

Doppler-free laser spectroscopy of positronium and muonium: Reanalysis of the $1S$ - $2S$ measurements

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A scheme for a precision measurement of the $1S$ - $2S$ transition frequency in positronium and muonium is reported, and corrections to the two most recent experiments are presented which bring their results into agreement with theory.

Positronium (Ps), the bound state of electron and positron, and muonium, the bound state of muon and electron, are the most fundamental purely leptonic two-body systems available. They provide an ideal testing ground for bound-state quantum electrodynamics (QED). New developments now permit precision optical frequency measurements to be made on these systems. While developing a new experimental scheme that will greatly improve the accuracy of such measurements, we have discovered corrections to the two most recent experiments that bring their results into agreement with theory.

Recently, the frequency of the $F=1 \rightarrow 1$ transition from the $1S$ ground state in muonium to the $2S$ metastable state was measured by Doppler-free two-photon laser excitation.¹ The result was two standard deviations smaller than the QED prediction. Shortly before, the frequency of the 1^3S_1 - 2^3S_1 transition in positronium was measured with a similar technique.² After a recalibration of the molecular tellurium reference line used in that experiment, the experimental value for the 1^3S_1 - 2^3S_1 interval turned out to be 56.4(10.7) MHz smaller than the theoretical value including the $\alpha^3 R_\infty$ QED correction.³

This discrepancy was particularly interesting in view of the recent measurement of the 3S_1 Ps ground-state lifetime, where a ten-standard deviation discrepancy between theory and experiment was found.⁴ Such a discrepancy would also have a bearing on the interpretation of the correlated e^+e^- lines recently discovered in heavy-ion collisions.⁵ It has been hypothesized that those lines might be caused by the creation and decay of a new light neutral particle.⁶ The existence of such a particle, coupling to e^+e^- , would naturally shift the energy levels of Ps.⁷ Although the accuracy of the present measurements of the anomalous magnetic moment of the electron ($g-2$) seems to exclude such a possibility⁸ (the limits derived are more than 2 orders of magnitude smaller than the discrepancy), there exists a loophole if cancellations due to several new particles occur, or, less likely, if the new object has a very large size. In these cases the effect on the ($g-2$) would be diminished, but Ps would still be affected by a level shift.

In order to resolve these discrepancies, we are currently developing an improved experiment aiming at an accuracy of 1 MHz as a first step and 10 kHz in a final stage. For the first stage of our program the output of a Coherent 699-21 actively stabilized cw ring dye laser is amplified in a four-stage pulsed amplifier system transversely pumped

by a Lambda-Physik EMG 202 MSC excimer laser. The 70-mJ, 24-nsec laser pulses have a bandwidth of 30 MHz and are sent through a confocal filter cavity to produce Fourier-transform limited pulses of 5-MHz bandwidth. Part of the cw light is split off and passed through a strip-line electro-optic modulator which creates sidebands with a variable spacing of 50 to 1000 MHz. One of the sidebands is filtered out and frequency locked to a tellurium reference line. The error signal is generated via frequency-modulation spectroscopy and controls the fundamental frequency of the cw ring laser. A variation of the rf driving the EO-modulator then effectively scans the cw fundamental frequency with an offset from the tellurium reference line that can immediately be read off from the rf signal generator.

The optical output frequency of the pulsed amplifier, unfortunately, is shifted with respect to the injected cw fundamental. The refractive index in the dye changes rapidly because the gain varies during the laser pulse. The resulting phase shift manifests itself as a frequency chirp of the pulsed output. Through computer simulation of the two-photon excitation, we have found that the amplitude and phase of the laser electric field must be determined in order to account for systematic offsets in the measured response of the atom. We feel that proper accounting of the frequency chirp describes the observed line shape of the recent pulsed measurement of the $1S$ - $2S$ transition in atomic hydrogen⁹ and accounts for the discrepancy between the pulsed and cw measurement.¹⁰ These findings will be discussed thoroughly in a later publication.

We are using a heterodyne technique to completely characterize the time-dependent complex field. Part of the pulsed light is overlapped with the cw light and the resulting beat signal recorded with a fast photodiode. The center frequency of the pulsed light is consistently shifted to the blue of the cw light by ≈ 30 MHz. If only three amplifier stages are used, this blue-shift is reduced to ≈ 20 MHz.

The earlier muonium and Ps measurements used similar amplifier configurations. In those experiments, shifts of similar size were measured with a Fabry-Perot interferometer, but the sign of the shift was incorrect due to a sign error in the scan recording. The obtained transition frequencies then have to be corrected. With our remeasured values for the pulsed frequency shift we obtain a frequency of $2455\,527\,936 \pm 120 \pm 140$ MHz for the F

$= 1 \rightarrow 1$ $1S-2S$ transition in muonium, in excellent agreement with the QED prediction of 2455 527 959.6(3.6) MHz.¹¹ For the $1^3S_1-2^3S_1$ transition in positronium, we obtain 1 233 607 218.9(10.7) MHz, which is 16.6(10.7) MHz higher than the theoretical value.¹² But, this two-standard deviation discrepancy is not too serious, considering that the uncalculated $\alpha^4 R_\infty$ term may provide a contribution of this order of magnitude (it would have to have a coefficient of ≈ 6).

Clearly, more theoretical work is required, particularly

once our experiment reaches its final accuracy, and we can expect this comparison of experiment and theory to rival the $(g-2)$ measurement as one of the most significant tests of QED.

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