Laser Excitation of Positronium in the Paschen-Back Regime

D. B. Cassidy, T. H. Hisakado, H. W. K. Tom, and A. P. Mills, Jr.
Department of Physics and Astronomy, University of California, Riverside, California 92521-0413, USA
(Received 2 February 2011; published 28 April 2011)

Zeeman mixing of singlet and triplet $2P$ states of positronium (Ps) atoms, followed by decay back to the ground state, can effectively turn a long-lived triplet atom into a short-lived singlet state, which would seem to preclude laser cooling of Ps in a magnetic field. Here we report experiments which show that, in fact, because of the large splitting of the $n = 2$ states in a high magnetic field (the Paschen-Back regime), the amount of such mixing diminishes approximately exponentially with an increasing magnetic field >0.01 T and is essentially eliminated above ~ 2 T. Thus, laser cooling of Ps should be feasible at high fields, which will facilitate the production of a Ps Bose-Einstein condensate.

DOI: 10.1103/PhysRevLett.106.173401 PACS numbers: 36.10.Dr, 34.80.Dp, 34.80.Nz

As a purely leptonic atomic system, positronium (Ps), the bound state between a positron and an electron [1], is very well described by quantum electrodynamics, and much is known theoretically about this system [2]. From the experimental side, however, the situation is less satisfactory, mostly due to the difficulty in obtaining low energy positrons in large quantities and the relatively short lifetime of Ps (142 ns or less). Thus, while the principal decay rates and energy intervals [3–6] have all been measured with great care, the level of precision available remains well below that of theory [2]. In addition, while certain more complex positronic systems have been produced, such as Ps^- ions [7], PsH [8], and Ps_2 molecules [9], two experiments that have been discussed for many years have yet to be attempted, namely, laser cooling of Ps [10] and the formation of a Ps Bose-Einstein condensate (BEC) [11].

Laser cooling is likely to be an important element in the production of a Ps BEC, since the critical temperature [12] is related to the Ps density by $T_c = (n_{\mathrm{Ps}}/10^{18}\ \mathrm{cm}^{-3})^{2/3} \times 14.6$ K. Thus, the colder the Ps, the lower the density required for a phase transition into the condensed state to occur. Our method for producing a high density of Ps involves the formation and compression of a positron plasma, which necessarily uses an axial magnetic field of ~ 0.04 T or more. It has been shown, though, that in magnetic fields greater than ~ 0.01 T, singlet and triplet P states with principal quantum number $n = 2$ will mix [13], such that after radiative (or stimulated) decay back to the ground state some fraction of the initially triplet population will be in the singlet state, leading to an increased Ps decay rate [14]. While this effect may be usefully employed as a means of detecting excited state Ps [15,16], it constitutes a loss mechanism that is incompatible with laser cooling.

Here we report experiments which show that this mixing of $2P$ positronium states is in fact suppressed at high magnetic fields. This occurs for $\mu_B B > \Delta E$, where μ_B is the Bohr magneton, B is the magnetic field in which the Ps is created, and ΔE is the approximate energy width of the $2P$ manifold, which corresponds to fields of the order of a few Tesla. This has important implications for experiments to produce a Ps BEC, insofar as it means that Ps may be laser cooled in a high magnetic field. Thus, the positron beam need not be extracted to a magnetic field-free region, considerably simplifying the experimental requirements.

