First Observation of Resonant Excitation of High-n States in Positronium

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We have performed the first excitation of high-n states of positronium using resonant, two- photon excitation applied to the $n=1$ to 2 and 2 to n transitions. Absolute values for the line positions were quantitatively determined for $n=14$ and 15 and compared well with calculated predictions. The prediction of the n^{-3} scaling of the relative transition rates was observed for $n=13$ to 19.

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Since the discovery of positronium (Ps), spectroscopic studies have been performed on sublevels of the ground and $n=2$ energy levels of the system. These include the singlet-triplet splitting in the ground state,[1] the two-photon $1S$-$2S$ transition,[2] and the $2S$-$2P$ splitting in the first excited state.[3] These measurements represent a sensitive test of quantum electrodynamics due to the precision of the experimental results, lack of importance of strong forces, and high calculational accuracy available from the theory. In this Letter, we report the first observation in Ps of the excitation of high-n states, $n=13$–15, a preliminary measurement of the energy of two of these levels, $n=14$ and 15, and preliminary observations on the relative transition probabilities for $n=13$–19. Excitation of high-n states was performed by a resonant, two-photon excitation $n=1$ to 2 and $n=2$ to 13–19 and using an extension of our previous investigations into the production of an optically saturated population of $n=2$ Ps.[4]

The basic quantum electrodynamic interactions in Ps, particularly energy shifts induced by perturbative external fields, may be tested in new ways by measurements of the energies and transition properties of high-n states. Excitation to high-n states results in a population of long-lived, neutral Ps because the deexcitation and annihilation lifetimes scale as n^{-3}. Such excited populations of Ps can be used to study areas such as antihydrogen production or exotic many-body states of Ps.[5] We can also tune through the resonant excitation profile using narrow laser linewidths allowing us to map the velocity distribution of the ground- and ($n=2$)-state populations. Since these transitions leave the Ps system in a bound state, they can be of particular use in laser-cooling experiments.[5]

In this experiment we excite high-n states in Ps through a two-step excitation process using the $n=2$ state as an intermediate state. The experiment was conducted using extensions of the techniques previously employed to observe the population of the $n=2$ levels of Ps.[4] Ps and laser light interacted in the ultrahigh-vacuum, experimental chamber used with the intense, low-energy positron beam at the 100-MeV electron linac at Lawrence Livermore National Laboratory. Thermal Ps was formed by guiding a 1-keV pulsed positron beam with a 200-G magnetic field onto a hot, 1000-K, clean copper single crystal cut along the 100 face. The target was biased with respect to a grid to attract any reemitted positrons. Pulses of 15-ns duration typically contained 10^5 positrons which were converted into Ps with an overall efficiency of 0.8 in an ($n=1$)-state population of 0.25 singlet and 0.75 triplet sublevels.

The output of a Lambda Physik EMG 103, XeCl excimer laser was split and used to simultaneously pump two dye lasers. Ultraviolet light to pump the $1S$-$2P$ transition was obtained by frequency doubling the output of one of the dye lasers to 242.953 nm (Ref. 4) with a linewidth of 0.07 nm. Red light to pump the $n=2$-to-high-n transition was obtained from a second dye laser having a 0.4-nm linewidth. The broad laser linewidths were used to maximize the overlap with the equally broad Ps Doppler profiles. Laser pulses were typically of 10-ns duration containing 300 μJ and 5 mJ in the ultraviolet and red pulses, respectively. The beams were expanded to a 1-cm^2 area, configured so that they had equal path lengths and merged before entering the Ps interaction region. The laser wavelengths were calibrated to ± 0.005 nm in a monochrometer, cross calibrated to a Au, hollow-cathode, discharge lamp and a Ne lamp for the ultraviolet and red wavelengths, respectively. Data were taken with the ultraviolet light fixed at the resonant excitation wavelength for $n=2$ given above and the red light tuned over the range of several high-n states.

The excitation of Ps was determined by observing deviations from the annihilation rate of the initial Ps population. The ground-state triplet and singlet lifetimes are 142 and 0.12 ns, respectively, so that only triplet Ps remains at the time of the laser pulse. Two modifications of the annihilation rate were observed. An enhanced annihilation rate occurs during the laser pulse due to mixing of singlet and triplet sublevels of the $n=2$ state in the 200-G magnetic field used in this experiment.

Although annihilation occurs from the singlet ground state after optical deexcitation of the $n=2$ state, the enhancement in annihilations is directly proportional to the $n=2$ singlet population in this experiment. This is because the singlet ground state is only populated through optical deexcitation of the $n=2$ level. Details of the transition rates and mixing ratios are found in Ref. 6.

