Laser Excitation of the Muonium 1*S*-2*S* Transition

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We present evidence for the excitation of the two-photon Doppler-free 1*S*-2*S* transition in muonium. Thermal muonium atoms were produced in vacuum by a 27.5-MeV/*c* pulsed positive-muon beam incident on a powdered SiO_2 target. Transitions were induced by the light from a frequency-doubled pulsed dye laser and were detected by photoionization of the 2*S* state. The $1S(F=1)\rightarrow 2S(F=1)$ transition frequency is within 300 MHz of the QED prediction.

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Muonium, the bound state of a positive muon and an electron, is one of the simplest systems for testing quantum electrodynamics.[1] High-resolution measurements in muonium are particularly attractive since the presumably structureless muon allows the energy levels of this atom to be calculated more accurately than those of hydrogen, and since the recoil terms in the Lamb shift are a factor of 10 larger than in hydrogen. Positronium is also free of uncertainties in nuclear structure effects, but the relativistic two-body problem presents a formidable calculational challenge. Recently the 2*S*-2*P* Lamb-shift splitting in muonium has been measured to a few percent uncertainty.[2] The ultimate precision of these radio-frequency measurements, however, will be limited by the 100-MHz-wide 2*S*-2*P* transition. In comparison, the 1*S*-2*S* Lamb shift is ≃8 times larger and has a natural width of 72 kHz set by the 2.2-μsec lifetime of the muon. As a first step toward such high precision we have performed an experiment to ionize muonium by the two-photon–resonant Doppler-free method that has been used to study hydrogen[3] and positronium.[4] The efficient ionization of muonium would also be advantageous in the search for muonium conversion to antimuonium,[5] for the study of muonium emission from solids,[6,7] and as the basis for an intense source of thermal muons[8] that could be used in a variety of surface and atomic physics studies.

Our experiment 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.[9] 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 stopped π^+ near the surface of the target. Bursts of 27.5-MeV/*c* μ^+ in pulses 50 nsec wide and 50 msec apart were directed to a SiO_2 target 35 mm high, 60 mm wide, and 5 mm thick. It has been shown that thermal-energy muonium diffuses into the vacuum space near the SiO_2 target surface when it is bombarded with muons.[7] We find that the ≃100 μ^+ per pulse incident on the target yield typically one muonium atom in the space in front of our target. Our 1% yield is consistent with the 17% yield reported in Ref. 7 given the different target geometry and beam parameters.

The muonium atoms were excited and ionized by light beams at 244 nm counter-propagating about 4 mm in front of the SiO_2 target surface as shown in Fig. 1. The light was produced by amplification of 50 mW of cw dye-laser light at 488 nm with a XeCl-pumped dye-laser amplifier chain, and then doubling the frequency in a β-Ba_2BO_4 crystal.[10] A pump pulse energy of 400 mJ at 308 nm yielded ≃80 mJ/pulse at 488 nm (24-nsec pulse, 30-MHz bandwidth), and as much as 15 mJ/pulse at 244 nm. We calculate that a laser fluence of 0.25 J/cm^2 in a Fourier-transform–limited bandwidth pulse is necessary to obtain a 50% excitation probability. Since our laser has about twice the bandwidth of the Fourier transform of its pulse envelope, we tried to operate at a fluence level of ≃0.4 J/cm^2.

The frequency of $\frac{1}{4}$ of the $F=1\rightarrow 1$ muonium 1*S*-2*S* interval is expected to be 839(1) MHz higher than a transition in Te_2 vapor recently calibrated to be 613 881 150.89(45) MHz.[11] The calibration line, identified with the help of a Te_2 atlas[12] and a vacuum wave meter, was obtained via saturation spectroscopy with a Te_2 cell heated to 793 K. The cw laser-frequency offset from the Te_2 line was determined with use of a 74.5-MHz semiconfocal vacuum Fabry-Perot interferometer. Thus the expected position of the muonium resonance was about eleven fringes to the blue of the reference line. A 300-MHz confocal Fabry-Perot interferometer was used to measure the frequency shift of the amplified pulsed dye laser relative to the cw dye laser.

