Absolute Optical Frequency Measurement of the Cesium D_1 Line with a Mode-Locked Laser

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We have measured the absolute optical frequency of the cesium D_1 line at 335 THz (895 nm). This frequency provides an important link for a new determination of the fine structure constant α. The D_1 line has been compared with the fourth harmonic of a methane stabilized He-Ne laser at 88.4 THz (3.39 μm). To measure the frequency mismatch of 18.39 THz between 4×88.4 THz $= 354$ THz and the D_1 line a frequency comb spanning around 244 000 modes of a Kerr-lens mode-locked laser was used. We find 1 167 688 (81) kHz for the hyperfine splitting of the $6P_{1/2}$ state and 335 116 048 807 (41) kHz for the hyperfine centroid from which we derive $\alpha^{-1} = 137.035\,992\,4(41)$. [S0031-9007(99)09057-2]

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The cesium atom serves as the primary standard of time, and it plays an important role in many experiments on laser cooling, trapping, and atom interferometry. Using a new approach to measure the frequency of light with femtosecond laser pulses we have determined the absolute frequency f_{D1} of the cesium D_1 resonance line near 895 nm to 1.2 parts in 10^{10}, surpassing the accuracy of the best previous measurements by almost 3 orders of magnitude [1]. This frequency is of particular interest for a new determination of the fine structure constant α taking advantage of the extremely well-known Rydberg constant $R_\infty = \alpha^2 c m_e/2h$ [2]. Chu and collaborators at Stanford are measuring the photon recoil shift of the D_1 line $f_{\mathrm{rec}} = f_{D1}^2 h/2m_{\mathrm{CS}}c^2$ with an atom interferometer aiming for an accuracy near the ppb (parts per 10^9) level [3]. Together with the proton-electron mass ratio m_p/m_e, which is known to 2×10^{-9} [4], and even more precise measurements of the cesium to proton mass ratio m_{CS}/m_p, which have been reported recently [5], our frequency measurement provides the missing link for the determination of the fine structure constant according to

$$\alpha^2 = \frac{2R_\infty}{c}\,\frac{h}{m_e} = 2R_\infty\,\frac{2f_{\mathrm{rec}}\,c}{f_{D1}^2}\,\frac{m_{\mathrm{CS}}}{m_p}\,\frac{m_p}{m_e}\,. \qquad (1)$$

Such a new determination of the fine structure constant is highly desirable in the light of persisting discrepancies between the values obtained so far by different methods [6].

In our experiment we compare the frequency of the cesium D_1 resonance line with the fourth harmonic of a transportable CH_4-stabilized 3.39 μm He-Ne laser [7]. The frequency of this reference laser has been calibrated at the Physikalisch Technische Bundesanstalt (PTB) in Braunschweig against a microwave cesium atomic clock with their phase-locked harmonic laser frequency chain [8] to within 2.6 parts in 10^{13}. To bridge the large frequency gap between the fourth harmonic near 848 nm and the 895 nm Cs D_1 line, we employ a Kerr-lens mode-locked Ti:sapphire laser as a precise active frequency comb generator, realizing, for the first time to our knowledge, a phase-coherent optical frequency measurement with ultrashort light pulses.

It has long been recognized [9] that the periodic pulse train of a mode-locked cw laser can be described in the frequency domain as a comb of equidistant longitudinal laser modes separated by the pulse repetition rate f_r. For an ideal laser emitting a periodic train of pulses of identical waveform the output can be described in the frequency domain by a Fourier series. The elements of this series correspond to the longitudinal laser modes whose frequencies would thus be high harmonics of the pulse repetition rate f_r. In a real laser the optical phase is shifted, by say $\Delta\varphi$, from pulse to pulse with respect to the envelope [10]. This is because the group velocity, which determines the round trip time, is generally not identical with the phase velocity of the pulse carrier frequency. However, if $\Delta\varphi$ stays constant, we still expect a comb of equidistant mode frequencies, spaced by the repetition frequency f_r, but with each frequency displaced from a multiple nf_r by an offset frequency $f_o = \Delta\varphi/2\pi T$, where $T = f_r^{-1}$ is the pulse repetition time. Recently we have tested the uniform distribution of the modes of such a frequency comb and showed that the mode separation agrees with the pulse repetition rate within an experimental uncertainty of a few parts in 10^{17} [11].

