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Sensitive detection of Doppler-free two-photon-excited 2S positronium by spatially separated photoionization

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Using a spatially filtered pulsed laser at 486 nm we have photoexcited positronium to the 2S state and detected it via a delayed and spatially separated 532-nm photoionization pulse. We have extended our observations down to low 486-nm power to explore the possibility of a cw measurements of the positronium 1S-2S interval. A model calculation taking into account the velocity distribution of the 235-K positronium from an oxygenated cold Al(111) target, the parameters of the laser beams, and the nonlinear excitation probability explains our data. The same model indicates that a cw measurement is presently feasible.

Positronium (Ps) is a purely leptonic atom that should be understandable in terms of the two-body formulation of quantum electrodynamics [1]. In particular, QED level shifts of the S states, accessible through a measurement of the triples 1S-2S interval [2], offer the possibility of the most precise determinations. Due to the optical metastability of the $2\,^3S_1$ state and the relatively long annihilation lifetime of the $1\,^3S_1$ state, the natural linewidth of the transition is only 1.3 MHz, about one part in 10^9 of the 1S-2S interval. The interval is presently known with a precision of 13 MHz from an analysis of the measurement using a pulsed laser [3]. Significant improvements in the precision might be obtained by using a cw laser to eliminate the nonlinear effects on the two-photon line shape of frequency chirps associated with a pulsed laser [4], and a cold source of Ps to reduce the transit time and second-order Doppler broadening of the line shape.

In the previous experiment [2], the 2S positronium atoms excited by counterpropagating 486-nm pulsed light were detected by subsequent photoionization in the same 486-nm beams. At low intensities, the two-photon excitation probability is proportional to the square of the intensity, whereas the three-photon ionization probability is proportional to the cube of the intensity. At intensities corresponding to cw excitation, the photoionization probability becomes negligible compared to the excitation probability, making a separate high-intensity pulse to photoionize the excited state atoms necessary. In the present paper we report experiments on the Dopple-free 1S-2S laser excitation and the separate photoionization detection of the 2S states. We make use of a source of Ps at a significantly lower temperature [5] than was used in the original precision measurement of the 1S-2S interval. We have been able to extend our measurements to low power, allowing us to test our model at excitation levels expected in a cw experiment, and thus predict the behavior of such an experiment.

Our measurements were made using a narrow-band pulsed laser source to optically excite thermal positronium atoms generated in vacuum. The e^+ were generated by an accelerator [6] in 10-ns pulses at 30 Hz, each containing 5×10^4 slow e^+. The e^+ are implanted with 1–2-keV kinetic energy into a 99.999% pure Al(111) sample prepared by electropolishing in a (1:5) perchloric-acid–ethanol mixture and by repeated cycles of Ar^+-ion bombardment and annealing at 820 K. The sample was cooled to 235 ± 5 K and then exposed to 5×10^{-8} torr of O_2 for 8 min, resulting in a partial monolayer of O on the Al surface. About 10% of the incident positrons are reemitted from the surface with 0.2-eV energies and 10% leave the surface as thermal positronium with a beam-Maxwellian velocity distribution [5].

As shown in Fig. 1(a), a Coherent 699-21 actively stabilized, cw ring dye laser serves as a frequency stable (< 1 MHz) source of 486-nm radiation. This is used as an injection seed beam for up to four stages of a modified Lambda-Physik traveling-wave pulsed dye amplifier. The amplifier is pumped with 20-ns 400-mJ pulses from a Lambda-Physik XeCl excimer laser, resulting in a 25–30-MHz laser linewidth. A monochromator is used to monitor the ratio of amplified signal to amplified spontaneous emission (ASE), verifying that ASE accounts for less than 1% of the total laser power.

The expanding telescope between the second and third stages of the amplifier chain is set to focus the unapertured seed beam at a plane 8 m from the exit of the amplifier, ensuring that the spatial Fourier transform of the exciting pulsed beam is formed at this plane as well.

