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Interferometric frequency measurement of a $^{130}Te_2$ reference line for muonium $1S$-$2S$ spectroscopy

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We report on an interferometric frequency measurement of a $^{130}Te_2$ absorption line which can be used as a reference in Doppler-free two-photon spectroscopy of the muonium $1S$-$2S$ transition. The frequency of this 488-nm $^{130}Te_2$ transition is determined with a precision of 1 part in 10^9 by comparison with a previously calibrated $^{130}Te_2$ line near 486 nm.

Muonium is the bound state of a positive muon and an electron, and is one of the simplest systems available for precision tests of quantum electrodynamics (QED).[1] Calculations of its energy levels are free of the proton structure effects that plague hydrogen calculations and of the virtual annihilation terms that affect positronium energy levels. A particularly interesting experiment with muonium would be a measurement of the two-photon $1S$-$2S$ transition frequency. Doppler-free laser spectroscopy has already been used to make precision measurements of the $1S$-$2S$ intervals in hydrogen[2,3] and positronium.[4] The recent production of thermal muonium in vacuum[5] represents a first step in performing a muonium $1S$-$2S$ experiment. The natural linewidth of this transition is 150 kHz, limited by the 2.2-μs lifetime of the muon.

We report here on a precise interferometric measurement of the frequency of a Doppler-free transition near 488 nm in molecular tellurium vapor ($^{130}Te_2$) that can be used as a reference in Doppler-free two-photon spectroscopy of the muonium $1S$-$2S$ transition. Since the intense 244-nm radiation required for the muonium two-photon excitation is most easily produced through second harmonic generation, the 488-nm fundamental laser can be referenced to this $^{130}Te_2$ transition. This precise frequency calibration now permits a muonium experiment analogous to the precision measurements of the $1S$-$2S$ transition frequencies in hydrogen[2,3] and positronium,[4] which relied on two separate $^{130}Te_2$ reference lines near 486 nm for their frequency calibration. The absolute frequency of the hydrogen reference line was measured[6] with an accuracy of 4 parts in 10^{10} by comparison to an iodine-stabilized He-Ne laser at 633 nm. The positronium reference line was measured[4] relative to the deuterium $2S_{1/2}$-$4P_{3/2}$ Balmer component with an estimated uncertainty of 8 parts in 10^9, and has recently been measured to 1 part in 10^9 by interferometric comparison with the hydrogen reference line.[7]

Our measurement relies on the previous calibration[6] of the hydrogen reference line (component b_2 of Ref. 6), as well as a precise frequency standard in molecular iodine vapor ($^{127}I_2$) near 515 nm.[8] The muonium reference line, hereafter referred to as component d_4,[9] corresponds to the Doppler-broadened line labeled No. 865 in the $^{130}Te_2$ atlas[10] at 20476.8694(5) cm^{-1}. We find the frequency of the Doppler-free d_4 component to be 613881150.80(60) MHz. This line has a frequency 840 MHz lower than one-quarter of the expected $1S$-$2S$ $F=1$ transition frequency in muonium, and is the closest $^{130}Te_2$ component (at 488 nm) to that muonium transition. This precise reference line will be useful in a current experiment to measure the muonium $1S$-$2S$ transition frequency.[11]

We use Doppler-free saturated-absorption spectroscopy to observe the $^{130}Te_2$ b_2 and d_4 reference lines and the $^{127}I_2$ transition, which is hyperfine component a_3 of the rotational and vibrational transition 43-0 $P(13)$ at 515 nm. The frequency of the muonium reference line is measured interferometrically, by comparison with the hydrogen reference line b_2. The interferometer is calibrated by using the $^{130}Te_2$ b_2 line and the $^{127}I_2$ a_s line as known frequency standards.

The experimental apparatus is shown schematically in Fig. 1. Many of the experimental details are identical to those used in our measurement of the positronium reference line.[7] The second tellurium spectrometer of Ref. 7 has been replaced by an iodine saturation spectrometer, and we now use two marker interferometers. The dye laser is operated with coumarin 102 and produces continuous-wave radiation at 486, 488, and 515 nm.