The 244-nm light entered the vacuum chamber through a Suprasil fused-silica Brewster window *W* as shown in Fig. 1. Mirrors m_1 and m_2 were adjusted to let the light make two or three passes in front of the target, *T*, before being retroreflected. Charged particles produced at the target were collected by an immersion lens,[13] accelerated to 4 kV, and guided to a microchannel plate detector (CEMA) through a series of electrostatic lenses, L_1–L_4, and mirrors, M_1 and M_2. Positive ions, presumably produced by the laser-induced photoelectron bombardment of various electrode surfaces,

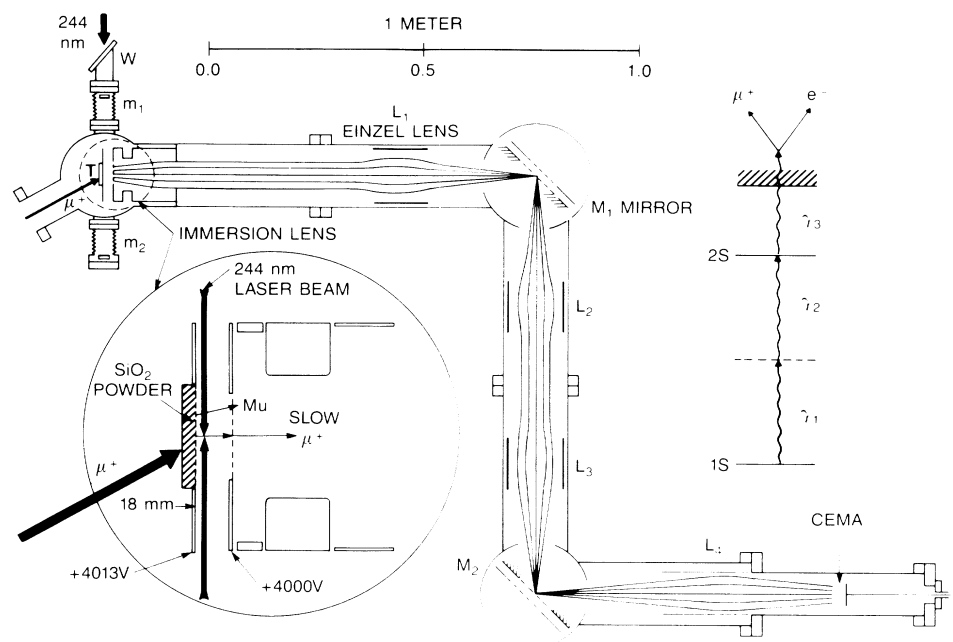

FIG. 1. Target chamber and electrostatic slow-muon collection optics. Inset: Two-photon–resonant, three-photon ionization scheme.

were a source of background counts at the CEMA. In particular, ions created near the CEMA could arrive in the expected muon time window. Because of the low density of muonium atoms ($\simeq 4\times 10^{-2}$ cm^{-3} in a 2-μsec time window) 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 within a 100-nsec time window in order to see a clear signal. Accordingly, the inside of the vacuum chamber 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.

A time histogram of the CEMA counts recorded during a run is shown in Fig. 2. At time $t=-850$ nsec, there is a small peak due to the prompt γ rays produced mainly when the primary protons hit the Be target. About 80 nsec later, the muons arrive at the SiO_2 target where a fraction of them are converted into muonium. The laser pulse was fired 770 nsec after the arrival of the muons to allow time for the muonium to diffuse away from the target. The uv light scatters throughout the chamber and is detected at $t=0$ by the CEMA. We identify the peaks centered at 3.92 and 4.44 μsec after

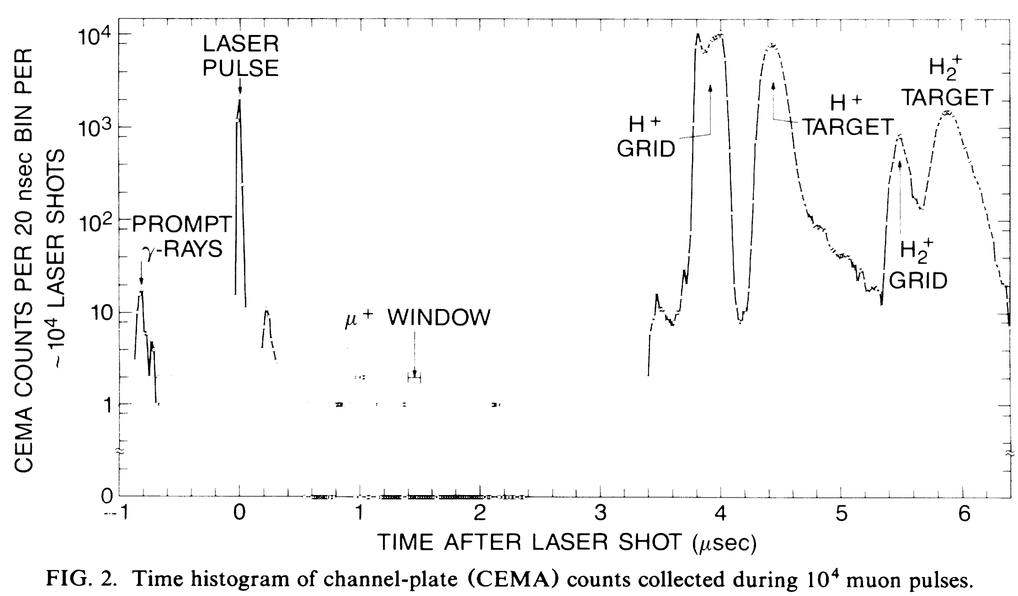

FIG. 2. Time histogram of channel-plate (CEMA) counts collected during 10^4 muon pulses.

