Measurement of the Positronium $1\,^3S_1$-$2\,^3S_1$ Interval by Continuous-Wave Two-Photon Excitation

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Using continuous-wave excitation to eliminate the problems inherent with pulsed laser measurements of nonlinear transitions, we have measured the 1^3S_1-2^3S_1 interval in positronium (Ps) to be 1 233 607 216.4 ± 3.2 MHz. The quoted 2.6 ppb (parts per 10^9) uncertainty is primarily due to the determination of the Ps resonance relative to the Te_2 reference line, with a 1.5 ppb contribution from a recent calibration of the Te_2 line relative to the hydrogen $1S$-$2S$ transition. The uncertainty corresponds to 3.5×10^{-5} of the $\alpha^2 R_\infty$ QED contribution to the 1^3S_1-2^3S_1 interval. Our measurement is sufficiently accurate to provide a test of the as yet uncalculated $\alpha^4 R_\infty$ QED corrections.

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Precision measurements of the structure of purely leptonic atoms such as positronium (e^+-e^-) and muonium (μ^+-e^-) provide a unique opportunity to further our understanding of the two-body formulation of quantum electrodynamics [1]. The positronium (Ps) atom is particularly sensitive to recoil and annihilation effects since it is composed of a particle-antiparticle pair. The theoretical computation of the positronium energy levels now includes $\alpha^4 \ln\alpha R_\infty$ terms [2-6] and work is in progress on the complete corrections of order $\alpha^4 R_\infty c = 9.3$ MHz [5]. The triplet $1S$-$2S$ interval is an experimentally attractive candidate for testing the theory because of its large QED corrections and a natural linewidth of only 1.3 MHz. Previously, the interval was known with a precision of 10.7 MHz, from the 1984 measurement [7] adjusted for a recalibration of a Te_2 reference line [8] and a correction of the frequency offset of the pulsed laser [9].

The highly forbidden $1S$-$2S$ transition may be driven by counterpropagating beams tuned to half the $1S$-$2S$ interval [10]. While free of first-order Doppler shifts, two-photon excitation requires a high intensity that may be accompanied by nonlinear effects due to frequency chirps if a pulsed laser is used [11]. The availability of high-intensity pulsed Ps sources [12] and high-finesse Fabry-Pérot cavities in which the circulating power can exceed 1 kW [13] makes it possible to excite the $1S$-$2S$ transition in Ps with a continuous-wave laser source [14]. In this Letter, we report a new measurement of the 1^3S_1-2^3S_1 interval using Doppler-free cw excitation to obtain a factor of 3 improvement in the experimental accuracy.

Positrons are generated by bremsstrahlung pair production at the beam dump of an electron microtron accelerator. After moderation [15,16], they are bunched into 25-ns pulses at 30 Hz, each containing 2×10^4 slow e^+. The positrons are guided by a 150-G axial magnetic field to an Al(111) sample shown in Fig. 1 where they are implanted with 1-2 keV kinetic energy. The sample was kept at 576 ± 5 K to thermally desorb the surface state positrons as free thermal Ps in vacuum [15,16]. About 10% of the incident positrons are reemitted from the surface with ~0.1 eV energies and about 30% leave the surface as thermal positronium with a beam Maxwellian velocity distribution [17].

A small fraction of positronium atoms desorbed from the Al sample pass through the cw excitation beam and are excited to the $2S$ state, from which they are photoionized after a known delay by a pulsed, doubled Nd:YAG laser at the 532-nm beam (30 mJ per 7-ns pulse) in a multipass arrangement. As shown in Fig. 1, the liberated positrons are accelerated to a channel electron multiplier array (CEMA) detector by a ~2 V/cm axial electric field. Because of the small excitation probability of Ps in the cw beam, the reemitted positrons constitute an overwhelming background if allowed to contaminate the photoionized e^+ signal. Thus, signal positrons are collected only from the region below the target while the reemitted positrons are trapped along magnetic field lines and cannot be accelerated to the detector.