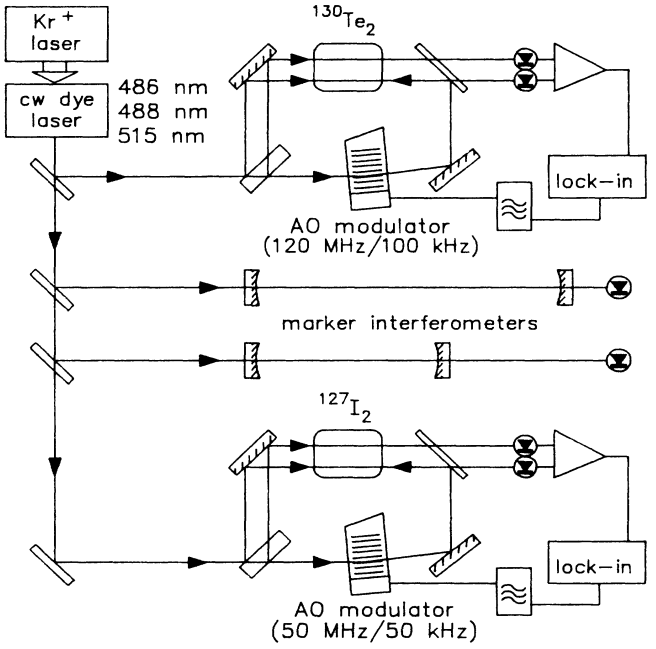

FIG. 1. Schematic of the experimental apparatus.

The $^{130}Te_2$ cell is maintained at the same temperature of 513(5) °C as in Ref. 7, which reproduces the conditions of the original b_2 calibration.[6] At this temperature the single-pass linear absorption on the d_4 component is 56(2)%.

The laser frequency is monitored with two confocal Fabry-Perot etalons, which are housed in a common evacuated, temperature-stabilized enclosure and have aluminum coated mirrors.[12] The free spectral ranges of these 1-m and 60-cm etalons are calibrated concurrently with our frequency measurement. The etalons are calibrated using a variation of the technique of exact fractions, as described by Goldsmith, Weber, Kowalski, and Schawlow.[12] We use the $^{130}Te_2$ b_2 component as one endpoint and the $^{127}I_2$ a_3 component as the other endpoint of the calibration interval.

The iodine saturation spectrometer is very similar to the tellurium one. The $^{127}I_2$ cell is used at ambient room temperature 21(6) °C corresponding to an iodine vapor pressure of 0.24(12) Torr. The pump beam intensity is modulated at 50 kHz with an acousto-optic modulator that also shifts the frequency of the beam upwards by 50 MHz. The pump and probe beams are each focused to points outside the cell. Within the cell, the pump beam diameter at the $1/e$ amplitude point varies from 0.43 to 0.62 mm and the probe beam diameter from 0.18 to 0.25 mm. The typical saturating beam power is 3.1 mW and the probe beam power is 1.8 mW.

Figure 2 shows a $^{130}Te_2$ spectrum in the vicinity of the muonium reference line. Also shown are the expected locations (at 488 nm) of the two hyperfine components of the muonium $1S$-$2S$ two-photon transition. The $^{130}Te_2$ d_4 component has a frequency approximately 2.6 THz lower than the b_2 component. The observed linewidth [full width at half maximum (FWHM)] of the d_4 component is 13.0(2) MHz. During each run, we record a total of 30–40 traces, alternating among the three lines and recording each line 3–5 times in succession. The data presented here represent three different runs.