Our experiment employed a positron accumulator explained in detail in Ref. [17]. By using a buffer gas trap [18], positrons from a ^{22}Na-based beam are captured and stored in an accumulator. The positrons were implanted into a p-doped Si target [19] in bunches containing $\sim 2 \times 10^7$ particles, with a time width of ~ 1 ns, creating bursts of ~ 0.16 eV Ps in vacuum. The laser systems used here are described in Ref. [20]. Briefly, a Q-switched Nd:YAG laser is used to pump a dye laser, producing 486 nm light that is frequency doubled in a β-barium borate crystal, providing up to ~ 1 mJ of UV light, tuned to the $1S - 2P$ transition at vacuum wavelength 243.02 nm; this light is used to excite $1S$ Ps to the $2P$ state. Residual light from the Nd:YAG provides ~ 40 mJ of 532 nm (green) light that may be used to photoionize $2P$ Ps. The bandwidth of the UV laser was ~ 150 GHz (FWHM), and the light was propagating in a direction perpendicular to the z axis and linearly polarized along the z axis, which is the magnetic field direction. The 243 and 532 nm beams had diameters of approximately 2 and 6 mm, respectively.

The amount of Ps created was measured by using the technique of single shot positron annihilation lifetime spectroscopy, wherein the output from a fast gamma-ray detector [21] is recorded (in real time) with a digital oscilloscope [22]. Combined with the production of intense positron pulses, this methodology makes it possible to simultaneously observe the annihilation of large numbers ($> 10^6$) of excited state Ps atoms and, in particular, to observe laser-induced changes in the amount of long-lived Ps. An example is shown in Fig. 1(a), where we see a prompt peak around time $t = 0$ followed by an exponential decay due to long-lived (142 ns) triplet ground state Ps; when the laser is tuned to the $1S - 2P$ (Lyman-alpha)

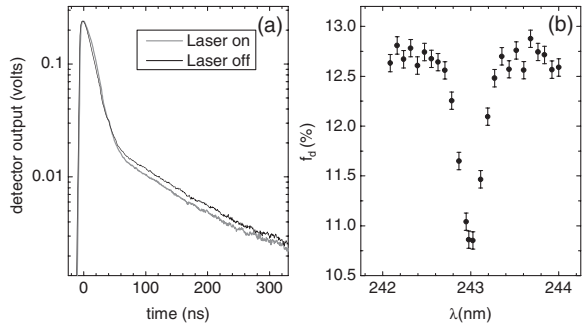

FIG. 1. Single shot lifetime spectra at room temperature and $B = 0.2$ T, with and without firing the UV laser at ~10 ns (a) and changes in the delayed fraction f_d versus the vacuum laser wavelength (b).

wavelength and fired just after the Ps is formed, the intensity of the prompt peak increases, while that of the long-lived Ps component decreases, due to the aforementioned mixing of $2P$ states. We quantify the amount of such Ps by using the parameter f_d, which is defined as the fraction of the lifetime spectrum in the interval 50–300 ns, and is proportional to the total Ps fraction [16]. Changes in the value of f_d as a function of excitation laser wavelength near the Lyman-alpha resonance are shown in Fig. 1(b). The ''flop out'' signal occurs at the Lyman-alpha wavelength (~ 243 nm) because some excited state atoms will decay more rapidly than they would if they had remained in the ground state, and f_d measures the amount of longer lived Ps present.

Magnetic quenching of ground state $m_s = 0$ triplet Ps atoms [1] reduces the observed Ps yield as B is increased. Magnetic quenching refers to the mixing of spin singlet and triplet states by a magnetic field, with a concomitant increase in the Ps decay rate. For ground state Ps, this involves the mixing of $m_s = 0$ singlet ($^1S_0$) and triplet ($^3S_1$) states [1], reducing the lifetime of the latter. This effect may be seen in the ''no laser'' data of Fig. 2(a), which shows the delayed fraction f_d as a function of B as well as a fit to a theoretical quenching curve [23]. Since the annihilation rate of $2P$ atoms is negligible, magnetic quenching is observed only following decay back to the ground state for these atoms.

