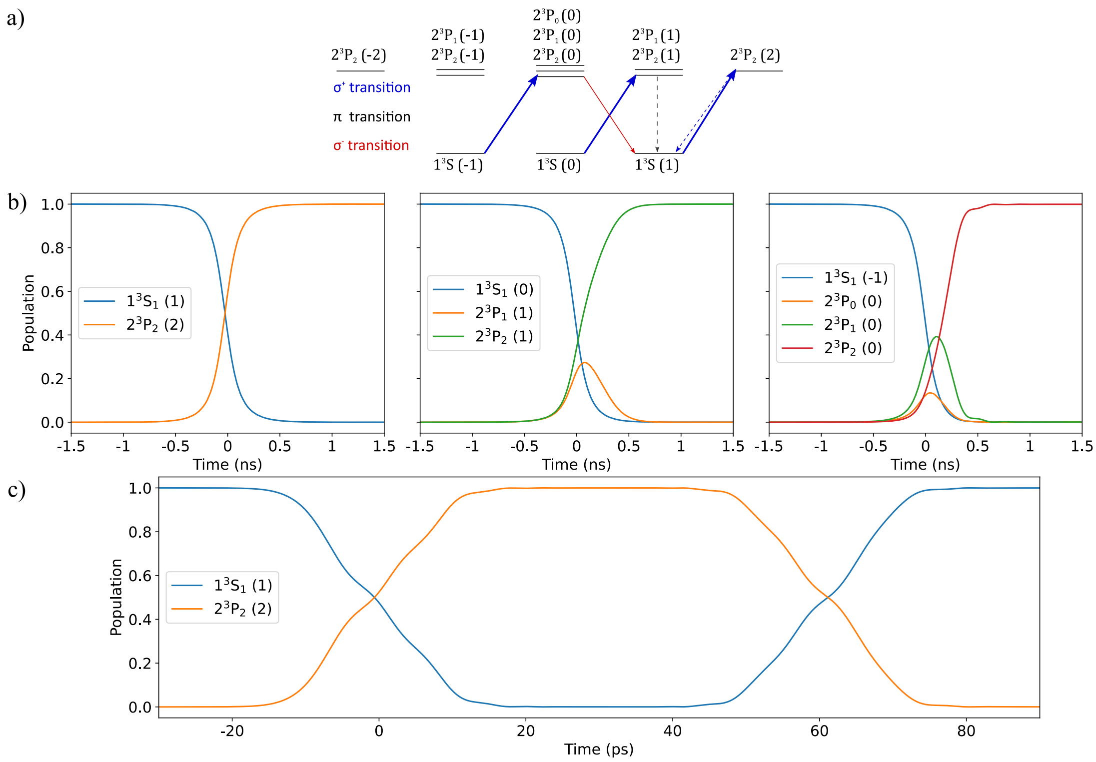

Figure 1: a) Ps Grotrian diagram for the 1^3S and 2^3P manifolds. b) Simulations of coherent population transfer in unpolarized Ps (1.25 ns chirped pulses). The same laser pulse is used for all three sub-panels (chirp rate 2π(0.375 GHz/ps), Rabi frequency: 2π(30 GHz)). c) Simulations of ultrafast coherent population transfer in polarized Ps (21.2 ps chirped pulses, chirp rate 2π(10 GHz/ps), Rabi frequency: 2π(75 GHz)). These parameters can be easily achieved with the current state of the art of femtosecond laser technology.

which case it is possible to use short chirped pulses to efficiently induce population transfer between the two levels by adiabatic rapid passage [14]. In Figure 1, we report on simulations of coherent population transfer between the 1^3S and 2^3P manifolds (see panel a). We show in particular that for unpolarized Ps samples, efficient population transfer can be achieved with laser pulses longer than 1 ns (see panel b) while the same result can be achieved with 21.2 ps long pulses in the case of fully polarized Ps. It is therefore highly beneficial to start from fully polarized Ps so that consecutive pulses can decelerate the Ps cloud along the direction normal to the target within a time short compared to the fluorescence lifetime of the excited state (3.19 ns). We note that, in principle, it is possible to prepare a fully polarized Ps cloud by applying a sequence of two pulses with the properties used for the simulations presented in Figure 1 b in a coherent optical pumping scheme. In addition, it might be experimentally more feasible to start with the deceleration step and then apply the stationary broadband laser with negative detuning approach using a single laser pulse for all three dimensions once the mean velocity is 0 on all three axes.

In order to keep as many Ps atoms alive and within the beam profile of the probe lasers, the amount of time required to reduce the Doppler broadening can be minimized using coherent laser cooling with trains of pulses [15]. Starting from a Ps cloud with smaller spatial dimensions can also help measuring the velocity distribution at large delays after Ps production. This can be achieved by

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focusing the e^+ bunch on smaller spots using advanced remoderation schemes [16]. Alternatively, a magnetic field can be used to compress the e^+ bunch. In the presence of a magnetic field, Ps triplet and singlet states start mixing, which results in the enhanced annihilation of Ps via a mechanism referred to as quenching [17]. Spin state mixing and quenching also happens in the Ps 2^3P manifolds which would seem to preclude Ps laser cooling in a magnetic field. A solution to this problem is to use strong enough magnetic fields (typically above 1 T) to reach the Paschen-Back regime where pure triplet states can be isolated spectrally from mixed states [17, 18].

Regarding the lowest temperature achievable with the stationary broadband laser with negative detuning approach, simulations [1] show that it is possible to reach equivalent temperatures below 1 K provided that the spectrum of the cooling laser is cutoff blueward of $\omega_0 - \mathrm{R}/\hbar$ where ω_0 is the resonant frequency and R is the recoil energy ($\mathrm{R}/\hbar \sim 3$ GHz). A high finesse Fabry-Perot interferometer in reflection was proposed as a technical solution to approach this goal [19]. Other solutions based on cutting-edge pulse shaping technology [20] can be implemented to reach the required spectral resolution.

Conclusion

In conclusion, the experimental demonstration of Ps laser cooling opens bright perspectives for precision spectroscopy on Ps [2, 21], increased production of antihydrogen, neutralization of cold trapped ions [22] and potentially a Ps BEC. Future developments include two- and three-dimensional laser cooling, coherent laser deceleration and laser cooling in a magnetic field in view of reaching even lower temperatures, to name of few. In order to reach Ps BEC, these developments should go hand-in-hand with increasing the density of Ps pulsed clouds.

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