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OPTICALLY EXCITED STATES IN POSITRONIUM

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ABSTRACT

We report optical excitation of the 1^3S-2^3P transition in positronium, and a second excitation from n=2 to higher n states. The experiment used light from two pulsed dye lasers. Changes in the positronium annihilation rate during and after the laser pulse were used to deduce the excited state populations. We found that we could saturate the n=2 level and excite a substantial fraction of n=2 positronium to higher levels. Preliminary spectroscopic measurements were performed on n=14 and n=15 positronium.

INTRODUCTION

Although positronium (Ps) has been known for many years, few experiments have created and maintained a population of Ps in other than the ground state. In previous experiments, a small fraction of Ps has been formed in the n=2 state by bombarding metallic targets with a low energy positron beam [1], or by exciting Ps from the ground to the n=2 state with an incoherent, broadband light source [2]. Two photon excitation of the 1^3S-2^3S transition [3] has also been used, but leads to prompt photo-ionization. At Livermore, we have created significant populations of n=2 Ps through optical saturation of the 1^3S-2^3P transition using a frequency-doubled, pulsed dye laser [4,5]. We have also excited higher n states with a second dye laser tuned to red wavelengths [5]. We find that we are able to repeatedly excite the Ps atoms in our laser beam to the n=2 levels, and subsequently excite n=2 Ps to higher n states where it is long-lived. Moreover, we have optically saturated the n=2 level using a broad bandwidth laser resonant with a large fraction of the Doppler profile of the Ps. These conditions are required for the recently suggested technique of broadband laser cooling [6].

BACKGROUND

The Ps ground state is split into a singlet state which undergoes 2γ annihilation with a 125 ps lifetime, and a triplet state which undergoes 3γ annihilation with a 142 ns lifetime (figure 1). The annihilation lifetimes of

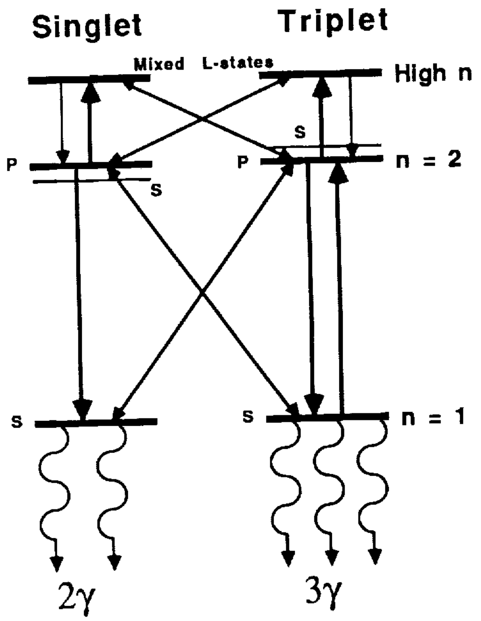

Figure 1. Schematic energy level diagram of Ps showing transitions between states mixed by magnetic and electric fields typical of this experiment. The laser bandwidths cover the entire n=2 and high n multiplets.

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the singlet and triplet 2P levels are > 100 ls, which are significantly greater than the 3.2 ns radiative lifetime. In a field free region, singlet-triplet electric-dipole transitions are not permitted. However, in the presence of a magnetic field, the Zeeman effect mixes singlet and triplet levels with the same magnetic quantum number [7,9]. In the ground state, this leads to magnetic quenching [10], i.e., a reduction in the triplet lifetime due to the small admixture of the singlet state. In first order perturbation theory, mixing of the singlet and triplet levels is inversely proportional to the energy difference between the levels. Due to the smaller energy separation for larger n values, mixing fractions increase with increasing n state. Hence, a 200 G field, which does not significantly mix the ground state, induces a several percent admixture of the amplitudes of the singlet state in the n=2 triplet levels [11]. At high values of n (>10), this field will cause the spin states to be completely mixed.

