First Phase-Coherent Frequency Measurement of Visible Radiation

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We have determined the frequency of the $^3P_1$-$^1S_0$ intercombination transition of atomic ^{40}Ca stored in a magneto-optical trap to be $\nu = 455\,986\,240\,493.95$ kHz with an estimated standard uncertainty of 0.43 kHz ($\delta\nu/\nu < 10^{-12}$) using a phase-coherent optical frequency chain from the Cs atomic clock to the visible. This allows the realization of the SI-unit meter according to its definition by visible radiation with 25-fold reduced uncertainty compared to previous measurements.

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The precise measurement of frequencies in the visible part of the electromagnetic spectrum is of fundamental importance for basic research and applied metrology: Precision frequency measurements are required, for example, for the determination of fundamental constants such as the Rydberg constant [1,2], allowing one to check the validity of the theory of quantum electrodynamics. They furthermore allow one to address the question of possible time variations of fundamental constants [3,4]. In the field of metrology they provide the base for the development of atomic clocks with transitions in the optical range [5] and the realization of the SI base unit, the meter [6], with unprecedented low uncertainty.

Frequency standards operating in the visible range are advantageous in providing extremely high line quality factors $Q = \nu/\delta\nu$ and offer low values of the frequency instability. Furthermore, the meter is preferably realized with standards of visible radiation to allow easy use of interferometric intercomparisons.

Like any other frequency standard, optical frequency standards have to be related to the primary standard of time and frequency, the Cs atomic clock. In order to avoid uncertainties additionally to those of the Cs clock and of the standard in the visible, the large gap of frequencies (frequency ratio $\approx$50 000) has to be bridged by a phase-coherent measurement.

So far, absolute phase-coherent frequency measurements of near infrared radiations up to 88 THz have been performed [7,8]. In the visible, the frequency of the well known iodine-stabilized HeNe laser ($\lambda = 633$ nm, $\nu = 473$ THz) was determined as early as 1983 [9]. A more recent measurement starting from a CO_2 laser at 29 THz [10] led to a reduced relative uncertainty of 2.5×10^{-11} [11]. The Rydberg constant has been determined independently with a relative uncertainty below 2×10^{-11} using frequency measurement chains starting at 473 THz [2,10] and 88 THz [1]. The frequency measurements of visible radiation were not based directly on the Cs atomic clock so far, and are thus ultimately limited by the reproducibility of the reference frequency standards.

We report on the first phase-coherent frequency measurement of an optical frequency standard in the visible. The optical frequency standard comprised a laser whose frequency was stabilized to the intercombination transition $^3P_1$-$^1S_0$ of ^{40}Ca atoms. This transition is an excellent reference for an optical frequency standard, because it has a natural linewidth of only 400 Hz with no hyperfine splitting. Furthermore, for the transitions with $\Delta m_j = 0$, the frequency shows only a small quadratic dependence on electric and magnetic fields of 30 MHz/(V/cm)^2 and 10^8 Hz/T^2, respectively [12,13]. Consequently, this transition was recommended by the Comité International des Poids et Mesures (CIPM) for the realization of the meter [11]. Up to now, this frequency has been determined by interferometric wavelength comparisons only [14,15]. We used a high resolution dye laser spectrometer [16] with its frequency stabilized to the intercombination line $^3P_1$-$^1S_0$ of an effusive Ca atomic beam. The transit time broadening was reduced below 10 kHz by optical Ramsey excitation in a four running-wave configuration [17]. For stabilizing the laser frequency to the central fringe of either recoil component, the laser frequency was square wave modulated (modulated frequency = 0.5 Hz, modulated width = 5 kHz), and the method of first harmonic detection was used.