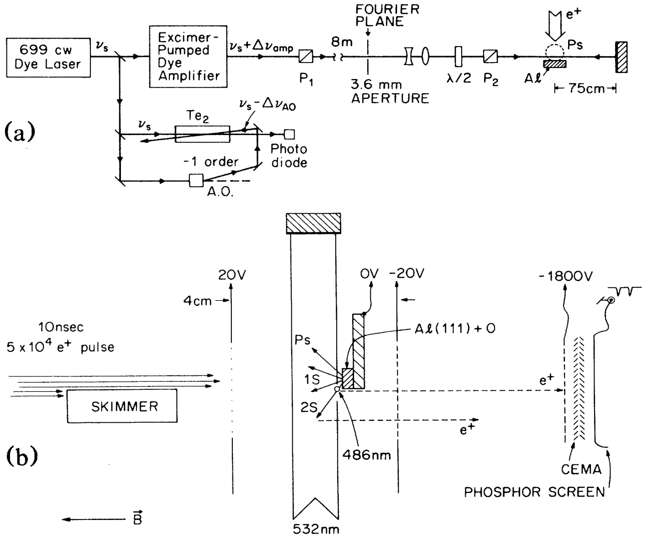

FIG. 1. Experimental setup. (a) Laser configuration. (b) Detail of positronium source and excitation region.

A 3.6-mm-diam spatial filter is placed in the Fourier plane. A beam-expanding telescope follows the spatial filter, focusing the pulsed beam to a waist at the retroreflecting mirror 75 cm behind the Al positronium source. The beam size measured at the source is $w=0.55\pm0.02$ mm, where only the central portion of the Airy pattern is visible in the burn pattern. The small beam size was chosen to be comparable to the beam that can be attained in the TEM$_{00}$ mode of a 1-m build-up cavity.

The source, excitation, and collection geometry have been modified from the previous experiment, in which the laser excitation region was directly in front of the positronium source. Since these experiments have always been done in an axial magnetic field (100 G) to simplify the positron optics, collecting the photoionized positrons from this region has involved $\mathbf{E}\times\mathbf{B}$ deflection of the positrons to the side of the target where they may be accelerated axially to a time-resolved channel-electron-multiplier array (CEMA) detector. Because of the relatively small excitation probability of Ps at low laser intensities, the re-emitted positrons represent an overwhelming background, despite efforts to distinguish them using the time resolution of the CEMA.

Our new excitation geometry, shown in Fig. 1(b), includes two essential features. First, to eliminate the problem with re-emitted positrons, the positronium-laser interaction volume is now split into two separate regions. The upper region contains the incoming positron beam and the Al(111) target. Occurring in the lower region are the photoionization and collection of the resulting positrons.

The incident-positron beam is skimmed to produce a 10%–90% width of 1.5 mm to the bottom edge of the initially circular positron beam. This edge is lined up with the bottom of the Al(111) target, allowing less than 10^{-3} of the incident positrons to hit the CEMA. As shown in Fig. 1, the counter-propagating 486-nm beams are directed parallel to the bottom edge of the target (into the figure) and 1.0 mm away from the target surface.

A second new feature is a 532-nm beam [30 mJ per pulse, second harmonic of Nd:YAG (yttrium aluminum garnet)] that fills a region to the left on the 486-nm beam roughly 0.5 cm wide and 2.0 cm deep (into the figure) in a multipass arrangement. The result is nearly unity photoionization probability for excited-state Ps in this region.

Any atom passing through the center of the 486-nm beam will subsequently pass through the photoionization region below the target. An axial electric field accelerates the resulting photoionized positrons toward the CEMA. Re-emitted positrons from the target are unable to reach the CEMA, since they are trapped along magnetic-field lines and cannot pass the target. The net result of this new geometry is a 100-fold reduction in noise and a signal positron collection efficiency comparable to that obtained in the previous experiment.

A plastic scintillator registers the 10-ns burst of annihilation γ rays from the target area. The relative timing of the 486-nm and YAG laser pulses is measured by directing a small fraction of these beams at a pinhole in the scintillator housing. We find a maximum of the YAG photoionization signal with a delay of 40 ± 5 ns for the 486-nm pulse and a delay of 110 ± 5 ns for the YAG pulse relative to the prompt annihilation γ rays.