Also shown in Fig. 2(a) is the variation of f_d with the magnetic field when the laser is fired for low and high UV power, with or without the 532 nm photoionization pulse. In the absence of both photoionization and magnetic quenching, we would expect practically no effect due to the 243 nm resonant UV laser pulse. The low power data (UV only, 0.1 mJ) show the extent to which magnetic quenching occurs; the high power data (UV, 1 mJ + green, 40 mJ) give us a lower limit on the degree of $1S - 2P$ excitation. When the green pulse is used, Ps is detected via the annihilation of liberated positrons that return to the Si target. Without photoionization, the change in the signal with respect to the no laser data is due only to magnetic

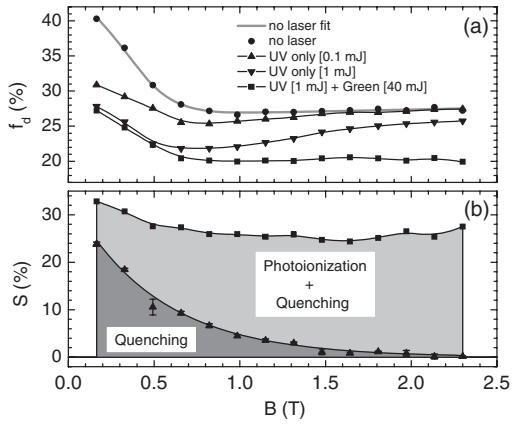

FIG. 2. Delayed fraction (a) and the laser excitation signal (b) as a function of the magnetic field B. The Si sample temperature is 815 K. Lines other than the curves fitted to the no laser measurement in (a) and the ''quenching'' curve in (b) are spline fits. The gradual diminution of the laser signal with higher magnetic fields when exciting by using 0.1 mJ of 243 nm UV light [upright triangles in (b)] shows that triplet-singlet quenching is effectively suppressed in the Paschen-Back regime of high magnetic fields. A constant 4% background has been subtracted from all measurements in (a).

mixing of $2P$ substates. The 1 mJ UV light pulses show a high degree of saturation of the $1S - 2P$ transition, as well as some photoionization of the $2P$ states. For the lower power (0.1 mJ) UV light, there is a negligible amount of ionization, and the signal is entirely due to magnetic quenching. A minimum occurs for both high and low UV power because of the different field dependences of the $1S$ vs the $2P$ state quenching mechanisms.

Owing to the relatively high laser powers used (peak power >0.5 MW cm^{-2} for the 1 mJ pulse), the Rabi period for the $1S - 2P$ transition is much less than the ~3 ns $2P$ lifetime. Thus, even though the efficiency for $2P$ singlet mixing may be less than 30% per excitation and decay cycle, by repeatedly cycling through $1S - 2P$ transitions eventually all excited state atoms may decay through this channel. Similarly, detection via photoionization involves liberated positrons that return to the sample and annihilate; some fraction of these positrons will form more Ps, which would not be detected. However, as this process happens on a subnanosecond time scale, such Ps will likely also be excited and ionized, so that the overall detection efficiency of excited state Ps via photoionization will be of order unity. Thus, the change in the delayed fraction for the 1 mJ UV pulse and for the 1 mJ UV + 40 mJ of green light at low field (where the quenching is maximal) shown in Fig. 2(a) are almost identical.

The efficacy of the laser excitation is indicated in Fig. 2(b), which shows the fractional difference S between the 0.1 mJ UV only and the 1 mJ UV plus 40 mJ green laser configurations and the no laser data represented by the fitted curve in Fig. 2(a) [24]. This parameter is defined as

$S \equiv (f_d[\text{off}] - f_d[\text{on}])/f_d[\text{off}]$, where off and on refer to the laser(s). The UV plus green signal [square symbols in Fig. 2(b)] is due to a combination of photoionization and quenching of $2P$ states and is relatively independent of the magnetic field, whereas the UV only signal [gray triangles in Fig. 2(b)] vanishes above ~ 2 T, demonstrating the absence of $2P$ state quenching at high fields. An exponential curve may be fitted to the UV only data, giving $S \equiv a \exp\{-B/B_0\} + c$, where $a = 0.340 \pm 0.009$ and $B_0 = (0.508 \pm 0.014)$ T, assuming $c = 0$. Including a constant background in the fit yields $c = -0.0018 \pm 0.0019$, changes the other parameters by less than 1 standard deviation, and does not decrease the chi square per degree of freedom. Thus, the data are consistent with the hypothesis that the quenching in Fig. 2(b) strictly vanishes at high magnetic field (i.e., $c = 0$) and that it has been reduced to a value of 0.004 ± 0.002 at $B = 2.3$ T. This exponential decay is an empirical observation for which we do not currently have a theoretical explanation.

