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# Positronium Hyperfine Interval Measured via Saturated Absorption Spectroscopy

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We report Doppler-free measurements of the positronium (Ps) Lyman- $\alpha$  transition using saturated absorption spectroscopy. In addition to a Lamb dip at wavelength  $\lambda_L = 243.0218 \pm 0.0005$  nm, we also observed a crossover resonance at  $\lambda_C = 243.0035 \pm 0.0005$  nm, arising from the excitation of  $1^3S_1$  atoms to Zeeman mixed 2P states, followed by stimulated emission to the  $1^1S_0$  ground state. Since  $(\lambda_L - \lambda_C)$  is related to the Ps hyperfine interval,  $E_{\text{hfs}}$ , this observation constitutes the first optical measurement of this quantity and yields  $E_{\text{hfs}} = 198.4 \pm 4.2$  GHz. We describe improvements to the methodology that could lead to the  $\sim$  ppm level of precision required to address the long standing discrepancy between QED calculations and precision experiments using microwave radiation to induce transitions between Zeeman shifted triplet Ps states.

In order to perform high resolution optical spectroscopy of low mass atoms or molecules it is essential to eliminate or significantly reduce Doppler broadening effects. This is particularly important for positronium (Ps) [1], the lightest atomic system known, for which the  $\sim 50$  MHz natural linewidth of the 1S-2P ( $\lambda = 243$  nm) transition will typically be Doppler broadened to  $\sim 500$  GHz or more [2]. One may use Doppler free two-photon spectroscopy to accomplish this, but only for transitions between states with orbital angular momenta differing by 0 or  $2\hbar$ , such as 1S-2S [3]. An alternative is saturated absorption spectroscopy (SAS) in which velocity selective excitation allows narrow lines to be resolved [4, 5].

In this letter we describe SAS measurements of the 1S-2P (Lyman- $\alpha$ ) transition of Ps. In addition to the expected Lamb dip [6] we also observed a crossover resonance due to Zeeman mixing of singlet and triplet states in the 2P manifold. The separation between the Lamb dip and the hyperfine crossover peak corresponds to an energy difference of half the Ps hyperfine interval,  $E_{\text{hfs}}$  (where  $h\nu_{\text{hfs}} = 203$  GHz). Our observation therefore constitutes the first *optical* measurement of  $E_{\text{hfs}}$  which could potentially be performed with high precision, and help to resolve the current discrepancy between theory [7] and experiment [8]. Being composed only of leptons, the Ps system is well described by Quantum Electrodynamics (QED) [9], which makes the existence of such a discrepancy all the more surprising.

The experiments were conducted using an accumulator [10] that produces intense sub ns positron pulses [11]. These were implanted into an n-doped Si(100) target kept at  $\sim 900$  K, resulting in the efficient production of Ps atoms in vacuum [12]. The laser systems used in this work are essentially the same as those described in ref [13]. This includes a Nd-YAG pumped dye laser providing up to  $\sim 1$  mJ of ultraviolet (uv) light pulses around 5 ns wide, with a spot size of approximately  $0.04 \pm 0.01$  cm<sup>2</sup>. Around 30 mJ of residual 532 nm (green) light is also available from the pump laser. In order

to eliminate a possible source of systematic wavelength measurement error the 486 nm light transmitted to the wavemeter was sampled before the doubling crystal. In the present experiment the excitation laser is retro-reflected back into the chamber, so that the positronium cloud in front of the Si target is illuminated by two counter-propagating beams at the same frequency. The reflected beam has a slightly longer path length, and is delayed by  $\sim 2$  ns with respect to the primary beam. A PbWO<sub>4</sub> scintillator is used to measure Ps annihilation radiation in real time, and thus to generate single shot lifetime spectra [14]. The fraction of such spectra integrated from 50-300 ns,  $f_d$ , is related to the amount of long lived Ps present [13], and we characterize the effect of laser irradiation using the parameter  $S = [f_d(\text{off}) - f_d(\text{on})] / f_d(\text{off})$ , which gives the fractional change in  $f_d$  due to irradiation (off and on refer to the uv laser). Photoionization and processes that convert long-lived triplet states to short-lived singlet states will change the Ps decay rate, and hence also S.