This test and the experiment described here have been carried out with a commercial Kerr-lens mode-locked Ti:sapphire laser (Coherent Mira 900, 73 fs $\approx 1.5 \times$ Fourier limited pulses, 75 MHz repetition rate), pumped by a 5 W diode-pumped frequency-doubled single-frequency Nd:YVO_4 laser (Coherent Verdi). With this laser we have bridged a 20 THz frequency gap [=2.9 times the spectral width (FWHM)] with a millihertz precision. This is to our knowledge the largest frequency gap measured with a frequency comb. Even broader frequency combs can be created with shorter pulses and/or by four wave mixing in optical fibers [12,13].

The experiment, which is sketched in Fig. 1, is operated as follows: First the color center laser is phase locked to

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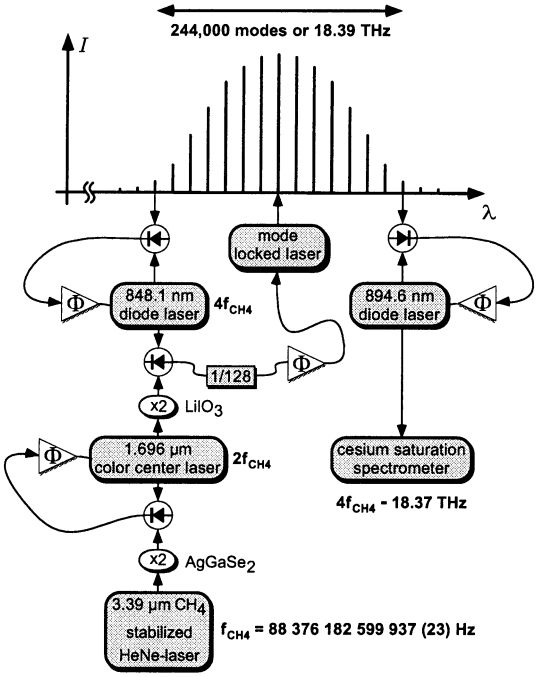

FIG. 1. Frequency chain that allows the comparison of the precisely known frequency of a methane stabilized He-Ne laser at 88.4 THz (3.39 μm) with the cesium D_1 transition at 335 THz (895 nm).

the second harmonic of the He-Ne standard by controlling the beat frequency between them with an electronic phase-locked loop. This forces the beat frequency to oscillate in phase [14] with a reference frequency of a few MHz (not shown in the figure) provided by a cesium atomic clock. The frequency of the color center laser is then known as precisely as the He-Ne standard. Next two grating-stabilized laser diodes [15] at 848 and 895 nm are locked to two suitable modes of the frequency comb after their absolute frequencies have been checked coarsely with a wave meter. As described below, the number of modes between the laser diodes is controlled to be an integer multiple of 20. A low noise beat signal (signal to noise ratio >30 dB in 400 kHz resolution bandwidth), necessary for phase locking, is created with the help of a grating that preselects some of the modes in the vicinity of the laser diode frequency. From the measured spectral intensity of the frequency comb we estimate the power of the modes in use to 40 nW. The mode to which the 848 nm laser is phase locked is stabilized to the second harmonic of the color center laser (fourth harmonic of the He-Ne standard). This is done with the help of a piezomounted folding mirror of the mode-locked laser that controls the beat frequency between the 848 nm laser diode and the second harmonic of the color center laser. Because of the rather low servo bandwidth of about 10 kHz at this stage we use a large range ($\pm 32\pi$) digital phase detector [14], as we do for locking the diode lasers, and a digital by 128 divider that increases the maximum possible phase fluctuations by a factor of 128. We continuously monitor the in-lock beat signals for possible lost cycles with additional counters operated at a different bandwidth, discarding data points that are off by more than 0.5 Hz from the given reference frequency. In the phase-locked condition the frequency of the laser that probes the cesium transition f_{895} is related to other known frequencies by