It is remarkable that values of S as high as 33% are obtained, given that the FWHM laser bandwidth is only $\sim$10% of the Doppler broadened $1S - 2P$ linewidth. We attribute this to the saturation of the transition, such that even the wings of the laser bandwidth efficiently produce excited states. The fact that the transition is saturated is also evident from the difference in f_d obtained for the two UV laser powers at $B = 0.16$ T, changing only from 33% to 24% when the laser pulse energy is reduced by an order of magnitude. We note that a scheme to produce highly excited (Rydberg) Ps for antihydrogen production [25] calls for short pulses of $\sim$100 K Ps to obtain $\sim$30% efficient Ps excitation. The $n = 2$ Ps production we observe would largely satisfy the criteria for these proposed experiments [26].

We now consider the mechanism by which the quenching effect disappears in high fields. Because of the absence of spin-orbit coupling in the Paschen-Back regime [27], the Ps $n = 2$ states are divided into two separate decoupled classes: (1) eight eigenstates with $m_s = \pm 1$ which have no singlet component and eigenenergies that do not change with B because the orbital and spin magnetic moments of the electron and positron exactly cancel; and (2) eight $m_s = 0$ eigenstates with energy shifted by $\pm 2\mu_B B$, each of which is of necessity 50% singlet. Here m_s is the z projection of the total spin angular momentum. Because of the magnetic quenching of the ground state Ps in high fields, calculated in Fig. 3(a), the experimentally accessible Ps eigenstates are all in the first category [28].

Light traveling in the x direction and polarized with the electric field in the z direction (as used in our experiment) has a transition matrix element that couples only to $m_l = 0$ P states. We denote the Ps states of principal quantum number n by $|n^{2s+1}L_j(m_j)\rangle$, orbital angular momentum states by $|l, m_l\rangle_L$, and spin states by $|s, m_s\rangle_S$. Starting from the triplet $m_s = 1$ state $|1^3S_1(+1)\rangle = |0, 0\rangle_L|1, 1\rangle_S$,

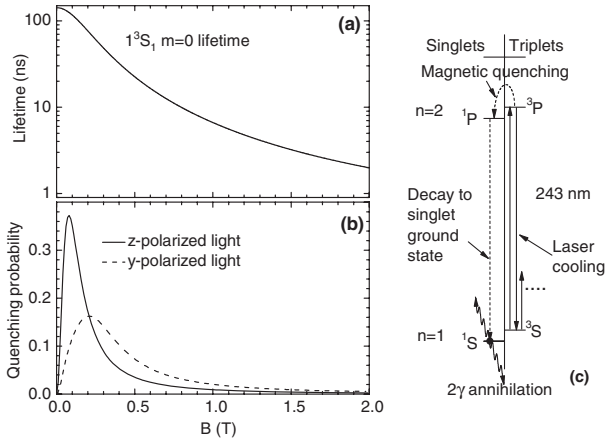

FIG. 3. Calculated triplet $m = 0$ ground state Ps lifetime (a), the quenching probabilities associated with excitation and decay of the $2P$ states (b) as a function of magnetic field strength B, and partial term diagram for the $n = 1$ and $n = 2$ states of positronium (c). The multiple $1^3S - 2^3P$ transitions required for laser cooling indicated in (c) will not be practical if magnetic mixing of the $2P$ states leads to rapid annihilation.

