Part II

Positronium, Muonium, and Other Hydrogen-Like Systems

Laser Spectroscopy of Positronium and Muonium

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I. Introduction

The laser spectroscopy of leptonic atoms is of interest because of the simplicity of the atoms. Leptons possess no known structure. Unlike the hydrogen atom with its complicated and poorly understood proton, the behavior of leptonic atoms should in principle be calculable to much greater precision. In the case of muonium, the theoretical uncertainty will be limited by our knowledge of fundamental constants such as m_μ/m_p and α. Given the 72 kHz linewidth of the 1S state due to the 2.2μsec lifetime of the muon, one can ultimately expect a measurement of the 1S-2S splitting better than $\Delta\nu/\nu < 10^3$ Hz/10^{15} Hz = 10^{-12}. In the case of positronium, the 140 nsec lifetime should lead to an ultimate precision of $\Delta\nu/\nu = 10^{-11}$. Thus, this system provides a unique opportunity for the precise study of a purely leptonic two-body system, and QED corrections to that system. In addition to QED tests, the laser excitation of these atoms could provide thermal, sub-thermal, and eventually cryogenic sources of positrons and muons that can be used in a variety of applications.

This article will outline the experimental techniques we have used in the laser spectroscopy of these atoms and briefly indicate current plans for the refinement of these measurements. As Fig. 1 shows, the laser spectroscopy of positronium and muonium is not competitive with comparable measurements in hydrogen, largely due to the low density sources of these atoms. In the case of positronium, the first measurements were done at peak densities of a few atoms/cm^3 during a laser pulse. The muonium work was limited by atom densities ~ 10^{-2} atom/cm^3 per laser pulse. As feeble as these sources might seem to spectroscopists of less exotic atoms, one must remember that these instantaneous densities represent many orders of magnitude improvement of above pre-existing sources of thermal positronium and muonium. Clearly, improved sources will lead to more precise measurements.

II. Positronium

Figure 2 shows the lowest lying energy states of positronium and the energy intervals that have been measured to date. The optical spectroscopy of the positronium 1^3S_1-2^3S_1 resonance has been previously described. [1] Briefly, the long lived triplet positronium was excited by two counterpropagating 486

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nm laser beams via the two-photon Doppler-free technique. Atoms in the n=2 state were then ionized with high probability and the detected positrons were recorded as the laser frequency was tuned through the resonance.

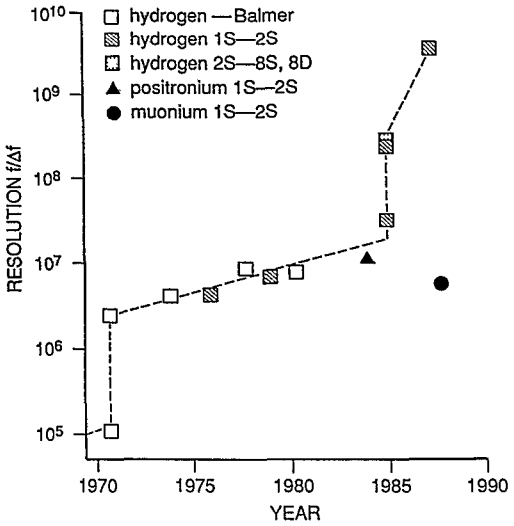

Fig. 1 Laser spectroscopy of hydrogen, positronium and muonium as a function of time.

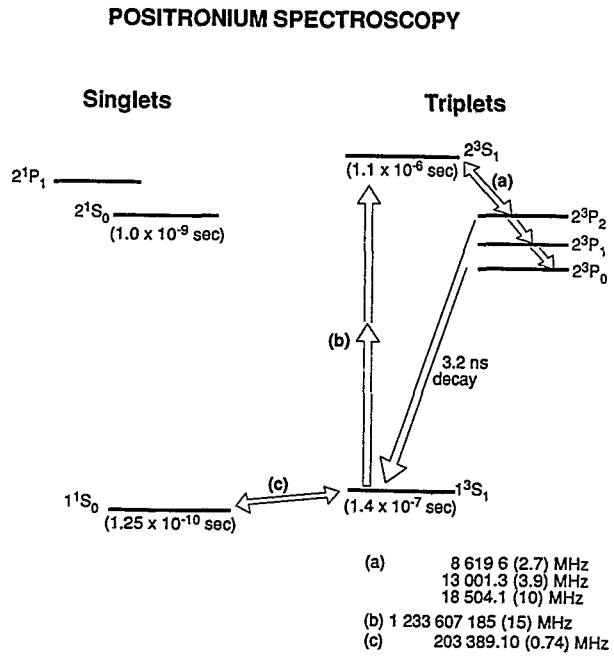

Fig. 2 Lowest energy level structure of positronium showing the measured energy level splitting.