$$f_{895} = 4f_{\mathrm{HeNe}} - nf_r \pm 128f_{\mathrm{ref}} - 280\ \mathrm{MHz}, \quad (2)$$

where $f_{\mathrm{HeNe}} = 88\,376\,182\,599\,937(23)$ Hz denotes the frequency of the methane stabilized He-Ne laser. By changing the reference frequency f_{ref} used for phase locking the 848 nm laser diode, we scan the frequency of the 895 nm laser diode. Exchanging the input signals of the phase detector allows the use of both signs in Eq. (2). The offset of 280 MHz is due to the remaining reference frequencies used for phase locking. The pulse repetition rate f_r of the free running laser is measured with a radio frequency counter (signal to noise ratio >60 dB in a resolution bandwidth of 400 kHz).

The cesium D_1 line was observed in a 7.5 cm long cell (Opthos Instruments Inc.) at room temperature. Several cells have been used in another experiment [16] where we measured the cesium D_2 transition frequency without noticeable frequency differences. To probe the cesium D_1 transition we use a saturation spectrometer with two linearly polarized counterpropagating laser beams with equal intensities (10 μW/cm^2). The pump beam redistributes the occupation number mainly by pumping the atoms to the other ground state hyperfine level so that the absorption of the probe beam is decreased. By chopping the pump beam the difference in absorption is detected with a lock-in amplifier. Because one beam can change the absorption of the other only through atoms whose Doppler shift is the same for both beams we detect only atoms that do not move along the laser beam axis. With this Doppler-free method we observe four hyperfine components of the single stable isotope ^{133}Cs for the transitions from the ground states $F_g = 3$ and $F_g = 4$ to the upper states $F_e = 3$ and $F_e = 4$. The observed linewidth of about 6 MHz (FWHM) was somewhat larger than the natural linewidth of 5 MHz. This is believed to be caused by the short term frequency fluctuations of the phase-locked lasers, allowed by the large range phase detectors. The crossover resonances were not visible due to the large separation of the excited state hyperfine components. Stray magnetic fields are reduced by a double cylindrical μ-metal shielding to values below 2 μT along the laser beam axis. The transversal field component has not been measured but is assumed to be much smaller. We find the line center of the resonances by fitting a Lorentzian with a linear background to it as shown in Fig. 2.

In order to obtain the correct frequency f_{895} the exact number of modes n between the two phase-locked laser diodes at 848 and 895 nm has to be determined. We could neither unambiguously identify the modes with our

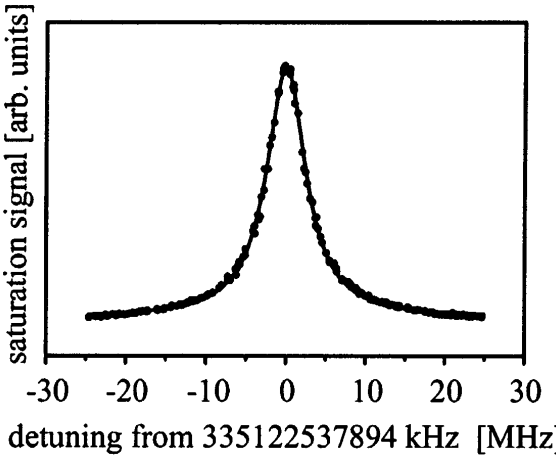

FIG. 2. The $F_g = 4 \rightarrow F_e = 4$ component of the cesium D_1 transition with fitted Lorentzian obtained from the saturation spectrometer. The absolute optical frequency is determined from Eq. (2).