the light couples only to the state $\sqrt{1/2}[|2^3P_2(+1)\rangle + |2^3P_1(+1)\rangle] = |1, 0\rangle_L|1, 1\rangle_S$, which is an eigenstate in the Paschen-Back limit and experiences no quenching because the spin and orbit components of the wave function are by definition decoupled. That is, in this regime excitation to $2P$ states, and subsequent decay back to the $1S$ state, will not affect the spin configuration of the system. If we are not in the high field limit, we must decompose this state into its eigenstate components, which then develop independently. We find the $^1P_1$ amplitude of each of the three eigenstate components and sum the three squared amplitudes to find the quenching probability of initially $m_s = 1$ triplet Ps after a single $1S - 2P$ excitation followed by a return to $1S$, indicated in Fig. 3(b). The field dependence of the overall quenching shown in Fig. 2(b) reflects not only on the single transition probability of Fig. 3(b) but also the dynamics associated with Rabi oscillations. The calculated quenching probability for y-polarized light is also shown in Fig. 3(b) for completeness.

The ability to produce excited state Ps in a high magnetic field without any singlet-triplet mixing is useful for experiments in which atoms may be cycled more than once from the ground state to an excited state, as illustrated in Fig. 3(c). For excitation experiments using high laser powers, multiple Rabi oscillations could lead to an excessive loss rate if quenching interactions are possible. In particular, it is essential that there be no such mixing for laser cooling schemes which necessarily require repeated cycling through the $1S - 2P$ transition [10]. Efficient laser cooling of Ps would mitigate demanding density requirements, which are the main impediment to obtaining a Ps BEC. In principle, Ps may be cooled to the recoil limit of $\sim$0.6 K by laser cooling via the $1S - 2P$ transition [10].

At this temperature the required Ps density would be $\sim 10^{16}$ cm^{-3}, which is close to what we have already achieved [29].

For the production of a Ps BEC, it is envisaged [11] that Ps will be created in a cavity where oppositely polarized (ground state) atoms will destroy each other via spin exchange quenching [29], leaving spin aligned triplet Ps. Laser cooling of these atoms is not as straightforward as would be the case for free Ps in vacuum as confined atoms frequently collide with the cavity walls, leading to a Dicke line narrowing effect [30,31] that is the antithesis of laser cooling. Because of the low mass of Ps, this problem could be overcome by the use of pulsed laser cooling [32], which is related to chirped frequency modulation cooling [3].

In summary, we have performed laser excitation of Ps in the Paschen-Back (strong magnetic field) regime. We find that the mixing between triplet and singlet excited states, that readily occurs at lower magnetic fields (> 0.01 T), does not occur noticeably for fields around 2 T or more. As a result, the production of a dense, spin aligned, triplet Ps gas in a strong magnetic field [29] and laser cooling of that Ps gas are not mutually exclusive, which is advantageous for the production of Bose-Einstein condensed positronium and fundamental Ps spectroscopy.

This work was supported in part by the National Science Foundation under Grant No. PHY 0900919 and by the U.S. Air Force Research Laboratory.