With the 486-nm laser frequency set on resonance, and the 532-nm beam on, the time spectrum of e^+ incident on the CEMA, shown in Fig. 2(a), has three peaks. These are due to incident 1-keV e^+ that pass under the target, excited-state photoionization by the 486-nm beam, and excited-state photoionization by the 532-nm beams, respectively. The 486-nm photoionization peak occurs at 150 ns rather than 40 ns because the axial energy (10 eV) of these signal positrons is lower than that of the incident positrons. Likewise, the 532-nm photoionization peak occurs slightly earlier than 70 ns after the 486-nm signal because the 532-nm photoionization region is at a slightly higher electrical potential than the 486-nm region. In Figs. 2(b) and 2(c) we show the spectra with the 532-nm beam turned off and the 486-nm beam set off resonance, respectively. The voltages on the input grid, the output grid, and the target are set to maximize the size and temporal sharpness of the observed signals. The magnitude of the electric field $E=10$ V/cm, produces a Stark shift of roughly 0.4 MHz.

A portion of the seed beam is used to determine the frequency of the cw laser beam to ±1 MHz relative to the saturated absorption spectrum of the Ps reference line in Te$_2$ vapor. The saturating pump beam used in the spectrometer is the -1 order of a 104-MHz acousto-optic modulator (AOM). In addition to providing modulation at 50 kHz for lock-in detection, the AOM produces a -52-MHz shift in the seed-beam frequency required to observe the Doppler-free Te$_2$ resonance. The pulsed laser output is not typically centered at the seed-beam frequency. We measure a shift of the output of the third amplifier stage of 40.3 ± 2 MHz to the blue for our amplifier setup. Thus, for the most recent measured values of the Ps transition frequency [3] and the Te$_2$ reference line [7], when the seed beam is set to observe the Te$_2$

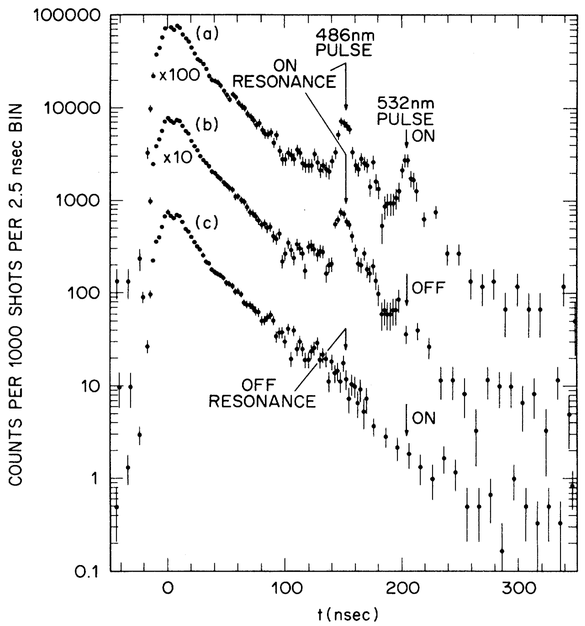

FIG. 2. Time spectra of detected positrons. (a) 486-nm excitation beam on resonance and 532-nm photoionization beam on, (b) 486-nm excitation laser on resonance and 532-nm beam off, and (c) 486 and 532-nm lasers on with the 486-nm laser frequency detuned several GHz from the Ps line.

resonance, the pulsed laser is $\Delta\nu = 29 \pm 8$ MHz above the Ps $1S$-$2S$ two-photon transition frequency, including ± 6-MHz uncertainty due to beam misalignment, and ± 5.4-MHz uncertainty in the measured Ps transition.

A laser frequency scan of the Te_2 resonance and the 486-nm photoionization signal is shown in Fig. 3(a). The large ac Stark shift at the laser power used in this scan shifts the center of the observed spectrum roughly 100 MHz to the blue of the actual Ps line center. The frequency offset $\Delta\nu$ was chosen to optimize the signal at high power levels so various experimental parameters could be adjusted in real time by observing the photoionized e^+ signal on a pulse-by-pulse basis.

A pair of crossed calcite polarizers is placed in the pulsed beam with a rotatable halfwave plate between to allow continuous adjustment of the laser power in the interaction region with negligible motion of the laser beams. A fraction of the retroreflected laser light is reflected onto a photodiode for relative laser-power measurements. Using only three stages of the dye amplifier, we typically find a maximum available power at the interaction region of 2.5 mJ per pulse at the 30-Hz repetition rate, more than sufficient to saturate the two-photon transition.