In our experiment, we observe the excitation of Ps through changes in the annihilation rate. The initial population of excited state Ps is negligible at production, and the singlet Ps atoms rapidly annihilate, leaving a population of ground state triplet Ps. Since the annihilation lifetimes of the 2P and higher states are long compared with their radiative decay lifetimes, essentially all annihilation occurs in the ground state. When no magnetic field is present, exciting Ps to the n=2 levels reduces the observed annihilation rate during the laser pulse. However, when a magnetic field is present, an enhancement in the annihilation rate can occur during the laser pulse since transitions between singlet and triplet levels are possible between Zeeman-mixed levels. Thus, each excitation of the Ps to n=2 levels can result in an enhanced annihilation rate by allowing de-excitation to the singlet ground state. At high transition rates, i.e. high laser intensities, a significant fraction of the illuminated Ps may be made to annihilate in a short time compared to the triplet ground state lifetime. Moreover, due to the Δm selection rules for different photon polarizations, the annihilation rate is also influenced by the laser light polarization. Thus, for fixed magnetic field and laser polarization, the number of enhanced annihilations is directly proportional to the n=2 singlet population.

The loss of Ps from enhanced annihilation during the laser pulse will lead to a reduction in the number of annihilations after the laser pulse. This reduction is due to the decrease in the triplet ground state Ps population caused by enhanced annihilation. The Ps ground state population can also be reduced by excitation to higher n states. Thus, there are two time intervals that are separately identified with annihilation changes proportional to the singlet n=2 population and changes in the triplet ground state population. Excitation to higher n states will affect both these populations by reducing the triplet ground state population and changing the singlet n=2 state population.

The number of excess annihilations of Ps resulting from excitation to the n=2 level can be calculated using a rate equation model for the 1S-2P transitions, and using perturbation theory to calculate the Zeeman mixing [9]. The magnitudes of electric-dipole transitions to states with $\Delta m = 0, \pm 1$ depend on both photon polarization and direction of photon propagation with respect to the magnetic field. Mixing into singlet Ps follows the Zeeman selection rules [7,8], and all 2^1P_1 Ps are assumed to decay to 1^1S_0 and annihilate. The results of the calculation are presented as a function of the laser pulse energy in figure 2, where we plot the change in annihilations induced in a 40 ns window by a 10 ns FWHM Gaussian laser pulse. The same detection probability was assumed for both singlet and triplet decays. The results are presented for several values of magnetic field. For low values of B, where little mixing occurs, the number of annihilations decreases due to the time spent in the n=2 level. At large values of magnetic field (~ 200 G), the annihilation rate from mixing into the singlet state becomes large causing an enhancement in the annihilation rate.

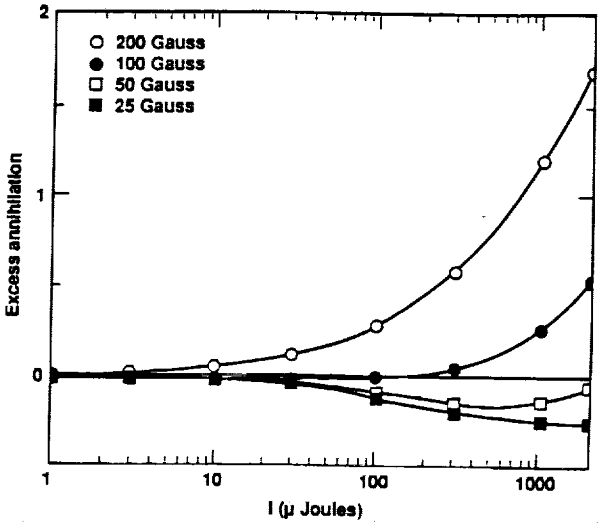

Figure 2. Calculated fractional change in the number of annihilations observed in a 40 ns window due to a 10 ns FWHM laser pulse (centered in the window) as a function of energy per laser pulse.