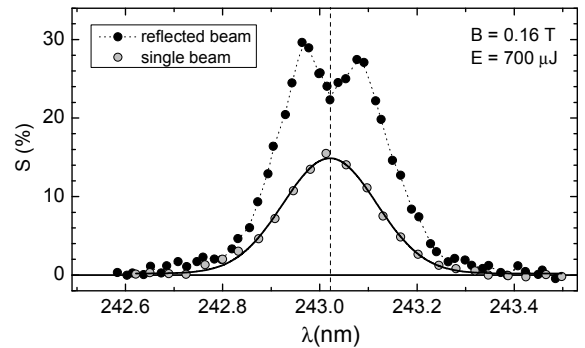


FIG. 1. Lineshapes measured with a laser bandwidth of  $\sim 100$  GHz with and without the retro-reflected beam. The solid black line is a Gaussian fit that yields a centroid of  $243.0208 \pm 0.0016$  nm and a FWHM of  $0.226 \pm 0.005$  nm. The dotted line in the reflected beam data is a simple spline fit. The dashed vertical line is at  $243.0218$  nm, (to be compared to measurements from FIG. 3).

Initial measurements were made using a  $\sim 100$  GHz wide laser, as shown in Fig 1. For a single beam (grey filled circles) we observe a Gaussian line profile with a  $\sim 1.1$  THz Doppler broadened 1S-2P linewidth (FWHM). When the laser is reflected and tuned off resonance by more than the laser linewidth (black filled circles), the two beams address separate parts of the Ps velocity distribution, but are still close to the resonance frequency. When the laser is tuned near to the resonance wavelength a Lamb dip is apparent because the same part of the Doppler spectrum cannot be excited by both the incident and reflected laser beams owing to saturation. The  $\sim 250$  GHz width of the Lamb dip is consistent with the power broadening observed in previous measurements [2].

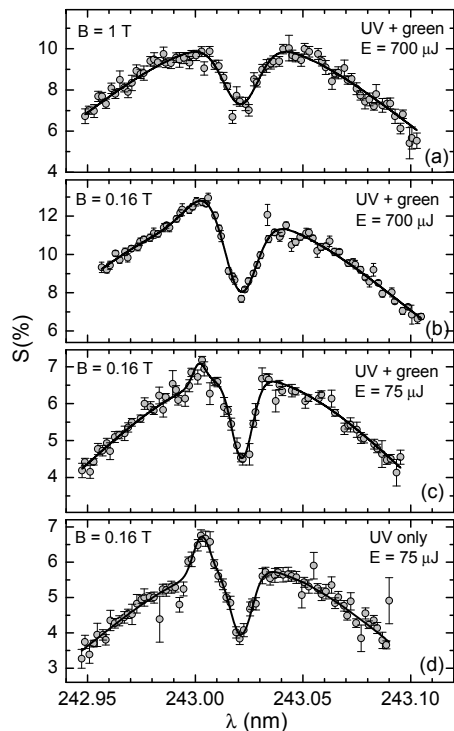


FIG. 2. Line shapes measured at high and low magnetic fields and uv laser pulse energies, and with and without green light, as indicated in the panels. The solid lines are 3-Gaussian fits to the data, and the laser bandwidth is  $\sim 15$  GHz.

In order to make more accurate Doppler-free measurements the laser bandwidth was narrowed, as shown in Fig 2, which shows a Lamb dip at  $\lambda_L \sim 243.02$  nm for various laser and magnetic field configurations. These data were taken with a  $\sim 15$  GHz wide laser, which reduces the fraction of Ps atoms excited (and hence also  $S$ ). Also apparent in Figs 2(b-d) is a second peak at  $\sim 243.003$  nm. This is a crossover peak [4] that is due to resonant excitation of triplet 1S atoms to Zeeman mixed 2P states that are subsequently driven to the *singlet* ground state via stimulated emission, and is the basis for our

measurement of the Ps hyperfine interval [15]. For an atom at rest the excitation frequency from the  $1^3S_1$  state to a 2P state will not be the same as the frequency needed to stimulate emission to the  $1^1S_0$  level. This difference is due to the different energies of the two 1S states, *i.e.*, the Ps hyperfine interval. However, with counter propagating lasers some atoms have velocities such that one beam is red shifted into resonance for excitation, and the other is blue shifted into resonance for stimulated emission.