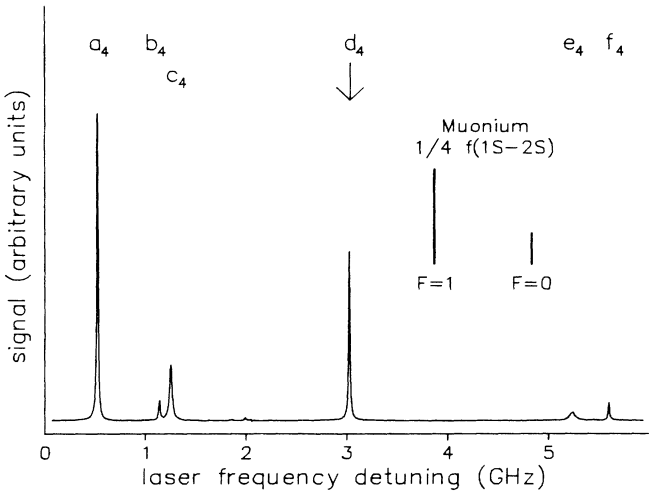

FIG. 2. Doppler-free spectrum of $^{130}Te_2$ at 488 nm in the vicinity of the muonium $1S$-$2S$ reference line d_4. The arrow indicates the d_4 component. Also shown are the expected locations (at 488 nm) of the hyperfine components of the muonium $1S$-$2S$ transition.

The saturation resonances and the etalon fringes are analyzed as described in Ref. 7. The $^{130}Te_2$ d_4 component has no neighboring lines and can be fitted without subtracting the background. The thermal drift of the length of each etalon was accounted for with a linear fit. Within each run, the fractional etalon order of each line was determined with a statistical error of 0.1%–0.2% of a fringe. The frequency separation between the d_4 and b_2 components is determined by the equation

$$f_{d_4}-f_{b_2}=(N_{d_4}-N_{b_2})\frac{f_{b_2}-f_{a_3}}{N_{b_2}-N_{a_3}}\ , \tag{1}$$

where N_i is the etalon order number (integer+fractional) for line i. The integer fringe orders of the observed transitions relative to one another are determined unambiguously by using the two independent etalons and relying on previous measurements that constrain the possible values of the etalons' lengths[7,12] and the frequency of the d_4 component.[10]

We apply two corrections to the raw data. The first is a trivial correction for the frequency shifts of the acousto-optic modulators in the saturation spectrometers. Each modulator shifts the frequency of the respective pump beam upwards, which causes the observed line centers to be red shifted by one-half of the modulator frequencies (see Fig. 1). This reduces the frequency interval between the observed $^{130}Te_2$ b_2 and $^{127}I_2$ a_3 components by 35 MHz, but does not affect the frequency interval between the $^{130}Te_2$ d_4 and b_2 components. We also correct the frequency of the $^{127}I_2$ a_3 component for a pressure shift, since the standard is defined at −5 °C, or 19 mTorr.[8] We rely on a previous measurement[13] of the pressure shift and apply a correction of −0.15 MHz.

Our results for the frequency separation of the muonium and hydrogen reference lines are summarized in Table I. The random uncertainties represent one standard deviation of the mean. Estimation of the systematic uncertainties is discussed below. We weight each of these six measurements by the inverse square of its total uncertainty, resulting in a measured frequency separation of

$$f_{d_4}-f_{b_2}=-2632745.50(54)\ \text{MHz}\ ,$$

which has a random uncertainty of 0.09 MHz and a sys-

TABLE I. Data summary of measurements of the frequency separation between the $^{130}Te_2$ b_2 and d_4 components. The random uncertainties are given by σ_R and the systematic uncertainties by σ_S. All values are given in MHz.

Etalon	Run	f_{d_4}-f_{b_2}	σ_R	σ_S
1 m	1	−2632745.54(48)	0.18	0.45
	2	−2632745.06(49)	0.20	0.45
	3	−2632745.62(48)	0.17	0.45
60 cm	1	−2632745.83(1.06)	0.26	1.02
	2	−2632746.05(1.06)	0.28	1.02
	3	−2632745.87(1.04)	0.15	1.02

tematic uncertainty of 0.53 MHz. To estimate these uncertainties, the random uncertainties are assumed to be uncorrelated between measurements, each systematic uncertainty is assumed to be totally correlated between different measurements, and different systematic uncertainties are assumed to be uncorrelated.[14]