[1] S. Berko and H. N. Pendleton, Annu. Rev. Nucl. Part. Sci. **30**, 543 (1980); A. Rich, Rev. Mod. Phys. **53**, 127 (1981).
[2] See, e.g., S. G. Karshenboim, Phys. Rep. **422**, 1 (2005).
[3] M. S. Fee *et al.*, Phys. Rev. Lett. **70**, 1397 (1993); S. Chu, A. P. Mills, and J. L. Hall, Phys. Rev. Lett. **52**, 1689 (1984).
[4] Y. Kataoka *et al.*, Phys. Lett. B **671**, 219 (2009).
[5] A. P. Mills, Jr. and G. H. Bearman, Phys. Rev. Lett. **34**, 246 (1975); M. W. Ritter *et al.*, Phys. Rev. A **30**, 1331 (1984).
[6] A. P. Mills, Jr., S. Berko, and K. F. Canter, Phys. Rev. Lett. **34**, 1541 (1975); S. Hatamian, R. S. Conti, and A. Rich, Phys. Rev. Lett. **58**, 1833 (1987); D. Hagena *et al.*, Phys. Rev. Lett. **71**, 2887 (1993).
[7] A. P. Mills, Jr., Phys. Rev. Lett. **46**, 717 (1981); F. Fleischer *et al.*, Phys. Rev. Lett. **96**, 063401 (2006); Y. Nagashima *et al.*, New J. Phys. **10**, 123029 (2008); K. Michishio *et al.*, Phys. Rev. Lett. **106**, 153401 (2011).
[8] D. M. Schrader *et al.*, Phys. Rev. Lett. **69**, 57 (1992).
[9] D. B. Cassidy and A. P. Mills, Jr., Nature (London) **449**, 195 (2007).
[10] E. P. Liang and C. D. Dermer, Opt. Commun. **65**, 419 (1988).
[11] P. M. Platzman and A. P. Mills, Jr., Phys. Rev. B **49**, 454 (1994).
[12] E.g., C. J. Pethick and H. Smith, *Bose-Einstein Condensation in Dilute Gases* (Cambridge University Press, Cambridge, England, 2002).
[13] S. M. Curry, Phys. Rev. A **7**, 447 (1973); M. L. Lewis and V. W. Hughes, Phys. Rev. A **8**, 625 (1973).
[14] C. D. Dermer and J. C. Weisheit, Phys. Rev. A **40**, 5526 (1989).
[15] K. P. Ziock *et al.*, J. Phys. B **23**, 329 (1990); K. P. Ziock *et al.*, Phys. Rev. Lett. **64**, 2366 (1990).
[16] D. B. Cassidy *et al.*, Phys. Rev. A **81**, 012715 (2010).
[17] D. B. Cassidy *et al.*, Rev. Sci. Instrum. **77**, 073106 (2006).
[18] C. M. Surko and R. G. Greaves, Phys. Plasmas **11**, 2333 (2004).
[19] D. B. Cassidy *et al.*, Phys. Rev. Lett. **106**, 133401 (2011).
[20] D. B. Cassidy *et al.*, Phys. Rev. A **82**, 052511 (2010).
[21] D. B. Cassidy and A. P. Mills, Jr., Nucl. Instrum. Methods Phys. Res., Sect. A **580**, 1338 (2007).
[22] D. B. Cassidy *et al.*, Appl. Phys. Lett. **88**, 194105 (2006).
[23] A. Rich, Ph.D. dissertation, University of Michigan, 1965 (University Microfilms, Ann Arbor, MI). We have included in the fitted curve for f_d a term linear in B to account for a slight density dependence of the Ps yield from p-Si(100).
[24] Since the fitted curve is a good representation of the ''laser-off'' data, we use it in the calculation of S so that the point-to-point statistical fluctuations in Fig. 2(b) are predominately due to the ''laser-on'' data, thus yielding an improved estimate of the asymptotic behavior of S.
[25] F. Castelli *et al.*, Phys. Rev. A **78**, 052512 (2008).
[26] A. Kellerbauer *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **266**, 351 (2008).
[27] F. Paschen and E. Back, Physica **1**, 261 (1921); E. U. Condon and G. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, Cambridge, England, 1935), ch. 16.
[28] We ignore transitions from the triplet $m_s = 0$ state because these become unimportant for fields greater than about 0.3 T for our detection scheme that counts only decays 50 ns after the formation of the positronium.
[29] D. B. Cassidy, V. E. Meligne, and A. P. Mills, Jr., Phys. Rev. Lett. **104**, 173401 (2010).
[30] R. H. Dicke, Phys. Rev. **89**, 472 (1953).
[31] D. B. Cassidy *et al.*, Phys. Rev. Lett. **106**, 023401 (2011).
[32] K. Molmer, Phys. Rev. Lett. **66**, 2301 (1991).