The existence of the hyperfine crossover peak can be understood by considering the effect of a magnetic field on Ps atoms: Zeeman mixing connects singlet and triplet states, so that when  $n = 2$  atoms are produced in a magnetic field they are superpositions of mixed singlet-triplet 2P states [16]. A consequence of such mixing is that the singlet and triplet ground states are indirectly coupled via the intermediate singlet-triplet 2P superpositions. The spontaneous decay of mixed 2P atoms to the singlet ground state can be used as a method to detect Ps excitation [17, 18]. It is known, however, that Zeeman mixing in this system is suppressed in high magnetic fields [19], so we can immediately explain the absence of a crossover peak in Fig 2a, where the data were taken in a magnetic field  $B = 1$  T. The crossover peak is present in Fig 2b, for which the magnetic field is 0.16 T. A 1T field will not completely suppress Zeeman mixing between 2P states [19], but will reduce the rate so that photoionization by the green light pulse becomes dominant and the crossover peak amplitude will then be negligible.

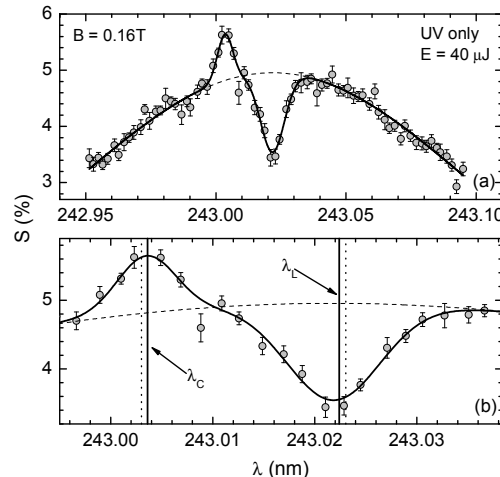


FIG. 3. Line shape measured at 0.16T with a 15 GHz laser bandwidth and 40  $\mu$ J of uv (a) and expanded section (b) showing the crossover ( $\lambda_C$ ) and Lamb dip ( $\lambda_L$ ) wavelengths with (solid vertical lines) and without (dotted vertical lines) the recoil shift. The solid black curve is a three Gaussian fit to the data and the dashed curve is the Gaussian corresponding to the Doppler broadened line only. The data and error bars are determined from a set of 17 wavelength scans of 104 shots each, using the un-weighted average of the  $S$  and  $\lambda$  data over bins of width 0.002 nm.

The width of the Lamb dips in Figs 2a and 2b is partly due to power broadening, as is evidenced by the narrower peak

seen in Fig 2c, for which data the laser pulse energy has been reduced by almost an order of magnitude. Reducing the power also results in a narrower crossover peak, with a lower amplitude, because the rate for stimulated emission is less than that of photoionization from the green light. This fact is illustrated by comparing Figs 2c and 2d. In the latter, the green light is not present and the observed signal is due only to magnetic quenching. The crossover peak is much larger in Fig 2d than it is in Fig 2c for this reason.

Our most accurate line center measurements were obtained using  $\sim 40 \mu\text{J}$  pulses of uv light, and are shown in Fig 3. For convenience we assume that there is only one intermediate (Zeeman mixed) 2P state, which we refer to as the  $2P_X$  level. Actually several 2P states will be populated, but since they cannot be resolved in the present experiment we use the  $2P_X$  level to approximate the average energy difference between the  $1^3S_1$  and all 2P excited states with a singlet component. Weighting the perturbed 2P energies for  $B=0.16 \text{ T}$  by the square of the amplitudes connecting the initial  $1^3S_1(m)$  levels to the singlet ground state for light linearly polarized in the  $z$ -direction (along the magnetic field and perpendicular to the sample) we obtain a weighted mean  $2P_X$  energy relative to the unperturbed  $2^3S_1$  level of  $-9.3 \text{ GHz}$  for  $m=0$  and  $-10.9 \text{ GHz}$  for  $m=\pm 1$ , with the average  $2P_X$  level being  $-10.4 \text{ GHz}$  [20].