The sources of systematic uncertainty in our measurement are summarized in Table II. Note that uncertainties in the frequency interval between the b_2 and a_3 components, as well as uncertainty in the position of the a_3 component are reduced by the ratio of the frequency interval between the d_4 and b_2 components to the interval between the b_2 and a_3 components ($\simeq \frac{1}{13}$). All errors listed in Table II are the total contributions from all three lines and represent one standard deviation. The etalon drift error arises from uncertainty in determining the thermal drift of the etalon. The line center of each of the saturation resonances is found by fitting the data to an asymmetric Lorentzian function over a range extending to three linewidths (FWHM) from the line center.[7] We investigate systematic errors due to this computer fitting procedure by varying the range over which each line is fitted for a subset of the data. From this, we conservatively estimate a systematic fitting uncertainty of 1% of the linewidth (FWHM) for each line, resulting in the errors shown in Table II.

Figure 3 shows the pressure shift of the d_4 component, as measured in separate experiments with the second tellurium spectrometer described in Ref. 7. The shift is measured with respect to the primary cell, which is kept at 513(5) °C or 0.89(11) Torr. A linear least-squares fit to the data shows that the muonium reference line is shifted by −0.84(12) MHz/Torr. The temperature uncertainty in the tellurium and iodine cells thus leads to a total pressure shift uncertainty of 0.15 MHz. The uncertainty of 0.25 MHz in each of the frequency standards[6,8] results in only a 0.03-MHz error for the measured frequency separation.

Dispersion in the reflective phase shift of the aluminum etalon mirrors[15] is not included in Eq. (1). An estimate of the possible size of this effect for our configuration is listed in Table II as a systematic uncertainty. The construction

TABLE II. Systematic uncertainties in the frequency separation measurement.

Source of uncertainty	Error[a] (MHz) 1-m etalon	60-cm etalon
Etalon drift	0.17	0.27
Computer fitting	0.24	0.24
Pressure shift	0.15	0.15
Frequency standards	0.03	0.03
Reflective phase shift	0.15	0.26
Transverse modes	0.20	0.54
Spherical aberration	0.15	0.71
Scan nonlinearity	0.06	0.18
Total	0.45	1.02

[a]One standard deviation.

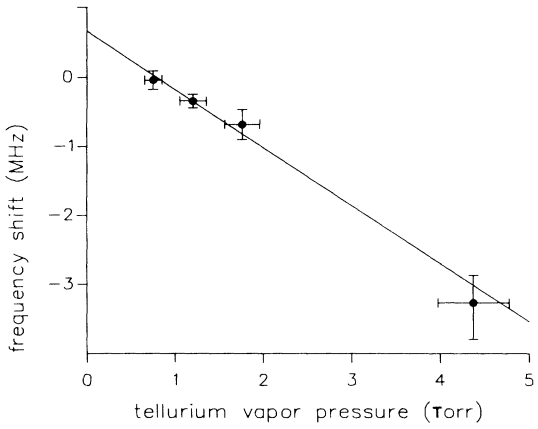

FIG. 3. Pressure shift of the d_4 component in a second $^{130}Te_2$ cell measured with respect to that of our primary cell [at 0.89(11) Torr].

of our etalons[12] precluded a virtual etalon measurement, which would avoid this problem. Equation (1) also neglects effects due to higher-order transverse modes and spherical aberration in the confocal etalons. We minimize these effects by aligning the etalons nearly on axis and using apertures to limit the possible departure of the beam from the etalon axis. The errors shown in Table II are estimates of the residual size of these effects, assuming the etalons are within 0.1 cm of the confocal position.[12,16] These estimates are consistent with earlier measurements of these effects with the same etalons.[12] The linearity of the laser frequency scans is investigated by comparing different fringe spacings within the same scan. These spacings exhibit random irregularities of approximately 1% of the free spectral range, due to index fluctuations and electronic noise. We use helium gas to pressure tune the dye laser and expect scan nonlinearity to contribute as much as 0.06 to 0.18 MHz for the 1-m and 60-cm etalons, respectively. We estimate the systematic uncertainty to be the full size of the expected effect.