The phase-coherent frequency chain connects the frequency of the Ca stabilized dye laser with the frequency of the Cs atomic clock (Fig. 1). The chain allows one to measure simultaneously the frequencies of the CH_4-stabilized HeNe laser at 3.39 μm, the OsO_4-stabilized CO_2 laser at 10.6 μm, and the Ca frequency standard at 0.657 μm. Basically, the frequency measurement utilizes a chain of several intermediate oscillators whose frequencies are phase coherently compared to each other by harmonic mixing. All reference frequencies used in the chain are derived from the Cs atomic clock. The schematics of the frequency chain and the phase-coherent frequency measurement have been described in detail elsewhere [18,19]. The upper part of the frequency chain is downlocked from the Ca optical frequency standard to the color-center laser (CCL). The lower part is locked from the 100 MHz standard frequency output of a hydrogen maser up to the methanol laser at 4 THz. The intermediate part of the chain, consisting of all CO_2-lasers, is locked to a methane stabilized HeNe laser

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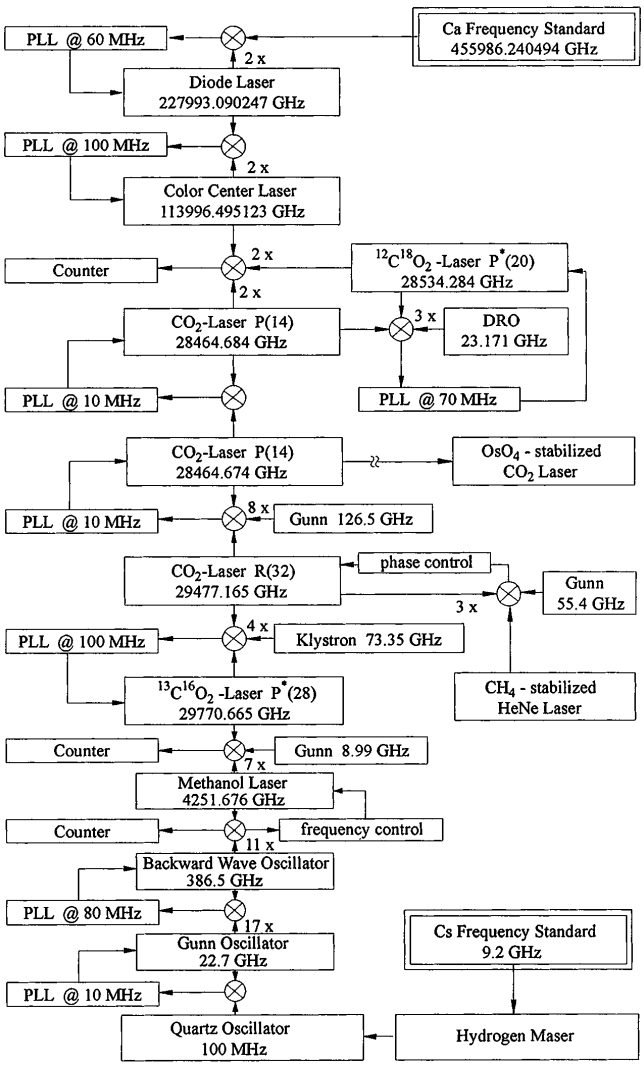

FIG. 1. PTB's frequency chain to the Ca intercombination line (PLL = phase locked loop, details are given in the text).

to improve the frequency stability. To obtain the value of the Ca transition frequency, we simultaneously counted the beat signals of the methanol laser with the backward wave oscillator (BWO) and the CO_2 laser, and the beat signal of the two CO_2 lasers with the CCL using totalizing counters. Combining the beat frequencies as described earlier [18] yields a frequency independent of fluctuations of the intermediate transfer oscillators. This method allows us to track the phase of all intermediate oscillators and therefore leads to a truly phase-coherent measurement.