Figure 3 shows a block diagram of the experimental apparatus. Starting with greater than 10^9 positrons/sec emitted from a ^{58}Co β^+ source, on the order of 10^6 positrons/sec are extracted from a cooled, single crystal tungsten moderator and directed into an e^+ magnetic bottle for storage. The bottle allows us to match the e^+ source to the duty cycle of the pulsed laser used to excite the atoms. A harmonic buncher in the magnetic bottle could be pulsed at a kilohertz rate, producing sub-keV positrons in 10 nsec pulses. The approximately 80 e^+/pulse are directed to an Al(lll) target heated to 300°C. By heating the metal surface, positrons that would normally annihilate in surface states are thermally desorbed as free thermal Ps in vacuum. During the experiment, roughly 20 thermal Ps would be emitted from the 1 cm dia Al target per bunching pulse.

The laser system consisted of a cw dye laser oscillator amplified by a XeCl excimer pumped 4-stage dye laser amplifier. Nominally 50 mw of cw power was amplified to 20-25 mJ, 10-nsec laser pulses with a frequency bandwidth of 70 MHz. The cw laser frequency was measured relative to the deuterium $2S_{2/3}$-$4P_{3/2}$ line in a three-step process. We first used a theoretical value for the deuterium line from Erickson's calculation and measured the shift between that line and a particular molecular line (labeled 1326 in tellurium atlas [5]) in the tellurium spectra. Next, the Ps resonance was measured relative to the tellurium standard. Finally, the frequency of the amplified pulsed laser beam is measured relative to the cw laser. Since the Doppler free counter-propagating beams used to excite the Ps atoms are produced in a flat-flat Fabry Perot interferometer inside the vacuum chamber (free spectral range = 450 MHz, finesse ~ 35) the beam pointing instability of the pulsed laser translated into a time averaged laser bandwidth of 30-35 MHz. A 300 MHz free spectral range confocal interferometer was used to measure the frequency shift of the pulsed laser light transmitted through the vacuum Fabry-Perot cavity relative to the cw light.

The dominant uncertainties in the measurement were the dc Stark shift of the deuterium line in the discharge tube, the ac Stark shift of the Ps line, the second-order Doppler shift of the thermal Ps and the amplified laser frequency shift relative to the cw laser. Counting statistics show up in the measurement of the Ps - tellurium frequency shifts as one extrapolates to zero laser power and zero atomic velocity.

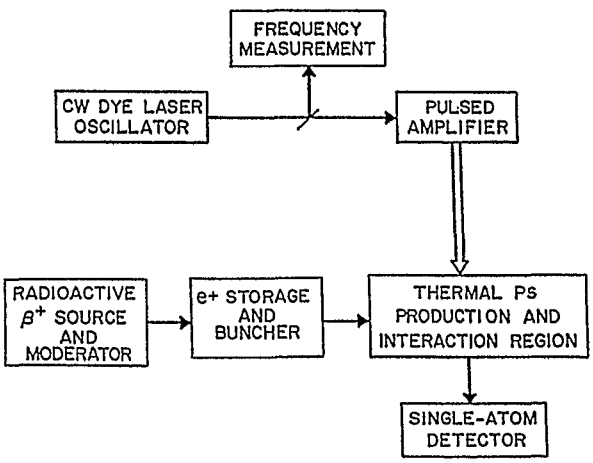

Fig. 3 Block diagram of the positronium experiment.

The quoted uncertainty in the measurement of the Te_2 line relative to the hydrogen reference was ± 10.5 MHz while the uncertainty of the Te_2 line relative to the Ps resonance was ± 10.6 MHz. The final result of the measurement was $\Delta\nu$ (1^3S_1 - 2^3S_1) = 1 233 607 185 ± 15 MHz. [1] Since that measurement, the tellurium line was recalibrated by McIntyre and Hansch [6], causing the experimental value to be shifted significantly. The new value is $\Delta\nu$ (Ps) = 1 233 607 142.9 (10.7) MHz.