the laser pulse with the laser-induced emission of H^+ ions from the grid at the entrance to the immersion lens and from the mesh covering the SiO_2 target surface. The subsequent peaks can be identified as H_2^+ ions. To avoid a serious reduction in the CEMA detector efficiency, we reduced the ion current without affecting ions lighter than H^+ by reverse biasing the SiO_2 target relative to the grid 0.4 μsec after the laser pulse. Any slow μ^+ liberated by the uv light would be expected to arrive at the detector in a time window (shown in Fig. 2) which occurs at a fraction $(m_\mu/m_p)^{1/2}=0.336$ of the flight time for H^+ ions from the target, or at about 1.49 μsec. The μ^+ flight time will be about 30 nsec longer because of the $\simeq$1-eV initial kinetic energy of the H^+, $\simeq$10 nsec shorter because of the kinetic energy of the thermal muonium, $\simeq$50 nsec shorter because the laser beam is 4 mm in front of the target, and $\simeq$20 nsec shorter because the photoelectrons responsible for the proton emission must travel from their point of origin to the target. We thus estimate that the μ^+ flight time is 1.43 ± 0.03 μsec. The error is due mainly to the uncertainty of the initial H^+ energy.

Figure 3 shows a scatter plot of the laser frequency and time for the events recorded during our best 2-h run. The laser frequency was scanned quickly through the Te_2 reference line until the seventh fringe (74.5 MHz/fringe) where the scan slowed by a factor of 5. A 1.5-GHz scan took about 10 min. A time-to-amplitude converter was used to record the time of arrival of the counts occurring between 1.0 and 1.8 μsec after the laser pulse. The numbers of counts near the expected position of the muonium $F=1\rightarrow1$ resonance (between fringes ten and twelve) were summed to form the histogram on the right. An excess number of counts recorded between 1.40 and 1.45 μsec after the laser pulse can be identified as muon counts since this coincides with the expected time delay for muons. The frequency histogram on the top of Fig. 3 was obtained by summing all the counts in a 1.38- to 1.42-μsec time gate that encloses the densest cluster of events. The muons are emitted from a narrow region of space and have initial kinetic energy much less than 1 eV. The muon time gate should be narrower than 0.06 μsec, $\frac{1}{3}$ of the width of the H^+. The 62 events in Fig. 3 were recorded in eleven scans during a single 2-h interval. The rectangular region defined by the intersection of the time and frequency cuts shows six counts. The expected number of counts on resonance for this data set is 5.5. On the other hand, the average number of counts in a rectangle of the same area arbitrarily placed in the scatter plot is 0.39 ± 0.05. On the assumption that the events in the scatter plot are governed by a Poisson distribution with a mean of 0.4, the probability that six or more counts occur by chance in such a rectangle at the expected laser frequency and time delay is 4×10^{-6}. We therefore interpret the data of Fig. 3 as evidence that we have excited the muonium $1S$-$2S$ transition.

The data of Fig. 3 were obtained under nearly ideal conditions: The mirrors and windows did not burn because the vacuum chamber had been baked, and the laser spatial mode structure was smooth. Other data taken in the same manner showed a much smaller counting rate, typically because of damaged optics. Nevertheless, to be sure that we are not biasing our result by data selection, we show in Fig. 4 the frequency spectrum for the sum of all the data taken in the mode shown in Fig. 3. The nineteen counts represent 16 h of integration time. Earlier data, taken with only one electrostatic mirror, a single bend in the muon flight path of Fig. 1, and a fixed 200-nsec time gate rather than a time-to-amplitude converter, showed a 2.5-standard-deviation effect with a signal-to-noise ratio of 1.5:1. Those data have not been included because the effect was small and the scan range did not correspond to the data of Fig. 3. A Lorentzian curve fitted to the nineteen counts in Fig. 4 gives a line center at 10.7 ± 0.4 fringes, with a full width at half maximum of 1.1 ± 0.7 fringes. The peak signal amplitude is 7.4 ± 5.0 counts per fringe and the background is