The Lamb dip arises through saturated excitation of zero velocity atoms, whereas the crossover resonance occurs for atoms with a velocity such that transitions occur at frequencies corresponding to excitation from the triplet ground state to the  $2P_X$  level, and to stimulated de-excitation back to the singlet ground state for different beam directions. This two-step process will lead to an increase in the Ps decay rate that we observe as the crossover peak, centered at frequency  $\nu_C$ . The transitions in the lab frame are  $1^3S_1 + h\nu_C \rightarrow 2P_X$ , followed by  $2P_X + h\nu_C \rightarrow 1^1S_0 + 2h\nu_0$ . Because Ps is so much lighter than other atomic systems, recoil effects become important even at relatively low levels of precision. From conservation of (non-relativistic) energy and momentum, the Lamb dip resonant frequency  $\nu_L$  will be

$$\nu_L = \nu_X (1 + \frac{1}{2}R), \quad (1)$$

where  $R \approx h\nu_X / m_{Ps}c^2$ . Ordinarily there would be recoil components shifted up and down, corresponding to absorption and stimulated emission respectively [21]. However, in the present experiment the latter will not result in an observable signal since stimulated atoms are returned to the triplet ground state. Depletion of the excited state population in this way could affect the observed lineshape, but the stimulated component will be diminished by the multimode and spatial structure of the laser pulses leading to an imperfect standing wave, as well as the  $\sim 2 \text{ ns}$  time delay between the beams.

The hyperfine crossover peak resonant frequency  $\nu_C$  is

$$\nu_C = (\nu_X + \frac{1}{2}\nu_{hfs})(1 - \frac{1}{2}R), \quad (2)$$

where  $E_X = h\nu_X$  is the energy difference between the  $1^3S_1$  and the  $2P_X$  level,  $m_{Ps} = 2m_e$  is the Ps mass,  $c$  is the speed of light, and we are neglecting the  $\pm 0.1 \text{ GHz}$  Zeeman shifts of  $n=1$  states in the  $0.16 \text{ T}$  magnetic field. The energy difference between the  $1^1S_0$  and the  $1^3S_1$  states is  $E_{hfs} = h\nu_{hfs}$ , with the hyperfine frequency from Eqs. 1 and 2 given by

$$\nu_{hfs} = 2(\nu_C - \nu_L) + (\nu_C + \nu_L)R, \quad (3)$$

where  $(\nu_C + \nu_L)R = 12.3 \text{ GHz}$  is the total recoil shift. The predicted line center wavelengths,  $\lambda_L = 243.02240 \text{ nm}$  and  $\lambda_C = 243.00361 \text{ nm}$ , are shown as solid vertical lines in Fig 3b, along with the wavelengths excluding recoil (dotted lines). The measured wavelengths, obtained from the Gaussian fits to the data of Fig 3, are  $\lambda_L = 243.02183 \pm 0.00027 \text{ nm}$  and  $\lambda_C = 243.00351 \pm 0.00031 \text{ nm}$ , from which we obtain  $\nu_{hfs} = 198.4 \pm 4.2 \text{ GHz}$ , which is  $\sim 1\sigma$  less than the known value ( $\sim 203.4 \text{ GHz}$  [7, 8]). A systematic wavelength measurement uncertainty of about  $\pm 0.0004 \text{ nm}$  ( $\pm 2 \text{ GHz}$ ) should be applied to the  $\lambda_L$  and  $\lambda_C$  measurements, but does not affect our determination of  $\nu_{hfs}$  since it is derived from the difference in the two wavelengths. With a higher resolution measurement it will be possible to distinguish between the individual 2P states, which would result in a series of lines, with a corresponding crossover resonance for each distinct 2P level coupling singlet and triplet ground states [22]. The effects of stimulated emission on the Lamb dip lineshape may also be observable with an improved resolution.