We have operated the frequency measurement chain in the phase-coherent mode for an integrated time of about three hours. The dye laser was subsequently stabilized to the high- and low-frequency recoil component of the Ca atoms in the effusive beam. Potential frequency shifts due to optical phase errors were largely compensated by the method of laser beam reversal [17]. The frequency of the laser stabilized to the effusive beam deviates from the frequency of the intercombination transition of an unperturbed Ca atom by a few parts in 10^{12}. This frequency offset is due mainly to the second-order and a small residual first-order Doppler effect. Their contributions to the uncertainty can be substantially reduced by additionally using laser cooled and trapped Ca atoms [13]. We have therefore split off part of the laser beam and stabilized its frequency to the absorption line provided by some 10^6 Ca atoms stored in a magneto-optical trap (MOT) [20]. Trapping of the atoms and probing of the clock transition was performed alternately. The error signal for the stabilization was derived from optical Ramsey fringes obtained by time separated field excitation [13]. The frequency of the exciting laser beam was controlled by an acousto-optic modulator. The frequency offset between the effusive beam and the stored atoms was measured simultaneously during the optical frequency measurement. Figure 2 shows the frequency data obtained while the laser was stabilized to the low frequency recoil component during a period of approximately 30 min. The lower and upper curves of Fig. 2 represent the measured frequency values for the effusive beam and the atoms at rest, trapped in the MOT, respectively.

The distribution of each data set is Gaussian with a FWHM of approximately 900 Hz. The statistical part of the uncertainty of the frequency measurement presented here was of the order of 20 Hz. The frequency difference between the frequency of the thermal atoms and the atoms stored in the MOT (≈ -3.4 kHz) is considerably higher than expected from the contribution of the second-order Doppler effect [$\Delta\nu_D = -\frac{1}{2}\nu_0(v/c)^2 \leq -2$ kHz]. Since the fields of the trapping laser of the MOT were not completely shut off during the time of the frequency measurement, the atoms in the MOT experienced a small ac Stark shift. This shift has been determined a few days after the frequency measurement to be 0.84 kHz. The

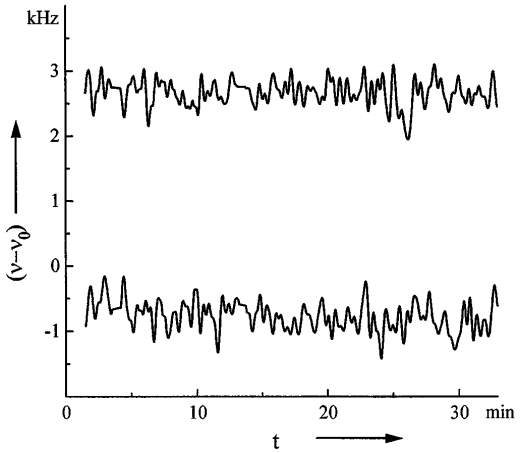

FIG. 2. Frequency of the laser stabilized to the low frequency recoil component of atoms in an effusive beam (lower curve) and of atoms in a MOT (upper curve). ν_0 was arbitrarily chosen to be 455 986 240 480.50 kHz.

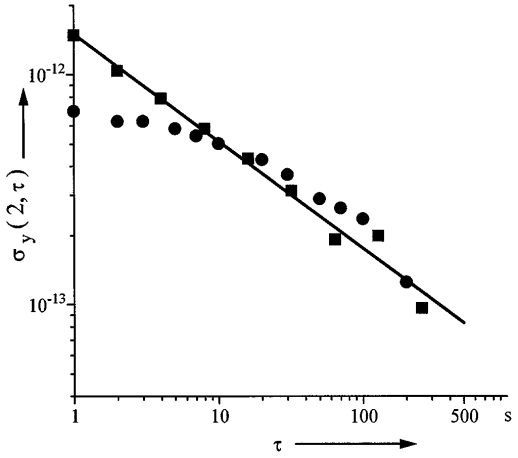

FIG. 3. Allan standard deviation $\sigma_y(2,\tau)$ of the measured frequency versus the integration time (dots: effusive beam, squares: trapped atoms) together with a fit for the data of the MOT (straight line).

remaining difference is attributed to a residual first-order Doppler shift in the thermal beam.