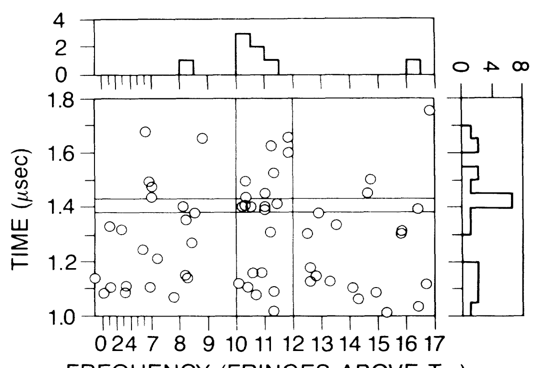

FIG. 3. Scatter plot of laser frequency and time for events recorded during the best 2-h run.

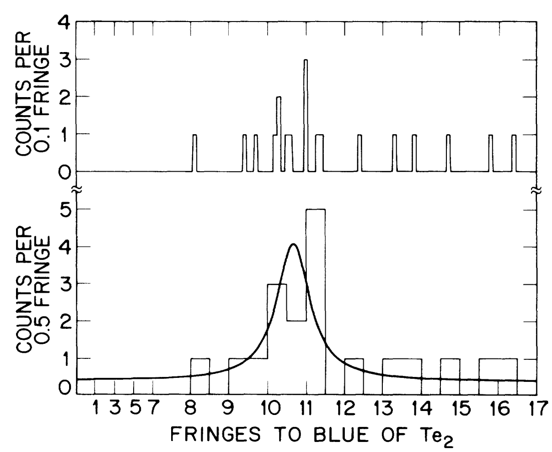

FIG. 4. Frequency spectrum for all the runs (16 h) taken in the manner of Fig. 3. A Lorentzian is fitted to the individual events shown at the top of the figure

0.8 ± 0.5 count per fringe. The average counting rate excluding the region between fringes 10.0 and 12.0 is 1.8 ± 0.6 counts per two fringes. At the expected line position between fringes ten and twelve there are ten counts, which would occur by chance with a probability of 6×10^{-4} given a mean of 1.8. Thus, the effect seen more strongly in Fig. 3 survives inclusion of all the data. Our observed linewidth of $\simeq 80$ MHz includes contributions from the laser linewidth (30 MHz), the ac Stark shift (10 to 20 MHz), the ionization rate (14 MHz), saturation of the two-photon transition ($\simeq 50$ MHz), and a residual first-order Doppler shift ($\lesssim 5$ MHz).

Before comparison of our observation with the QED calculation, we must consider three important systematic effects. The amplified pulses are shifted to the red of the cw laser by 30 ± 5 MHz because of the rapid phase modulation of the light in the amplification process. Since the resulting pulses depart from the Fourier-transform limit in an unmeasured way, it is difficult to estimate what the effect would be in a two-photon transition with use of the doubled light. We assign an error of 20 MHz to this shift. We calculate the ac Stark shift to be 3.3 Hz W^{-1} cm^{-2}, where the intensity is that of a single beam (one way). A 10-mJ/pulse beam in 18 nsec and 0.25 cm^2 gives a shift of 18 ± 10 MHz at 488 nm. Finally, we add 25 MHz to our measurement because of the 50-MHz acousto-optic modulator red shift of the saturating beam used in the Te_2 spectroscopy. The sum of these systematic corrections is -23 ± 35 MHz, where the errors have been summed. The measured frequency difference between the fitted line center and the Te_2 calibration line is thus $74.5 \times (10.7 \pm 0.4) - 23 \pm 35 = 774 \pm 30 \pm 35$ MHz, where the first error is due to statistics, and the second to systematic effects. Thus, our measurement of $\frac{1}{4}$ of the frequency of the $F=1 \rightarrow 1$ transition of $613\,881\,924 \pm 30 \pm 35$ MHz is in reasonable agreement with the QED prediction of 613 881 989.90(90) MHz.

In summary, we have presented evidence for the laser excitation of muonium. Within the estimated experimental errors we see an excess number of counts occurring at the correct time and frequency windows expected for the laser ionization of the muonium atom. Clearly a more intense source of thermal muonium is necessary before a precision measurement of the $1S$-$2S$ interval is possible. Active work on muon moderators, improved muon-to-muonium conversion targets, and brighter pulsed muon sources is under way in several laboratories. The next-generation experiment could also use improved optics, a Fourier-transform–limited laser source, and a direct comparison of the hydrogen $1S$-$2S$ transition in the same apparatus to reduce the systematic effects mentioned above. Given such improvements, a high-resolution measurement of the $1S$-$2S$ transition should be possible.

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