The theoretical value of the energy interval is given as an expansion in powers of α $\Delta\nu$ (1^3S_1 - 2^3S_1) = 3/8 $R_\infty C$ {1 + $K_2\alpha^2$ + $K_3\alpha^3$ + $K_4\alpha^4$ +...} where the contributions are listed in Table I. The α^4 and $\alpha^4 \ln\alpha$ terms have not been calculated, although the $\alpha \ln\alpha$ term is estimated by scaling $1/n^3$ behavior of the hfs splitting.[7] The α^4 term would be 3.5 MHz if the uncalculated K_4 coefficient turned out to be 1. Thus, the uncertainty due to the uncalculated terms could easily be ± 30 MHz. If we ignore the uncertainties due to the uncalculated terms, $\Delta\nu_{theory}$ - $\Delta\nu_{expt}$ = 56.4 (11) MHz. Clearly, more experimental and theoretical work needs to be done.

Table I Positronium Theory

3/8 $R_\infty c$	1 233 690 730 (1.3) MHz
$K_2\alpha^2$	- 82 005.616 (18)
$K_3\alpha^3$	- 1 527.440 3 (5)
$K_4\alpha^4$	(3.5 MHz if K_4 =1)
$\alpha^4 \ln\alpha$	(-4.2 MHz ± ?)

$\Delta\nu$ (1^3S_1 - 2^3S_1) = 1 233 607 197.(1.3)

In addition to tests of QED, precision measurements of the energy levels of Ps can be used to establish upper limits in other areas of physics. For example, we have implicitly assumed that the Ps Rydberg R_{Ps} is exactly 1/2 R_∞. Agreement between experiment and theory can be used to establish an upper limit on m_{e+} - m_{e-}. [1] Precision Ps spectroscopy can also be used to set upper limits on electron coupling to light scalar particles. The unexpected observation of correlated back-to-back emission of e^+e^- pairs in heavy ion collisions [8] has led to the suggestion of a new neutral particle of mass M ~1.7 MeV. If the new particle has a scalar coupling $g\phi\psi\psi$, measurement of the electron g-factor limits the coupling constant to $g^2/4\pi < 10^{-8}$. [9] Given this upper limit the 1S-2S splitting in Ps can be off from the QED value by 100 KHz for a point-like particle. If the mystery particle has structure, the form factor would tend to suppress its contribution to the electron g-2 value relative to Ps energy level shifts. Schafer, *et al.* [10] have pointed out that the agreement with theory in the ground state Ps sets good limits on possible vector, axial vector, and pseudo-scalar couplings, but does not say anything about a scalar coupling.

Work is currently underway to re-measure the 1S-2S energy level splitting. An electron accelerator beam dump at AT&T Bell Laboratories has been shown to produce 4 x 10^4 slow positrons/pulse at 30 Hz, and further improvements are expected.[11] We anticipate a 10^3 increase in the number of available thermal Ps. In collaboration with M. Fee and K. Danzmann at Stanford, we are also working

on an improved laser system. Fig. 4 shows a block diagram of some of the essential features of the new laser system. The cw dye laser will be stabilized to < 100 KHz with an improved servo system, and an electro-optic rf side-band will be locked to the Te_2 line. Frequency scanning will be done by using an rf frequency synthesizer to shift the rf sideband locked to the Te_2 line. The shift between the amplified dye laser and the cw oscillator will be measured by beating the two laser beams together after the pulsed laser is passively filtered to ~ 6 MHz bandwidth.

Precision measurements with high powered pulsed lasers are difficult because of the incomplete knowledge of the optical electric field. A common problem with amplified laser pulses is that the beam is never Fourier transform limited because of the rapid index of refraction modulation of the amplifying medium. Filtering with a Fabry-Perot cavity after amplification does not eliminate the problem. As an example, we assumed a linear frequency-chirped gaussian beam a factor of two worse than the Fourier transform limit, computer modeled the transmission through a Fabry Perot cavity, and then calculated numerically the two-photon transition rate. A 6 MHz filtered laser beam showed a 1.5 MHz red shift relative to the peak of the frequency spectral distribution. If the time varying electric field were known for each laser pulse (by simultaneously measuring the beat frequency of the pulsed laser with a stable oscillator and the quadrature beat freqency) this systematic effect could be accounted for. However, such a measurement may not be practical, and ultimately, we plan to create cryognic Ps atoms, either by collisional cooling or by laser cooling. [12] Assuming that the technical difficulties can be overcome, cooling the atoms to few Kelvin temperatures will allow us to measure the interval using cw excitation without excessive transit time broadening. Under these conditions, precision approaching the $\Delta\nu/\nu = 10^{-11}$ level should be achievable.