Figure 3 shows the Allan standard deviation of the measured laser frequency (e.g., Fig. 2) stabilized to both the effusive beam (dots) and the MOT (squares) together with a fit of the data of the MOT (straight line) as a function of the integration time. The Allan standard deviation of the MOT is dominated by white frequency noise as can be derived from the fact that it is proportional to $\tau^{-1/2}$. The instability is limited by the signal-to-noise ratio of the trapped atoms ($S/N \approx 10$) and the selected spectral resolution ($\delta\nu \approx 10$ kHz) given by the pulse separation. An estimation of the fractional frequency instability $\Delta\nu/\nu \approx \delta\nu/(\nu S/N)$ yields 2.2×10^{-12} for an integration time of one second, which agrees well with the fitted value of 1.6×10^{-12}. The mean of the high- and low-frequency recoil components is 455 986 240 493.95 kHz with an estimated standard uncertainty of 0.43 kHz, which we shall discuss in the following.

The low velocity of the Ca atoms stored in the MOT allows, in principle, one to reduce the contributions to the relative standard uncertainty due to the first- and second-order Doppler effect below 10^{-14} [13]. After switching off the trapping laser and magnetic fields, the atomic cloud is expanding, and the atoms are accelerated due to gravity and possible transients of the trapping fields during the switching process. If the exciting laser beam is reflected under a small angle α, a residual first-order Doppler shift $\Delta\nu_D \approx \nu_0(v/c)\alpha$ may occur. The acceleration due to gravity leads to a velocity $v < 1$ cm/s during the maximum elapsed time of 1 ms. With α controlled to be ≤ 0.3 mrad this effect contributes to less than $10^{-14}\nu_0$. Collisions of the cold atoms represent another possible source of systematic frequency shifts of the trapped atoms. With at most 10^7 atoms stored in about 1 mm^3 the distance between the Ca atoms is about 5 μm, which is large compared to the wavelength of the exciting radiation ($\lambda \approx 0.657$ μm) and the diameter of the Ca wave packet. Even though this effect might eventually limit the accuracy of the Ca clock it is not expected to contribute on the order of 100 Hz. A third possibility of potential frequency shifts results from the fact that both recoil components are excited in our experiment which may lead to an intensity dependent frequency pulling [21]. We have varied the intensity (and at the same time the pulse length), and we did not see an effect within an uncertainty of about 100 Hz. Since this shift is expected to be antisymmetric with respect to the mean frequency, it should be largely eliminated in our frequency value which corresponds to the mean frequency of both recoil components. The possible occurrence of cycle slips in the frequency chain represents another source of error of the frequency measurement. These cycle slips depend critically on the signal-to-noise ratio of the relevant beat signals [22]. We estimated the rate of cycle slips in the most crucial stages of the chain and conclude that the resulting errors can be neglected at the present level of uncertainty. In our experiment the uncertainty was therefore dominated by the residual light shift of the trapping laser radiation (0.84 kHz). Taking into account possible variations of the light shift between its determination and the earlier frequency measurement, we estimate the standard uncertainty to be close to one-half of the light shift (0.4 kHz).

In conclusion, we have performed the first phase-coherent frequency measurement of visible radiation referenced to the primary standard of time and frequency, the Cs clock. We have stabilized a dye laser to the Ca intercombination line and determined the absolute frequency of this transition with a standard uncertainty of 0.43 kHz ($\delta\nu/\nu < 10^{-12}$). Comparing our results with the adopted value of the uncertainty of the Ca transition frequency by the CIPM ($\sigma = 4.5 \times 10^{-10}$) [11], we have reduced this uncertainty 500-fold. The Ca stabilized dye laser now represents the optical frequency standard with the lowest uncertainty in the visible. The uncertainty is more than an order of magnitude smaller than that of the previous most accurate standard, the iodine stabilized HeNe laser at $\nu = 473\,612\,214\,705$ kHz $[(1 \pm 2.5) \times 10^{-11}]$ [11]. Extending the phase-coherent frequency measurement to the MOT and avoiding light shifts due to the trapping laser fields, we soon expect a further reduction of the uncertainty by at least one order of magnitude. We have established a frequency standard in the visible, limited only by the uncertainty of both the primary standard of time and frequency, and of the optical frequency standard itself. This allows one to investigate the frequency ratio between the Cs clock and the Ca frequency standard over a long period of time. Since different contributions of fundamental constants are responsible for the electronic and hyperfine transitions of Ca and Cs, respectively, possible time variations of fundamental constants [4] can by now be performed with improved accuracy.

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