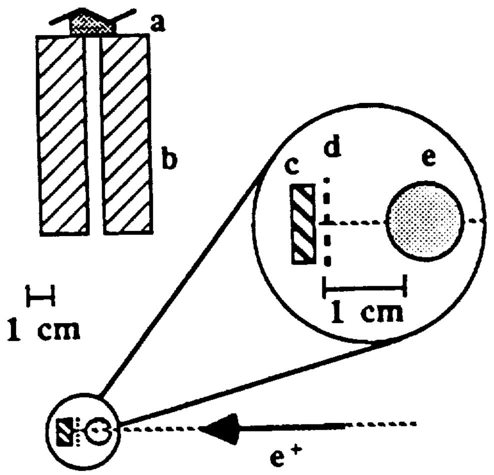

Figure 3. Schematic diagram of the apparatus showing the annihilation gamma detector (a) and its collimator (b) in relation to the target (c), grid (d), and laser profiles (e) (inset drawing). The incoming positron beam is centered on the dotted line.

EXPERIMENT

A schematic of the experimental geometry is shown in figure 3. Positronium is produced by focusing the Livermore intense, low energy (1 keV), positron beam [12] in an ultrahigh vacuum chamber on a clean, heated (1000 K) Cu target. An electrically biased grid, mounted in front of the target, returns any bare positrons to the target. The heated sample emits both energetic (kinetic energy ~ 2.6 eV) and thermally desorbed Ps (kinetic energy ~ 0.1 eV) [13] which then drifts through the grid into the laser interaction region. Several plastic scintillator detectors were used to detect annihilation gamma rays. Uncollimated detectors observe the Ps annihilation throughout the chamber, while collimated detectors are used to separate Ps by energy through time-of-flight techniques [14]. The laser interaction region, directly in front of the Cu target, is viewed by a collimated scintillator paddle with a field of view 1.4 cm wide along the path of the laser beam.

To achieve resonant n= 2 excitation over a significant fraction of the Doppler profile of the thermal Ps, we have frequency-doubled the 485.906 nm output of our excimer pumped, Lambda Physik Fl 2002 dye laser. The light is focused in a β-Barium Borate doubling crystal to obtain 242.953 nm light with a typical 0.07 nm FWHM bandwidth. This covers a significant fraction of the estimated 0.15 nm FWHM Doppler profile of the transition and allows excitation of all of the 2P levels. The output of the doubling crystal is passed through a prism to separate the fundamental from the first harmonic. The resulting ultraviolet beam is passed through a second prism, expanded to an ~ 1 cm diameter, and then passed in front of the Cu target. Calibration of the laser is performed to 0.005 nm with a monochromator, cross-calibrated to the 242.795 nm line of an Au, hollow cathode, discharge lamp.

To excite higher n states from the n=2 population, we split the pump beam of our excimer laser to pump a second dye laser which was tuned to wavelengths in the 720 to 750 nm region and calibrated to 0.005 nm by comparison to an Ne discharge lamp. The red laser had a band width of 0.4 nm and a beam profile of 1 cm^2. The two laser beams covered equal path lengths and merged to coincidently enter the Ps interaction region.

The laser was timed to the linac positron pulse with a variable delay. Timing between the laser light and the positrons was monitored by a photomultiplier tube mounted with both a plastic scintillator and a fiber optic. We establish timing of the 10 ns laser pulse relative to the 15 ns positron pulse to ± 2.5 ns.

RESULTS

Time distributions of annihilation gamma rays viewed with the collimated detector were separately collected for positron pulses with the laser on and off. The laser pulsed every eleventh positron burst. A typical result is shown in figure 4a (see next page) with B=200 Gauss, the ultraviolet light delayed 90 ns after the positron pulse, and 460 μ-joules per laser pulse. The normalized laser-off data is subtracted from the laser-on data to obtain the difference distributions shown in figures 4b-4d. Figure 4b is the difference plot obtained from figure 4a. In figure 4c, the peak in the annihilation rate is seen during the laser pulse, now delayed by 70 ns. In figure 4d, the laser has been detuned by 1.0 nm, producing a random distribution.

Measurements were made for the n=2 excitation for several values of laser pulse energy, with B=200 G. In figure 5b we see the excess counts during the laser pulse normalized to counts in a time interval including all annihilations except the prompt burst. Data taken with the laser detuned by 1.0 nm in a 200 G magnetic field and with the laser on resonance in a 50 G magnetic field are represented by the open triangle and the open circle, respectively. Both of these points show no enhanced annihilation during the laser pulse, as expected from our rate calculations.