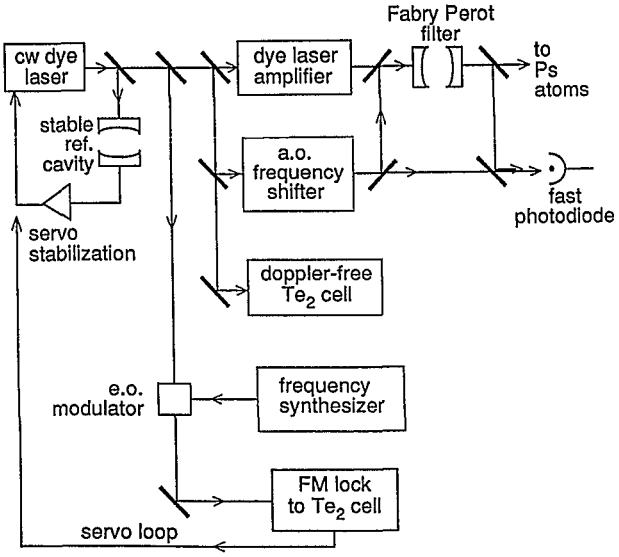

Fig. 4 Present plans for an improved laser system for positronium and muonium spectroscopy.

III. Muonium

The laser spectroscopy of muonium was recently made possible by the development of thermal muonium sources. As in the positronium work, μ^+ stopping in gas targets is considered unacceptable because of the perturbing collisions of the gas molecules. Extensive efforts to create thermal muonium in vacuum have given us two methods: (1) μ^+ stopping near the surface of a hot tungsten foil has been shown to desorb in the form of muonium analogous to the production of hydrogen and thermal Ps. [13] (2) Thermal muonium has also been produced by emission from silica-powder target. [14] In that case, it has been shown that incident μ^+ convert to muonium inside the SiO_2 balls, and then diffuse into the vacuum space between the SiO_2 grains. [15] If the powder remains uncompressed, enough interstitial space is left to allow a reasonable fraction of the muonium to escape into the vacuum.

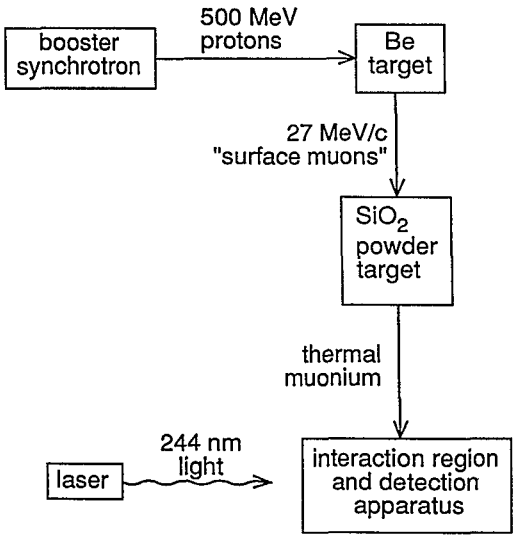

Fig. 5 Block diagram of the muonium experiment.

The first experiment on optically excited muonium [16] was performed at the Booster Meson Facility, Meson Science Laboratory, University of Tokyo, located at the National Laboratory for High Energy Physics (KEK) at Tsukuba, Japan. [17] Figure 5 shows a schematic of the experiment. A pulsed beam of 500-MeV protons incident on a Be target was used to create a low-energy muon beam resulting from the decay of π^+ stopped near the surface of the target. The bursts of 27.5 MeV/c "surface" muons were directed to a SiO_2 powder target. Typically, 100μ^+ per pulse yielded one thermal muonium in the space in front of the target. Our 1% yield is lower but not inconsistent with the 17% yield reported in Ref. 14, given the different target geometry and beam parameters.

The muonium atoms were excited and ionized by counter-propagating light beams at 244nm. The brightness of the dye laser system was increased to 80 mJ/pulse in a 30 MHz bandwidth. Using that source, we were able to generate 10-15 mJ/pulse at 244nm in a Ba_2BO_4 doubling crystal. The laser fluence needed to obtain a 50% excitation probability is 0.25J/cm² for a Fourier-transform limited pulse. Since our laser has about twice the bandwidth of an ideal pulse, we tried to operate at a fluence level of ~0.4 J/cm².