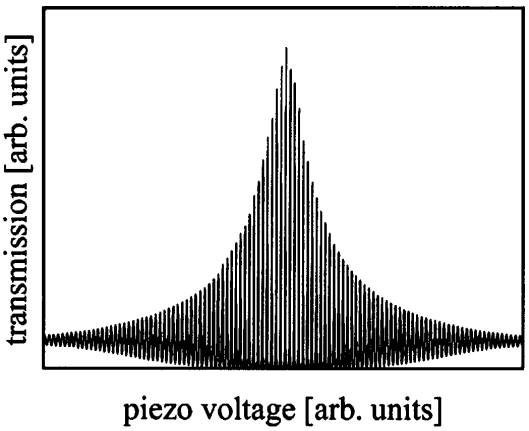

FIG. 3. The transmitted light from a mode locked laser through a cavity of finesse F as a function of the piezo supply voltage that controls the mirror separation. Far detuned the cavity will transmit one out of $\pi/2F \approx 0.005$ modes giving rise to the constant background. At the central peak the ratio of the pulse repetition rate and the free spectral range of the cavity is exactly 1:20.

wave meter nor did we want to rely solely on previous measurements in order to exclude the $f_r \approx 75$ MHz ambiguity. The observation of a frequency shift $n \times \delta f_r$ of one of the modes after changing f_r by δf_r may be difficult because it demands a resolution of δf_r to distinguish between the mode number n and $n \pm 1$. The shift $n \times \delta f_r$ could not be chosen too large if one has to track it with a phase-locked diode laser. To uniquely determine n of the order of 244 000 we employ a cavity that is stabilized to have a free spectral range of exactly 20 times the pulse repetition rate. On resonance this cavity transmits every twentieth mode of the frequency comb increasing the pulse repetition rate by a factor of 20. Then the beat signals between the frequency comb and one of the laser diodes are no longer separated by $f_r \approx 75$ MHz but by $20 f_r \approx 1.5$ GHz sufficient to be unambiguously identified by our wave meter. To make sure that the free spectral range of the cavity is indeed 20 times the pulse repetition rate we observe the transmission through the cavity as shown in Fig. 3. If the fraction of the free spectral range and the pulse repetition rate is a small integer such as 1:20 resonant fringes appear. A peak in the transmission appears if the piezocontrolled mirror spacing allows the transmission of every twentieth mode and the transmission vanishes if these modes are blocked [17]. Higher order ratios produce fringes with smaller contrast. We found the correct fringes by verifying the 1.5 GHz separation of the beat signals. To validate our mode number counting scheme we have phase locked two laser diodes 26 500 modes apart and checked the resulting frequency difference with an optical frequency comb generator [18]. In the future a somewhat larger mode spacing may allow the identification with a common laser wave meter.

We expected some spurious shifts of the transition lines due to the fact that the readout of the lock-in amplifier with a time constant of $\approx$0.1 sec is delayed with respect to the optical frequency measurement according to Eq. (2). This shift depends on the speed and the direction of the frequency scan. We have recorded all four hyperfine components several times with six different scanning speeds and fitted a straight line to the obtained center frequencies as shown in Fig. 4. The ground state hyperfine splitting as calculated from our result is 36 kHz for $F_e = 3$ and 27 kHz for $F_e = 4$ too large compared to the definition of the SI second. We attribute this deviation to systematic uncertainties due to imperfections of the magnetic field shielding. Because of different gyromagnetic ratios Zeeman shifts are present depending on the Zeeman level that is mostly depleted by the pump beam. Imperfections of the polarization of the beams and small polarization modulations caused by the chopped pump beam may lead to unsymmetric pumping of the M_F levels. In the worst case the $M_F = 4 \leftrightarrow M_F = 3$ component is probed leading to a Zeeman shift of

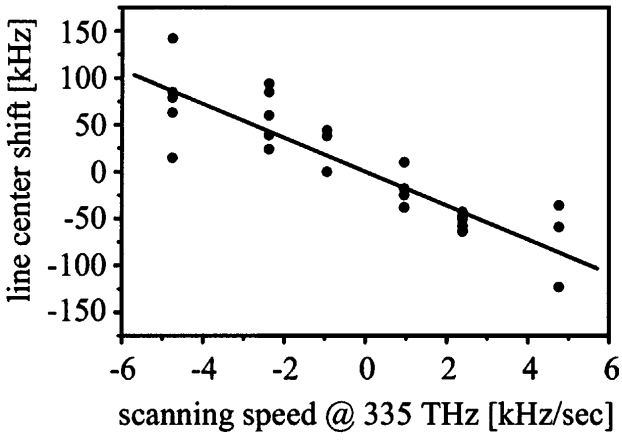

FIG. 4. Observed line center of the $F_g = 4 \rightarrow F_e = 4$ transition as a function of the laser frequency scanning speed.