Figure 4 shows a scan of the 486-nm and YAG photoionization signals as a function of the incident-peak laser power. The data were taken by averaging the time spectrum over 1000–5000 pulses to obtain reasonable statistics at each laser power. The pulsed laser detuning from the Ps line was held constant at $\Delta\nu = 29 \pm 8$ MHz during each run by observing the Te_2 spectrometer. The relative laser power was determined by measuring the peak signal on a photodiode, well below saturation, and normalizing the results to a power meter measurement made at the highest power setting. At each power setting, the average time spectrum was found with the YAG laser both on and off. The number of YAG ionization counts was found by subtracting the off signal from the on signal. The number of 486-nm counts was found by subtracting the laser detuned spectra from the YAG off spectra. The error bars indicate uncertainty due to Poisson statistics.

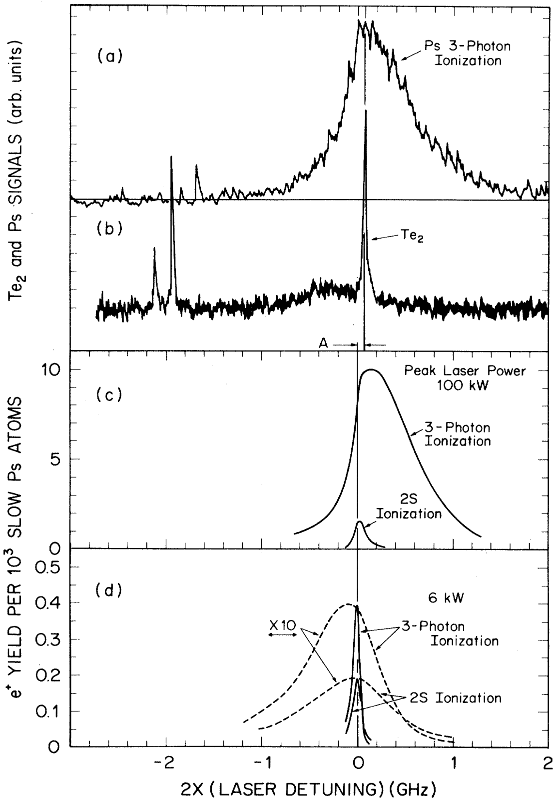

FIG. 3. Laser frequency scan showing Ps-pulsed laser spectrum. (a) Experimental results at a peak laser intensity $I = 28$ MW/cm^2, (b) model results at same intensity, and (c) model results at a peak laser intensity of 1.5 MW/cm^2 corresponding to the lowest laser power used in our measurements.

To explain the above results and determine if a cw measurements of the $1S$-$2S$ Ps interval is feasible, we have developed a computer model of our experimental geometry allowing us to integrate the excitation and photoionization probabilities over 10^5 randomly chosen positronium trajectories. For each classical atomic trajectory, the complete equations of motion of the atomic state amplitudes under two-photon excitation are integrated and the resulting transition probabilities are summed. A complete description of the model will be presented elsewhere.

Using our model, we have calculated results for the experiments described above. Figure 3 shows the model line shape for both 486-nm and 532-nm photoionization at a pulse laser intensity in the interaction region of $I = 28.0$ MW/cm^2. Also shown are the two model-calculated line shapes at a laser intensity of $I = 1.5$ MW/cm^2.

Figure 4 shows the model results for the laser power scan. In the model calculation, we have used the measured values for all model parameters. Because of the relatively large uncertainty in our estimated experimental detuning and the sensitivity of the results of the laser power scan to this parameter, we have varied the model value of the laser detuning. The best agreement was found with a model detuning of +30 ± 5 MHz with clear disagreement between model and experiment at the stated limits. This is in good agreement with the measured detuning, providing independent confirmation of the measured $1S$-$2S$ positronium interval with an uncertainty of ± 20 MHz.

The reason for the sensitivity to detuning may best be understood in terms of Fig. 3. At high laser intensities, the broadening of the observed line is dominated by ac Stark shift, which always shifts the transition to the blue. When averaged over a large number of trajectories, the strongly Stark-shifted line will be skewed to the blue and thus have a sharper edge at the red side of the line.

At low laser intensities, the broadening of the observed Ps line will be dominated by the second-order Doppler shift, which always shifts the transition to the red. Consequently, the result is a sharper edge to the blue side of the line. To experimentally observe the transition at both high and low intensities, we have set the pulsed laser detuning at the red side of the high-intensity spectrum and the blue side of the low-intensity spectrum for the laser power scan. In the model, we have set the value of detuning to give the experimentally measured ratio of high- to low-intensity signal.