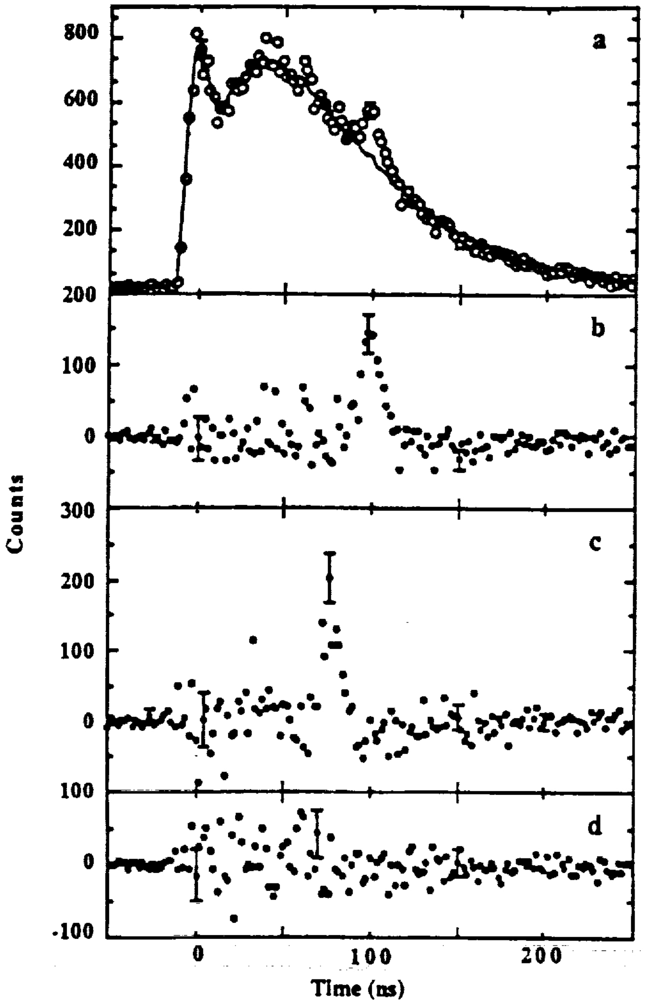

Figure 4. a) Typical annihilation time distributions collected by the collimated detector for ultraviolet illumination with the laser off (solid curve), and laser pulsed at 90 ns (open circles) taken with a 200 G magnetic field. b) Laser-on minus laser-off difference spectrum obtained from the data of 4a. c) Difference spectrum as in 4b, but with the laser pulsed 20 ns earlier. d) Difference spectrum as in 4c, but with the laser detuned 1.0 nm to the red of the transition.

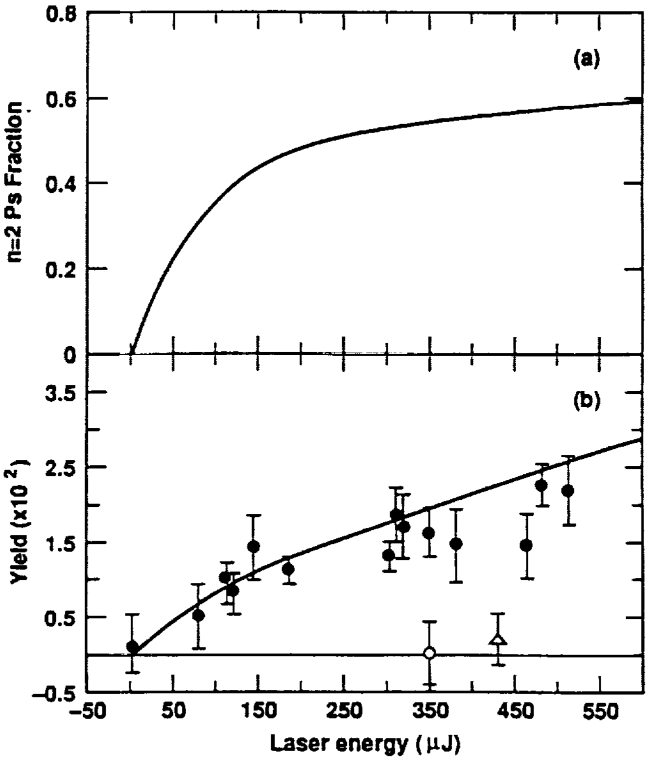

Figure 5. a) Results of the rate equation model showing the fraction of illuminated Ps in the n = 2 state. The calculation is performed for a 200 Gauss magnetic field and laser parameters typical of our measurement. b) Excess annihilations during laser illumination normalized to the total Ps production for several laser powers. The solid curve is the calculated excess annihilations. The open circle is taken with the laser detuned 1.0 nm and B = 200 G. The open triangle is taken with the laser on resonance and B = 50 G.