A reduction in the annihilation rate after the laser pulse occurs when there is a loss in the Ps ground-state population. Losses in the ground-state population can be caused by enhanced annihilation, excitation to long-lived, high-n states, or by photoionization. At the fluence levels we used, photoionization by any combination of ultraviolet or red photons has been calculated to be less than 0.001 of the high-n excitation rate. The optical (annihilation) lifetimes of high-n states are many microseconds, $\sim 150(500)$ μs at $n=15$. The optical deexcitation lifetimes for $n=2$ are shorter than the laser pulse so no Ps remains in $n=2$ states. Thus in these data there are two separate time regions, during and after the laser pulse, that are identified with Ps in $n=2$ and in the ground state, respectively. Excitation to higher-n states will affect both these populations by reducing the ground-state population and changing the singlet ($n=2$)-state population according to the details of the transitions.

Annihilation radiation was detected with a plastic scintillator collimated so that the field of view of the detector was 1.4 cm wide centered 1 cm in front of the target and included the laser-Ps interaction region. The time distribution of Ps annihilations was stored for positron pulses with both the laser on and the laser off with the laser pulse rate set at $\frac{1}{11}$ of the linac pulse rate. The laser was typically pulsed 70 ns after the arrival of the positron pulse on the target. Relative timing of the positron and laser pulses was established to within ± 2.5 ns by observing both annihilation and laser pulses in the same photomultiplier tube.

Typical data are shown in Fig. 1. The data in Fig. 1(a) are raw spectra obtained with the ultraviolet laser on (solid circles) and off (solid line). The peak at time $t=0$ results from annihilations at the copper target scattering into the detector. The broad distribution at later times is formed by Ps annihilation in the field of view of the detector. Additional annihilations during the laser pulse are easily seen. The data in Figs. 1(b) and 1(c) are obtained by subtracting normalized laser-off data from the corresponding laser-on data. In Fig. 1(b) only ultraviolet light illuminated the Ps leading to resonant excitation of the $n=2$ state. The data in Fig. 1(c) were taken with both ultraviolet and red light illuminating the Ps leading to resonant excitation of $n=2$ and $n=15$ states.

In comparing spectra 1(b) and 1(c) we see that population of the $n=15$ state further reduces the ground-state Ps population and we see a larger deficit in post-laser annihilations. Some depletion of the Ps ground-state population is always expected since some Ps remains in the long-lived high-n state after the laser is turned off. However, the annihilation rate during the laser pulse is also reduced implying that the time-averaged ($n=2$)-state population is depleted by excitation to the $n=15$ state.