Variation of the saturating power between 2.6 and 7.9 mW produced no observable shift of the $^{130}Te_2$ d_4 component. The b_2 component also exhibits negligible light shifts for the power levels used here.[6,7] We did not measure a light shift of the $^{127}I_2$ a_3 component, but expect it to be negligible.[17]

The frequency of the $^{130}Te_2$ b_2 component was previously measured[6] to be 616513896.30(25) MHz. When combined with our frequency separation measurement we find a value for the muonium reference line frequency of

$$f_{d_4} = 613881150.80(60) \text{ MHz} .$$

In a recent experiment,[3] the frequency of the hydrogen $1S$-$2S$ transition was measured with respect to the $^{130}Te_2$ b_2 component in the same tellurium cell used here. From that result, we determine the frequency separation between the muonium reference line and one-quarter of the hydrogen $1S$-$2S$ $F=1$ transition frequency to be

$$f_{d_4} - \tfrac{1}{4} f_H(1S\text{-}2S)_{F=1} = -2634124.97(61) \text{ MHz} .$$

This value can now be used to make a precise comparison between the $1S$-$2S$ transition frequencies in muonium and hydrogen. Such a comparison will be especially important in the future because it permits a more precise test of quantum electrodynamics than is possible with absolute frequency measurements, which are limited by uncertainties in the Rydberg constant and other optical frequency standards.

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[1]C. J. Oram *et al.*, Phys. Rev. Lett. **52**, 910 (1984); A. Badertscher *et al.*, *ibid.* **52**, 914 (1984).

[2]E. A. Hildum, U. Boesl, D. H. McIntyre, R. G. Beausoleil, and T. W. Hänsch, Phys. Rev. Lett. **56**, 576 (1986); J. R. M. Barr, J. M. Girkin, J. M. Tolchard, and A. I. Ferguson, *ibid.* **56**, 580 (1986).

[3]R. G. Beausoleil, D. H. McIntyre, C. J. Foot, E. A. Hildum, B. Couillaud, and T. W. Hänsch, Phys. Rev. A **35**, 4878 (1987).

[4]S. Chu, A. P. Mills, Jr., and J. L. Hall, Phys. Rev. Lett. **52**, 1689 (1984).

[5]A. P. Mills, Jr. *et al.*, Phys. Rev. Lett. **56**, 1463 (1986); G. A. Beer *et al.*, *ibid.* **57**, 671 (1986).

[6]J. R. M. Barr, J. M. Girkin, A. I. Ferguson, G. P. Barwood, P. Gill, W. R. C. Rowley, and R. C. Thompson, Opt. Commun. **54**, 217 (1985).

[7]D. H. McIntyre and T. W. Hänsch, Phys. Rev. A **34**, 4504 (1986).

[8]"Documents concerning the new definition of the metre," Metrologia **19**, 163 (1984).

[9]This labeling is arbitrary, but consistent with Ref. 6.

[10]J. Cariou and P. Luc, *Atlas du spectre d'absorption de la molecule de tellure* (Laboratoire Aime-Cotton, CNRS, Orsay, France, 1980).

[11]S. Chu (private communication).

[12]J. E. M. Goldsmith, E. W. Weber, F. V. Kowalski, and A. L. Schawlow, Appl. Opt. **18**, 1983 (1979).

[13]F. Spieweck, G. Camy, and P. Gill, Appl. Phys. **22**, 111 (1980).

[14]S. R. Lundeen and F. M. Pipkin, Metrologia **22**, 9 (1986).

[15]J. M. Bennet, J. Opt. Soc. Am. **54**, 612 (1964).

[16]M. Hercher, Appl. Opt. **7**, 951 (1968).

[17]F. Spieweck, IEEE Trans. Instrum. Meas. **IM-29**, 361 (1980).