40 kHz where the sign depends on the nature of the polarization imperfections. We expect smaller shifts for transitions connecting the same total angular momentum than for transitions between $F = 4$ and $F = 3$ because the difference of the gyromagnetic ratios is smaller and mostly unshifted $M_F = 0$ components will be probed. We have corrected for this systematic effect by adding 36 kHz to the $F_g = 4 \rightarrow F_e = 3$ and subtracting 27 kHz from the $F_g = 3 \rightarrow F_e = 4$ transition forcing the ground state hyperfine splitting to the defined value and obtained the following from a total of 112 recorded lines:

$$f_{D1}^{F_g=3\rightarrow F_e=3} = 335\,120\,562\,838 \text{ kHz} \quad (3)$$

$$f_{D1}^{F_g=3\rightarrow F_e=4} = 335\,121\,730\,526 \text{ kHz} \quad (4)$$

$$f_{D1}^{F_g=4\rightarrow F_e=3} = 335\,111\,370\,206 \text{ kHz} \quad (5)$$

$$f_{D1}^{F_g=4\rightarrow F_e=4} = 335\,112\,537\,894 \text{ kHz}. \quad (6)$$

This procedure does not alter our result of the hyperfine centroid but the resulting excited state hyperfine splitting. The systematic uncertainty is estimated by the maximum expected Zeeman shift of 40 kHz for the absolute frequencies and twice that value (systematic uncertainties may add up) for the upper state hyperfine splitting. Other systematic effects such as the ac-Stark effect, light pressure induced line-shape modifications [19], spurious selective reflection signals, and collisional shifts [20] are estimated to be much smaller. The statistical uncertainty is around 10 kHz. Our results for the hyperfine centroid

$$f_{D1} = 335\,116\,048\,807(41) \text{ kHz} \quad (7)$$

and for the upper state hyperfine splitting

$$f_{6P_{1/2}}^{\mathrm{HFS}} = 1\,167\,688(81) \text{ kHz} \quad (8)$$

are in good agreement with previous values obtained by others $f_{D1} = 335\,116\,062(15)$ MHz [1] and $f_{6P_{1/2}}^{\mathrm{HFS}} = 1\,167.54(32)$ MHz [21]. The uncertainties of the frequencies given in Eq. (6) are estimated in the same way to be 41 kHz. With the electron proton mass ratio [4], preliminary measurements of the recoil shift f_{rec} [22], and the cesium atomic mass [5], a new value for the fine structure constant $\alpha^{-1} = 137.035\,992\,4(41)$ is obtained from Eq. (1) and the precisely known Rydberg constant [2]. This value disagrees by 1.8 combined standard deviations from the current most precise value obtained from the electron gyromagnetic ratio [6].

After completion of the reported experiments we have succeeded in phase locking a selected mode of our femtosecond laser to the fourth harmonic of the CH_4-stabilized He-Ne laser while simultaneously controlling the pulse repetition rate with a cesium atomic clock. In this way we are generating a comb of several hundred thousand reference frequencies, each known to a few parts in 10^{13}. The frequency f_n of any longitudinal mode can be tuned as in a cw laser by changing the optical cavity length with a piezomounted mirror. To control the pulse repetition rate f_r and thus the comb spacing independently, we use a second piezotransducer to tilt the cavity end mirror near one of the prisms for compensation of the group velocity dispersion. We thus introduce an additional phase shift proportional to the frequency distance from f_n which displaces the pulse in time and thus changes the effective cavity roundtrip time.

In the near future, we will compare the absolute frequency f of a laser directly with the microwave frequency of a cesium atomic clock by measuring the interval between f and the second harmonic $2f$. Two or three cascaded frequency interval divider stages can reduce this interval sufficiently so that it can be bridged with the frequency comb of a femtosecond laser. In this way, the large and complex laser frequency chains of the past can be replaced with versatile, compact, and reliable solid-state systems for the measurement and synthesis of optical frequencies.

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