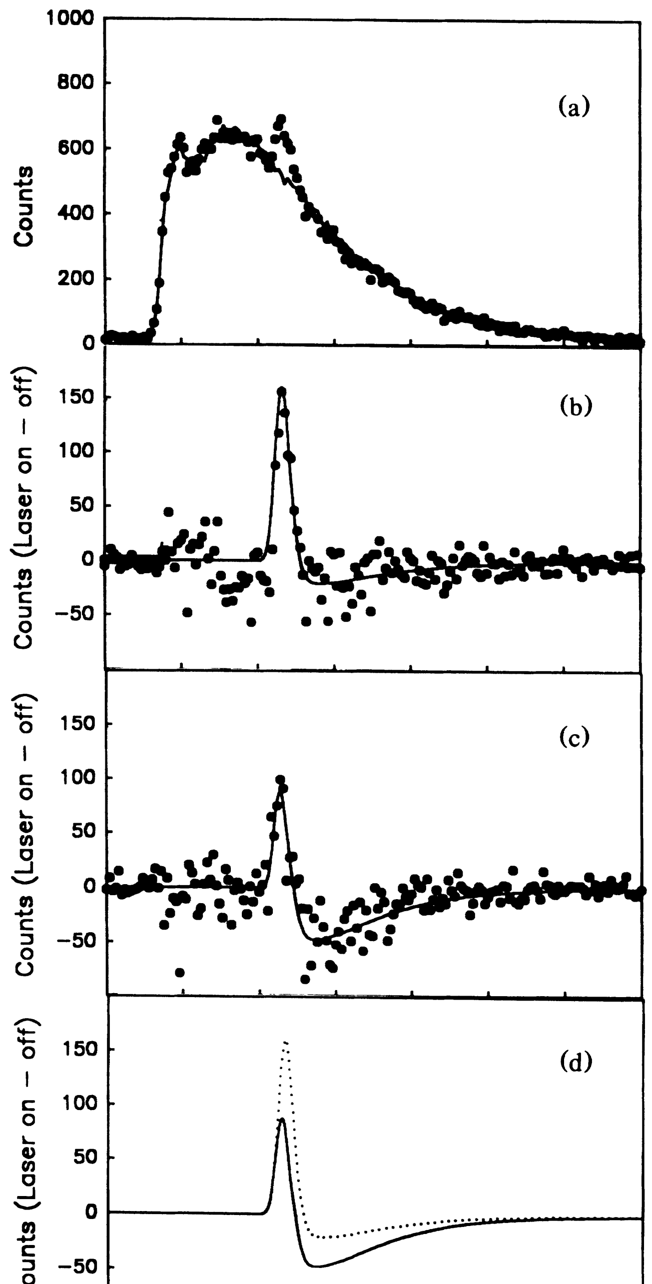

FIG. 1. Data and calculations of the time distribution of annihilation photons in a collimated detector. For a description of the spectra see the text.

Depletion of the $(n=2)$-state population cannot occur unless the total number of transitions populating the $n=2$ states is less than the number of transitions depopulating that state. 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 in the two states. In order to test the response of annihilation rates to changes in available sublevels induced by mixing, we have numerically solved the time-dependent rate equation for this experiment for several possible cases.

For our experimental parameters, Zeeman mixing causes high-n singlet and triplet spin states to be statistically populated. The dipole selection rule for the $(n=2)$-to-high-n transition limits the number of populated sublevels in the high-n state to a value which is too small to explain the observed decrease in the $(n=2)$-state population during the laser pulse. Our calculations show that deexcitation from a singlet high-n state, statistically populated under the dipole selection rules, would result in an increase in annihilations during the laser pulse, rather than the decrease we observe. Stark mixing can break down the $\Delta l = \pm 1$ dipole selection rule and allow population of all of the l sublevels of the high-n state accessible with $\Delta m = 0, \pm 1$. Such mixing can occur due to the static and motionally induced electric fields of $\sim$40 V/cm found in the chamber. The computed results of the rate-equation model are shown in Figs. 1(b) and 1(c) with both calculations normalized to the level of annihilation enhancement seen in Fig. 1(b). These results are plotted without data in Fig. 1(d) for easier comparison. The calculations were performed using measured values for laser energy and pulse characteristics. The curve in Fig. 1(c) included transitions from all populated sublevels in $n=2$ to all accessible sublevels of all l values of the high-n state. These calculations reproduce not only the measured reduction in enhanced annihilations, but also the change in the temporal profile of the enhanced annihilation peak seen in comparing the red light on and off spectra. Calculations containing many fewer sublevels in the high-n state, such as would occur with weaker Stark mixing, cannot reproduce both the decrease in annihilations during the laser pulse and the decrease in the ground-state population observed with the red light on resonance. From this we conclude that the high-n states are strongly Stark mixed indicating that j is no longer a good quantum number for these states. Our calculated results also show that a significant fraction, $\sim$0.3, of the $(n=2)$-state population is excited by the red light to higher-n states where it remains after the laser pulse has ended.

Results of measurements at several wavelengths are shown in Fig. 2. The data in Fig. 2 are the ratio of counts in two time windows in the laser-on–laser-off difference spectrum. The number of extra annihilation events found in a narrow, 45-ns, time window set to include the duration of the laser pulse and subsequent optical decay was used to normalize the loss of counts in the period starting after the end of the narrow window and extending until the data reached background values. This ratio is the decrease in ground-state Ps normalized to the time average of the singlet $n=2$ population and is free of many systematic differences in the geometry and target condition from run to run. Ps lost to excess annihilations through $(n=2)$-state excitation alone gives a background value of 0.62 for this ratio when the red light is off resonance or blocked. Excitation of Ps into higher-n states leads to a larger depletion of the ground-state Ps and thus a higher value for this ratio when the red light is tuned on resonance. The data in Fig. 2 include on- and off-resonance values between $n=13$ and 19. Data were taken at more closely spaced wavelengths