We also measured the excess annihilations for linearly and circularly polarized ultraviolet light. In both cases the direction of photon propagation was normal to the magnetic field. The linear polarization plane was parallel to the magnetic field. With circularly polarized light, the annihilation rate was found to be 0.42 ± 0.21 of that observed with the plane polarized light (~100 μJ per laser pulse) in agreement with the calculated ratio of 0.36 obtained in the rate equation model for those polarizations [9].

By comparing the excess annihilation data with our rate equation calculation, we see that we have reached optical saturation. The results of the calculation, normalized to the data using a Monte Carlo simulation, are plotted as the solid line in Fig. 5b. The model accurately describes the laser energy-dependence of the transition rates. We view the numerical agreement between the calculated and measured excess annihilations as fortuitous due to the strong sensitivity of the Ps population in the laser beam on small variations in experimental parameters. The calculated fraction of the Ps in the n=2 state for illuminated Ps is plotted as a function of laser energy in figure 5a. This curve shows the characteristic shape of a saturation spectrum. There is a linear dependence on laser power at low energy per pulse, with an asymptotic approach to the limit of 0.67 at high laser energy.

Typical data for the excitation of higher n states are shown in figure 6. The data in figure 6a were taken with only ultraviolet light illuminating the Ps, leading to resonant excitation of n=2 state. The data in figure 6b were taken with both ultraviolet and red light illuminating the Ps. This data shows the effects of resonant excitation of both the n=2 and n=15 states. Here we find that excitation of the n=15 level reduces the ground state Ps population. This causes a larger deficit in the late time annihilations. The annihilation rate during the laser pulse is also reduced, implying that the n=2 state population is also depleted by excitation to higher n states.

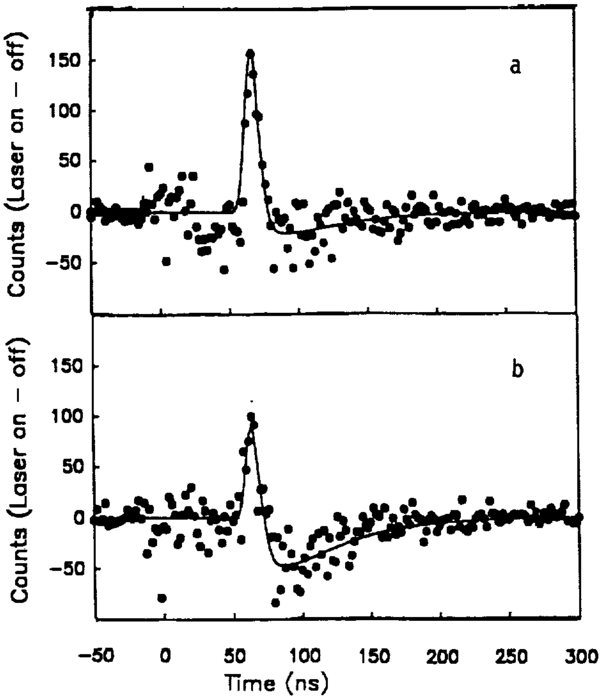

Figure 6. Difference spectra for a) ultraviolet illumination only and b) resonant two photon excitation to the n=15 state. Curves are calculated using a rate equation model with strong mixing in the n=15 state.

The decrease in excess annihilations during the laser pulse when the red laser is on resonance implies a decrease in the singlet n=2 state population. Ps remaining in long-lived, high n states after the laser turns off is a source of depletion of the Ps ground state population. However depletion of the n=2 state population will not occur unless the total number of de-excitations from the high n state is less than the number of excitations. Losses to photoionization are negligible. We calculate that the photoionization rate is less than 0.001 of the high n excitation rate at our laser energies. Large differences in the population of the n=2 and high n states can be obtained if there are corresponding differences in the number of sublevels.