Thermal muonium that is resonantly ionized by the pulsed laser is collected by an electrostatic immersion lens, accelerated to 4kV, and directed in a 2.5m long zig-zag path to a microchannel plate detector. (CEMA) Because of the low density of muonium atoms and the small volume irradiated by the uv light, it was necessary to reduce the background counting rate to less than one count per hour in order to see a clear signal. Accordingly, the long path between the SiO_2 target and the CEMA detector was painted with colloidal graphite (Aqua-dag), and baffles and vanes were added around the electrostatic mirrors to reduce the background due to scattered light. Lead shielding 15 cm thick surrounded the CEMA.

We also found it necessary to set a very narrow time window (100 nsec) for counted events that may be due to muonium ionization. The identification of the various time-of-fight peaks in a histogram of the CEMA counts, particularly the H^+ ions emitted from the grid and target allowed us to calculate that the expected arrival time of the μ^+ is 1.43± 0.03μsec after the laser pulse. A retangular region defined by the intersection of the appropriate time cut and a 150 MHz frequency cut (75 MHz per Fabry-Perot fringe) shows six counts. The average number of counts in a rectangle of the same area arbitrarily placed in the scatter plot is 0.39 ± 0.05. Assuming a Poisson distribution, the probability that six or more counts will appear in such a rectangle is 4×10^{-6}. We therefore interpret the data as evidence for the laser excitation of muonium.

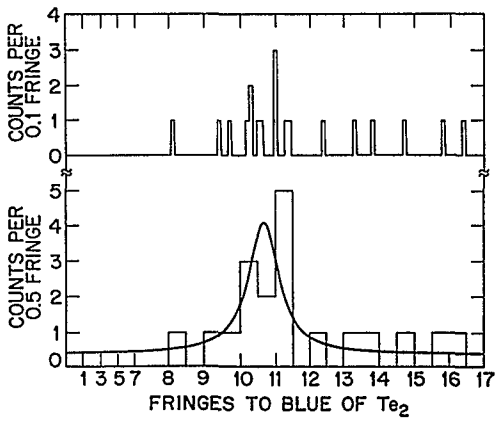

Fig. 6 Frequency spectrum for all runs taken where the laser frequency and μ^+ arrival time were recorded. The Lorentzian curve is fitted to the individual events.

In order to avoid biasing our data selection, we also include the data taken under conditions where the intense uv light was known to have partially destroyed the optics inside the vacuum chamber, the statistical significance of our result diminishes. Figure 6 shows the inclusion of 16 hours of integration time with the same time cut in the CEMA counts. A Lorentzian curve fitted to the nineteen counts given in Fig. 6 gives a line center at 10.7 ± 0.4 fringes with a fwhm of 1.1 ± 0.7 fringes. The peak signal is 7.4 ± 5.0 counts per fringe. The probability that ten or more counts between fringes 10 and 12 would occur by chance decreases to 6 x 10^{-4}. Thus, the effect shown in our best 2h run survives the inclusion of all the data. Comparison of our observation with the QED calculation was previously reported. [16] Since the resonance line is inherently narrow, our reported observation of 1/4 of the frequency of the F = 1→1 transition 613 881 924 ± 46 MHz represents a resolution $\Delta\nu/\nu = 10^{-8}$. The result is in reasonable agreement with the QED prediction of 613 881 989.90 (90). We stress that this first work should not be taken as a "measurement" of the 1S-2S splitting in muonium.

Clearly, a more intense source of thermal muonium is necessary before precise laser spectroscopy can be done on this atom. Active work to develop efficient muon moderators analogous to the positronium moderators is under way in several laboratories. Conversion of the 30 MeV surface muons into few eV muons with a moderator would allow one to generate thermal muonium very efficiently in convenient target geometries. Brighter pulsed muon sources are also under development. The next generation experiment would also use improved uv optics and possibly a brighter laser source. With these improvements one might be able to achieve a 10^4 increase in the number of counts for the same integration time. Another improvement would be the calibration of the experiment with a 1S-2S measurement in hydrogen in the same apparatus. Many of the systematic effects associated with a pulsed measurement of muonium would be similar to the effects in hydrogen where the 1S-2S splitting is measured with greater accuracy. The ionization signal can also be used to better determine the timing windows for the muonium signal.

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