A Coherent 699-21 actively stabilized, continuous-wave ring dye laser serves as the source of single frequency 486-nm radiation to drive the high-finesse buildup cavity. The piezoelectrically tunable cavity is located inside the experimental vacuum chamber and is constructed with laser gyroscope quality mirrors in a near-confocal geometry. With a cavity finesse of 10^5, we achieve a factor of 7.6×10^3 buildup. A maximum of 2.5 kW circulating power and a peak intensity of 1.7 MW/cm^2 is obtained before thermal effects distort the TEM_{00} mode. By means of a low-loss phase modulator inserted in the dye laser, we lock the laser to the 1.6 kHz resonance of the buildup cavity using a 1-MHz bandwidth servo loop [18]. The error signal is derived from light reflected from

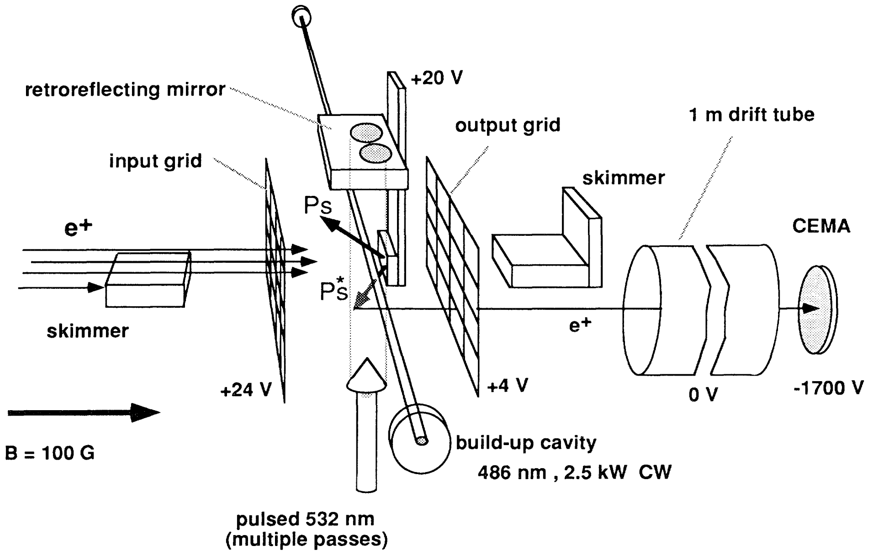

FIG. 1. Positronium source and laser interaction region. Incident positrons are thermally ejected from an Al target as ground state (1^3S_1) positronium atoms. Some of these atoms pass through a 2.5-kW cw laser beam formed inside a Fabry-Pérot resonator, and are excited to the $2S$ state. Excited-state atoms cross magnetic field lines to a region below and to the left of the positronium source, where they are photoionized by a pulsed laser at 532 nm. Liberated positrons are accelerated along the magnetic field lines, below the Al target to the detector. The two skimmers and an aperture (not shown) just before the CEMA reduce the number of background positrons reaching the detector. The two spots on the retroreflecting mirror are caused by the multiple passes of the 532-nm laser beam.

the buildup cavity using a sideband locking scheme [19].

A Te_2 absorption line (e_3) was used as the frequency reference for our measurement [20]. Our Te_2 cell was calibrated to the hydrogen $1S$-$2S$ transition frequency eight months after our Ps measurement, and found to be at 616 803 544.4 ± 0.60 MHz. The quoted uncertainty reflects the 250-kHz uncertainty in the calibration and 500-kHz constraint on pressure shift changes between our Ps measurement and the calibration based on the measured linewidth of the e_3 line.

The laser frequency was measured as a radio frequency offset relative to the zero crossing of the e_3 line center of a second, frequency modulated Te_2 spectrometer, and scanned in 5-MHz steps. The transmitted laser power through the buildup cavity was continuously monitored by a photodiode and calibrated to reflect the absolute power in the buildup cavity.

In Fig. 2 we show the Ps resonance taken at four different YAG pulse delays which correspond to Ps velocity groups having energies from 2 to 15 meV. The major systematic effects in determining the positronium $1S$-$2S$ line center relative to the Te_2 reference line are the second-order Doppler shift due to the motion of the Ps atoms in the laser reference frame and the ac Stark effect due to the nearly saturating cw laser beam. For the majority of the data taken, both systematic corrections are less than 20 MHz. Scans were taken at several time delays of the photoionization pulse and at several settings of the buildup cavity power to facilitate eliminating these systematic effects.