We assume a photoionization efficiency of 50% by the 532-nm beam, and a collection efficiency of 50% for the positrons photoionized in the 486-nm beam, since, as seen from the CEMA, half of that beam is blocked by the target. If we additionally assume a 50% detection efficiency by the CEMA, then we estimate that the number of thermal Ps atoms emitted by the source is roughly 1500 Ps/pulse, in agreement with estimates based on previously measured values of Ps production efficiency and the estimated 2×10^4 positrons in the skimmed incident beam. These efficiencies have been used to scale the model results to the experimental signal size.

In finding substantial agreement between our model and experimental results, we now believe we can predict the signal and line shape of a cw experimental configuration. In the model we use the geometrical parameters of the TEM_{00} mode of a super-high-finesse build-up cavity we have prepared for this purpose. The cavity has a 1-m confocal geometry with a circulating power buildup of 10^4, yielding 1 kW circulating power with only 100-mW cw input power. The beam waist is $w_0 = 0.278$ mm, giving a peak intensity $I = 0.825$ MW/cm^2. With the same production and detection efficiencies and the identical target-laser geometry used in the pulsed experiment, our model predicts a peak cw signal of 30 counts per 10^3 shots in a 15-ns window, with a background of roughly 10 counts per 10^3 shots in the same window.

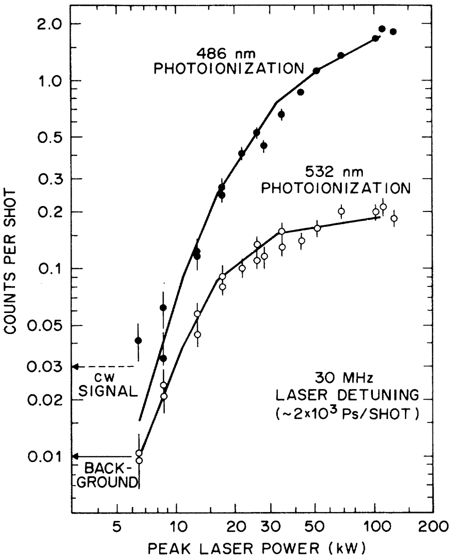

FIG. 4. Photoionization signals vs peak laser power showing experimental results and model calculation (solid curves). The dashed line indicates the model-calculated cw signal size, and the solid line indicates the background noise level.

In conclusion, we have made measurements of the Doppler-free excitation probability of the $1S$-$2S$ transition in Ps at low laser powers corresponding to a possible cw excitation. By using a separate high-intensity photoionization beam, the detection efficiency is improved by an order of magnitude. In addition, an alternative excitation and collection geometry results in an improvement by several orders of magnitude in the background noise over a previously used geometry. We have developed a computer model of the two-photon transition, incorporating our experimental geometry, that explains the experimental results and allows us to predict the signal strength expected using cw laser excitation. We conclude that a precision cw measurement of the Ps $1S$-$2S$ transition to the level of ± 1 MHz is feasible with existing cw apparatus.

Note added in proof. We have recently observed a cw excitation of the Ps $1S$-$2S$ transition. The signal was observed with 500 W of circulating laser power and is consistent with the computer model results.

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*Present address.

[1] For a general reference, see J. R. Sapirstein and D. R. Yennie, in *Quantum Electrodynamics,* edited by T. Kinoshita (World Scientific, Singapore, 1990), p. 560.

[2] Steven Chu, Allen P. Mills, Jr., and John L. Hall, Phys. Rev. Lett. **52**, 1689 (1984).

[3] K. Danzmann, M. S. Fee, and Steven Chu, Phys. Rev. A **39**, 6072 (1989).

[4] M. S. Fee, K. Danzmann, and Steven Chu (unpublished).

[5] A. P. Mills, Jr. *et al.*, Phys. Rev. Lett. **66**, 735 (1991).

[6] R. H. Howell, R. A. Alverez, and M. Stanek, Appl. Phys. Lett. **40**, 751 (1982); A. P. Mills, Jr. *et al.*, Rev. Sci. Instrum. **60**, 825 (1989).

[7] D. H. McIntyre and T. W. Hänsch, Phys. Rev. A **36**, 4115 (1987).