We have numerically solved time-dependent rate equations describing this experiment for several possible cases. The dipole selection rule for the n=2 to high n transition limits the number of high n state sublevels to too low a value to explain the observed decrease in the n=2 state population during the laser pulse. Zeeman mixing will cause the high n singlet and triplet spin states to be statistically populated for our experimental parameters. Our calculations show that de-excitation from a statistically populated singlet high n state would result in an increase in annihilations during the laser pulse rather than the decrease we observe. However, including strong Stark mixing can break down the $\Delta l=+/-1$ dipole selection rule and allow population of all of the l sublevels of the high n state accessible with $\Delta m=0,+/-1$. Such mixing can occur due to static and motionally induced electric fields of ~40 V/cm found in the chamber.

The computed results of the rate equation model are shown in figure 6a and b, normalized to the level of annihilation enhancement seen in figure 6a. The calculations were performed using measured values for laser power and pulse characteristics. The curve in Figure 6b includes transitions from all populated n=2 sublevels, to all accessible l-sublevels of the n=15 state. These calculations reproduce the observed change in enhanced annihilations and even the change enhanced annihilation time profile seen in comparing on and off resonance spectra. Calculations containing many fewer sublevels in n=15 state, which might result from weaker Stark mixing, cannot reproduce both the change in annihilations during the laser pulse and the decrease in the ground state population observed with the red light on resonance. Thus, we conclude that the high n states are completely mixed. Our calculated results also show that a significant fraction, ~0.3, of the n=2 state population is promoted to higher n states where it remains after the laser pulse has ended.

Shown in figure 7 are data for wavelengths on and off resonance between n=13 and n=19. Data were taken at more closely spaced wavelengths around the n=14 and 15 resonances in order to determine experimental centroid wavelengths and widths. A ratio, formed from counts in two time intervals as a function of wavelength, is shown here. The first interval is a narrow time window including the duration of the laser pulse and subsequent radiative decay from the n=2 level. The second interval extends from the end of the first interval until the data reached background values. The absolute value of the counts lost during the second interval were decided by the counts in the first interval to form the ratio. Thus, this ratio is the decrease in ground state Ps normalized to the singlet n=2 population and is insensitive to systematic differences in the geometry and target condition. Positronium lost to excess annihilations in the n=2 state alone results in a baseline value of 0.62 . Excitation of Ps into higher n states gives a larger ratio due to a larger depletion of the ground state Ps.

In the data in figure 7 we see a convincing demonstration to resonantly excite high n states from the n=2 population. There is a large increase in the yield at wavelengths resonant with excitation to the n=13, 14 and 15 levels. Calculated excitation profiles for the resonant peaks are also shown in figure 7. The peak wavelengths were calculated from the Balmer formula, and the widths of the curves included the 0.4 nm laser line width and the 0.4 nm Doppler broadening width for these transitions. The relative areas of the calculated peak shapes were determined using the rate equation model with the transition rates scaled by n^{-3}. The absolute normalization was set to the sum of the areas of the n=14 and 15 data. From these calculations we see that values of n greater than 15 are more difficult to excite and are not easily observed with the statistics now available in the experiment.

Values for the centroids and widths for the n=14 and n=15 peaks were calculated from the data in figure 7 after background subtraction. We obtained values of 744.049 +/- 0.035 nm and 741.993 +/- 0.040 nm for the n=14 and n=15 centroids, respectively, and 0.37 nm and 0.44 nm for the full widths at half maximum, respectively. The centroid values compare favorably with the reference values of 743.988 nm and 741.995 nm obtained by calculating the energy difference between the unshifted energy of the 2^3P_1 and the energy of the high-n state. Hyperfine splitting in the high-n state is negligible in this comparison, but spliting of the 2P levels adds .02 nm to the width of the excitation-line profile. The