The precision to which we can determine the Ps line center is limited by both counting statistics and our understanding of the line shape. A computer model of our experimental geometry allows us to integrate the Schrödinger equation for a large number of positronium trajectories chosen randomly from a beam Maxwellian distribution at the temperature of the Al sample. The calculation includes the ac Stark shift, ground-state annihilation, and 486-nm photoionization. Calculated line shapes at each YAG delay are multiplied by a signal amplitude parameter, and shifted to a frequency relative to the measured Te_2 reference line. The value of the amplitude parameter is shown in Fig. 3(a) to be independent of the Ps energy, thus supporting our use of a beam Maxwellian velocity distribution. The error bars are from a Monte Carlo calculation in which the model was fitted to 2000 randomly generated data sets to determine the variance of the fit amplitude and line center for each resonance.

Figure 3(b) shows that the fit frequencies as a function of Ps energy have a small residual slope 1.13 standard deviations above zero. To account for possible overcompensation of the second-order Doppler shift, we extrapolate linearly to zero Ps energy to find the Ps resonance frequency. We note that fit line centers are also perfectly consistent with their weighted mean, yielding a reduced chi-square parameter of $\chi^2=0.5$, indicating that the scatter in the line centers is actually $\sqrt{2}$ smaller than ex-

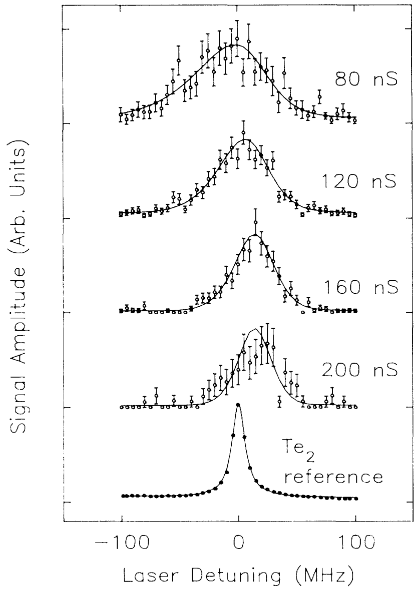

FIG. 2. Fit of the model line shape to five Ps resonances taken at different YAG photoionization pulse delays. The data in this figure were taken at a target temperature of 600 K and a cw laser power between 2 and 2.5 kW. The calculated line shape at each YAG delay is fitted with only an amplitude parameter and a frequency offset.

pected based on our Monte Carlo statistical error estimate. The extrapolation is a more conservative approach, giving a statistical uncertainty of 0.90 MHz, compared to a 0.46 MHz uncertainty in the weighted mean.

The uncertainty in the determination of the Ps transition frequency due to uncertainty in the model parameters is determined by varying each parameter and noting its effect on the extrapolated frequency. The values of the fit frequency for the 13 resonances are shown in Fig. 3(c) as a function of circulating laser power. The slope of the data is consistent with zero because the model has correctly accounted for the ac Stark effect over a factor of 3 variation in laser intensity. The uncertainty in our laser power measurements contributes less to the uncertainty in the slope of these data than the statistical errors. It is therefore not necessary to extrapolate these data to zero laser intensity in the determination of the Ps line center.

The reduction of our measurements to an absolute value for the 1^3S_1-2^3S_1 transition frequency is detailed in Table I. An acousto-optic frequency shift produces a +55 MHz offset in the observed position of the Te_2 reference line. Contributions to the ±1.5-MHz uncertainty in the extrapolated Ps line center are the statistical uncertainty and the uncertainty of the second-order Doppler and ac Stark shift corrections. The dimensions of the YAG beam affect the velocity distribution of the sampled atoms, and therefore the size of the second-order Doppler shift correction. The cw beam diameter and laser power calibration determine the cw laser intensity and thus the ac Stark shift correction. All contributions to the final uncertainty are assumed to be uncorrelated and are added in quadrature. The motional Stark shift in the 150-G magnetic field is less than 1 MHz and is eliminated by our extrapolation procedure. The calculated Zeeman shift of the 1^3S-2^3S ($m=0\rightarrow 0$) transition produces a +1.14-MHz shift in the observed Ps resonance when averaged over the three magnetic sublevels. With these corrections, the absolute transition frequency is 616803608.2 ± 1.6 MHz, and the full 1^3S_1-2^3S_1 interval in positronium is 1233607216.4 ± 3.2 MHz.