Quantum Electrodynamics [23] is one of the most successful theories in physics insofar as its predictions have been tested to astounding accuracy, and have (almost) always been found to agree with measurements [24]. There are, however, a few rare cases in which experiments appear to disagree with QED predictions; namely the proton radius as measured in muonic hydrogen ( $\sim 5\sigma$  discrepancy) [25], the muon (g-2) magnetic moment ( $\sim 3\sigma$  discrepancy) [26] and the positronium hyperfine interval ( $\sim 4\sigma$  discrepancy). The last has been calculated to  $\sim 1$  part per million (ppm) [7] while measurements have a combined precision of  $3.4 \text{ ppm}$  [8]. Such discrepancies should not simply be overlooked, or dismissed as flawed experiments, as they might provide some (much needed) guidance towards new physics, just as deviations of the electron g factor from the Dirac value of exactly 2 [27] and the hydrogen Lamb shift [28] were important in the early development of QED.

Precision Ps hyperfine experiments [8] have been carried out using the method first developed by Deutsch in 1952 [29] which measures  $E_{hfs}$  via transitions between  $|m|=1$  and  $m=0$  triplet states of positronium produced in a gas from a  $\beta^+$  emitter. This method was used because of the difficulty in producing the  $203 \text{ GHz}$  radiation needed to make a direct transition from triplet to singlet states [30]. One possible source of the current discrepancy is that a non-linear dependence of the Ps thermalization rate with gas pressure may lead to an incorrect determination of the pressure shift.

By measuring the Ps energy and lifetime simultaneously, thermalization effects can be eliminated [31]. Experiments of this type are currently underway, but have not yet reached a sufficient level of precision to address the discrepancy [32].

It is possible that a common systematic error is present in the previous measurements of  $E_{\text{hfs}}$ , but there have been suggestions that positronium might provide a window into physics beyond the standard model through various mechanisms related to mirror particles [33]. The existence of such particles could manifest themselves through forbidden decay modes, although no evidence for any such phenomena has been observed so far [34]. The Ps hyperfine interval may also be measured via modulations in the angular distribution of annihilation gamma radiation arising from quantum interference between singlet and triplet states [35]. Such measurements have been made [36] but, again, have not yet reached a sufficient level of precision to address the discrepancy. Clearly, the present optical measurements are also very far from the required level of precision. However, since optical metrology techniques are highly developed, there is a great deal of scope for increasing the accuracy of our measurement.

We now consider some of the improvements that could make it possible to perform a  $\sim$  ppm optical measurement of the Ps hyperfine interval. The present result has a statistical error in  $E_{\text{hfs}}$  of about  $\pm 2\%$  which could have been attained in about 0.5 days of continuous data taking. Using cold Ps at 50K [37] would increase the signal amplitude by a factor of 5, while using a 0.5 GHz wide cw diode-seeded laser to resolve the  $\sim 1$  GHz natural linewidth (arising from the 1/8 ns singlet ground state lifetime) would lower the signal amplitude by a factor of 100, for a net signal amplitude 1/20 of the present size. The solid angle of the detector used here is  $\sim 5\%$  but with a modified chamber could be increased to provide nearly  $2\pi$  coverage. This, along with an improved time resolution ( $\sim 5$  rather than 20 ns [38]) would bring us to about the same signal to noise ratio as at present, but with a linewidth 50 times narrower. The line centers could then be determined about 10 times faster by only scanning only over the narrow peaks. The net statistical precision in 0.05 days would then be  $2\%/50=400$  ppm, and would amount to about  $\pm 10$  ppm in 100 days. Running the experiment at a facility delivering  $10^9$  positrons per second [39] compared to our present  $5 \times 10^6 \text{ s}^{-1}$  would give us a final precision of the order of 2 ppm in a few months.

To summarize, we have performed saturated absorption spectroscopy of positronium for the first time. This technique is well suited to Ps because it is so light and is usually produced with velocities of  $\sim 10^{-4}c$  or more, making Doppler broadening the dominant contribution to measured linewidths. Using SAS it should be possible to make precision optical measurements of atomic (and molecular [40]) Ps energy levels. We also presented a new way to measure the positronium hyperfine interval via a crossover resonance arising from coupling of the ground state singlet and triplet levels via Zeeman mixed 2P states. We expect that this proof-

of-principal measurement can be sufficiently improved so as to provide a  $\sim$  ppm measurement, and thereby address the current  $\sim 4\sigma$  disagreement between Ps hyperfine structure measurements and QED theory; the possibility that this discrepancy might be due to new physics demands further investigation.

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