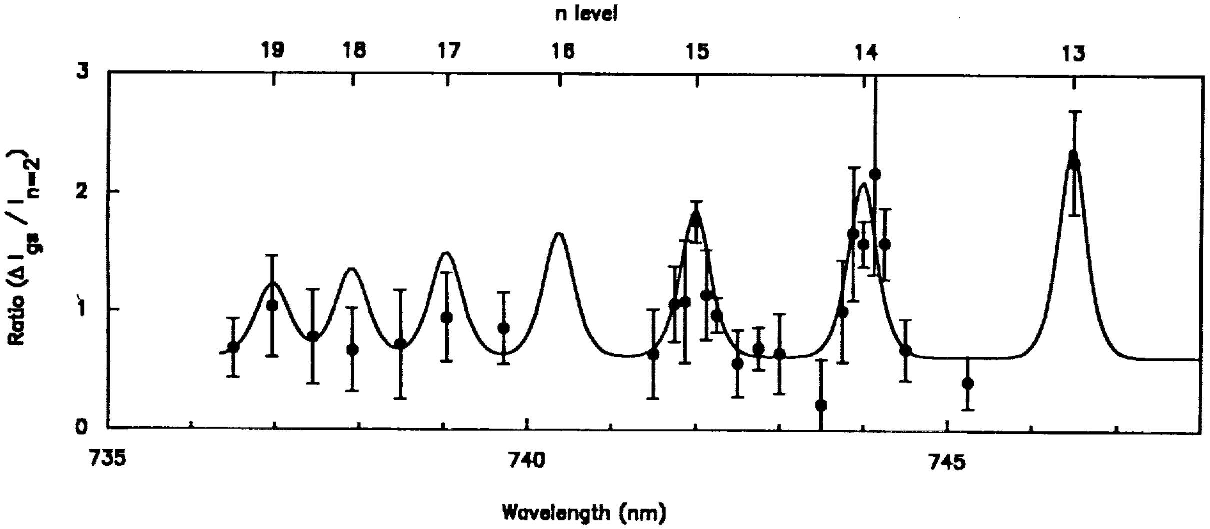

Figure 7. Ratio of loss in the ground state population normalized to the n=2 population. The solid line is the expected response calculated from known parameters of the experiment.

large measured widths for the n=14 and 15 excitation are consistent with the Doppler spread in the Ps and line profile of the two laser beams. The good reproduction of the energies and widths within the experimental errors demonstrates the potential to perform higher precision spectroscopy by using narrower laser line widths and lower velocity Ps.

These results represent a first step towards laser-cooling Ps. They show that transition rates, photon bandwidths and excitation of spectator transitions required to achieve and measure cooling are possible. As pointed out by Liang and Dermer [6], one must use a broadband laser to cool Ps since the lifetime is too short to sweep the laser frequency. The 0.07 nm bandwidth of our laser is ideally suited to cool the Ps but the pulse duration is currently too short. The agreement between calculation and experiment for excitation to the n=2 and higher n states indicates that sufficient Ps atoms survive to perform cooling experiments.

The observation of high n state excitations provides a useful diagnostic for the velocity of cooled Ps, and a basis for new spectroscopic investigations. Detecting the ionization products of excited Ps resulting from photon or static electric field ionization can be used to develop a higher efficiency detector for high n state Ps. With this detector we use narrow laser lines to tune over the velocity profile. This same system would form the bases for spectroscopic studies on the cooled Ps.

In summary, we have demonstrated optical saturation of the resonant transition of Ps from the ground to the first excited state. We have also observed the first resonant excitation of high n states of Ps using two resonantly excited transitions, from 1S to 2P and 2P to nL. The population of the excited states was deduced from observation of annihilation rates during and after the laser pulse. Magnetic mixing in the n=2 state increased the annihilation rate during the laser pulse, and loss of Ps in the ground state population reduced the annihilation rate after the laser pulse. Changes in the enhanced annihilation rate with laser power, photon polarization and magnetic field show that the n=2 state was in optical saturation. Magnetic and electric fields in the chamber mixed the high n states so that all l sublevels were populated in the high n state. Values for the line centroid and widths agreed with calculations. Qualitative reproduction of the n^{-3} scaling of the relative transition rates was also observed.

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