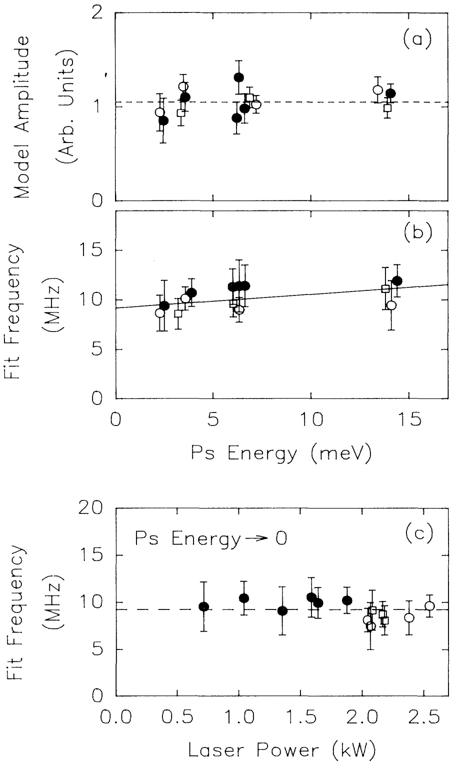

FIG. 3. (a) Signal amplitude parameter and (b) fit line center relative to the Te_2 reference line, for fits to 13 experimental resonances, as a function of positronium energy for a set of model parameters representative of the experimental geometry. (c) Fit line center as a function of the measured circulating power in the excitation beam. The points are extrapolated to zero Ps energy using the slope found in (b). The filled circles, open circles, and squares correspond to three separate runs.

The details of this work will appear elsewhere [21]. We are in agreement with two recent calculations of the $\alpha^4 R_\infty \ln\alpha$ QED corrections by Fell [5] and Khriplovich, Milstein, and Yelkhovsky [6]. The precision of the measurement reported in this Letter is sufficient to provide a test of the as yet uncalculated $\alpha^4 R_\infty$ QED corrections to both the excited and ground states. The measurement

TABLE I. Summary of uncertainties and results.

Te_2 reference line	616 803 544.4 ± 0.6 MHz
AOM offset	55.0 ± 0.0
Model fit to line center	10.0 ± 1.5
statistical error ± 0.9 MHz	
YAG parameters ± 1.0	
cw beam parameters ± 0.6	
dc Stark shift	−0.07 ± 0.1
Quadratic Zeeman shift	−1.14 ± 0.0
1*S*-2*S* transition frequency	616 803 608.2 ± 1.6 MHz
1*S*-2*S* interval	1 233 607 216.4 ± 3.2
Previous measurement	
Chu *et al.* [7]; Danzmann *et al.* [9]	1 233 607 218.9 ± 10.7
Theory	
Fell [5]	1 233 607 221.7 ± $O(10)$

provides a test of the $\alpha^2 R_\infty$ QED contribution to the triplet 1*S*-2*S* interval to 3.5 parts in 10^5. If the agreement between our experimental result and theory persists with the $\alpha^4 R_\infty$ QED corrections, the positron-electron mass difference [7,22] will be constrained to $|m_{e^+} - m_{e^-}| < 2\times 10^{-9} m_{e^-}$ assuming equal charges, the best limit on a particle-antiparticle mass ratio apart from the $K^0-\bar{K}^0$ system.

Further improvements over the present measurement are clearly possible. Cold Ps would greatly reduce the second-order Doppler shift and the transit time broadening of the observed resonances. The increased interaction time would permit a lower laser intensity and a smaller ac Stark shift. Laser cooled Ps would permit a measurement of the 1*S*-2*S* transition to reach a precision significantly better than the 1.3 MHz natural linewidth.

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