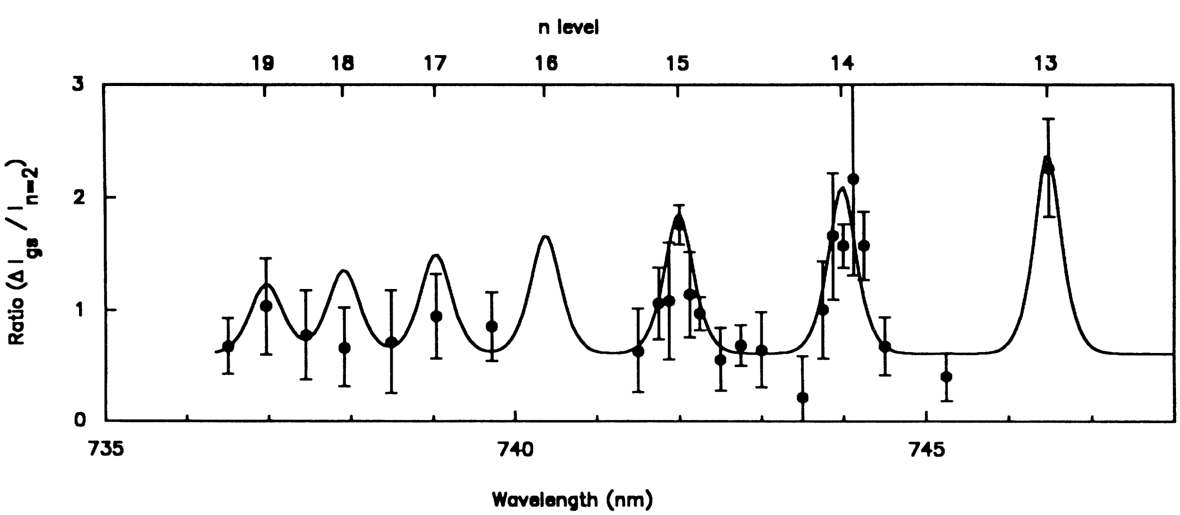

FIG. 2. Decrease in the ground-state population normalized by the $n=2$ singlet population. The solid line is calculated using known parameters of the experiment.

around the position of $n=14$ and 15 to allow the calculation of experimental centroid and width values.

The data in Fig. 2 clearly show our ability to resonantly excite high-n states from the $n=2$ population. There is a large increase in the yield at resonant wavelengths for $n=13$, 14, and 15. The anticipated excitation shape for the resonant peaks is also shown in Fig. 2. The centroids of the peaks were placed at the wavelengths expected from theory and the width of the calculated peaks included the laser linewidth, 0.4 nm, and the Doppler-broadening width, 0.4 nm, of the Ps states. The relative areas of the calculated peaks were determined from ratios calculated in the rate-equation model with the transition rates scaled by n^{-3}. Absolute normalization was determined from the sum of the areas of the $n=14$ and 15 peaks. From these calculations we see that our data generally follow the prediction of an n^{-3} scaling of the transition rate. We also see that states with higher-n values are more difficult to excite and are not easily observed at our present level of precision.

Values for the centroid and width of the peaks for $n=14$ and 15 were calculated from the data in Fig. 2 after background subtraction. The values are 744.049 ± 0.035 and 741.993 ± 0.04 nm for the centroids of the $n=14$ and 15 peaks, respectively, and 0.37 and 0.44 nm for the respective full widths at half maximum. The centroid values compare favorably with the reference values of 743.988 and 741.995 nm obtained by calculating the energy difference between the unshifted energy of the triplet $^2P_1$ and the energy of the high-n state calculated from the Balmer formula for Ps. Hyperfine splitting in the high-n state is negligible but splitting of the $2P$ levels adds 0.02 nm to the linewidth.[7] The absolute positions of the lines agree well with predictions and the large measured widths for the $n=14$ and 15 excitation are consistent with the Doppler broadening in the Ps and line profiles of the two laser beams.

The good reproduction of the energies and widths demonstrates the potential to perform higher-precision spectroscopy by using narrower laser linewidths and initial populations of low-velocity Ps. Such measurements are currently difficult to perform due to the low counting rates encountered in this experiment. However, direct detection of ionization products from field ionization or photoionization of the high-n states may result in significantly higher detection efficiencies. We are currently exploring these possibilities and expect more than a thousandfold increase in counting efficiency. This will allow future experiments to be performed with narrower laser profiles and higher counting statistics, improving the accuracy and precision of the measurements.

In summary, we have performed the first resonant excitation of high-n states of Ps using two resonantly excited transitions $1S$ to $2P$ and $2P$ to nL. 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 were quantitatively determined and compared favorably with calculated values. Qualitative reproduction of the n^{-3} scaling of the relative transition probabilities was also observed.

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