

Journal: Measurement Science and Technology

Topical Review

Optical frequency standards for time and length applications

Feng-Lei Hong^{1,2,3}

¹ Department of Physics, Graduate School of Engineering, Yokohama National University, Yokohama 240-8501, Japan

² National Metrology Institute of Japan (NMIJ), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8563, Japan

³ JST, ERATO, MINOSHIMA Intelligent Optical Synthesizer Project, Tsukuba, Ibaraki 305-8563, Japan

E-mail: hong-fl@ynu.ac.jp

Abstract

The last decade has witnessed tremendous progress in research on optical frequency metrology. Optical frequency standards using optical lattice and single-ion trap technologies have reached levels of stability and accuracy that surpass the performance of the best Cs fountain atomic clocks by orders of magnitude. Optical frequency standards are also used for various applications including length metrology. Optical frequency measurement and links using optical frequency combs and optical fibres play important roles in the development of optical frequency standards. This article introduces optical frequency standards recommended by the International Committee for Weights and Measures (CIPM) along with updates provided by recent research results. Frequency ratio measurements and remote frequency comparisons are addressed in relation to the work whose goal is to redefine the second. Optical frequency standard and optical frequency comb applications are also described.

Keywords: optical frequency standard, optical clock, optical lattice clock,

single-ion clock, wavelength standard, optical frequency measurement, optical frequency comb

(Some figures may appear in colour only in the online journal)



This is an open access article distributed under the terms of the Creative Commons Attribution Non-Commercial No Derivatives 4.0 License (CC BY-NC-ND, <http://creativecommons.org/licenses/by-nc-nd/4.0/>), which permits non-commercial reuse, distribution, and reproduction in any medium, provided the original work is not changed in any way and is properly cited.

1. Introduction

Optical frequency metrology [1] is of great interest for a wide range of applications, including the fundamental science and technologies that support broadband communication networks and navigation with global positioning systems (GPSs). Research on optical frequency standards has progressed from spectral lamps to frequency-stabilized lasers using Doppler-free laser spectroscopy with improved frequency stability and reproducibility [2]. The invention of optical frequency combs [3, 4] has revolutionized the field of optical frequency measurement and further stimulated research on optical frequency standards. Today, optical frequency standards using cold atoms or ions provide much lower uncertainties than the primary cesium (Cs) frequency standards that provide the present definition of the ‘second’. These research activities helped to advance the entire field of frequency metrology, especially in the optical area, which includes optical frequency standards, measurement, comparisons, and transfer.

Figure 1 shows the technologies used in high-resolution laser spectroscopy. The main issue here is to observe atomic and molecular spectra without the inhomogeneous broadening caused by the Doppler shifts of atoms or molecules with different velocities. In saturation absorption spectroscopy, at high laser intensities the population difference between two levels is reduced as atoms are excited to the upper level. A weak laser beam is subsequently used to probe the saturation dip in the population difference, which has a homogeneous width about three orders of magnitude narrower than the Doppler width. In two-photon spectroscopy, when an atom absorbs two photons to drive the transition from each of the counter-propagating laser beams, then the Doppler shifts are cancelled out in the rest frame of the atom. With the atomic beam method, the laser beam intersects the atomic beam at right angles thus reducing the Doppler effect. When inhomogeneous broadening is reduced, attention must be paid to homogeneous broadening, which is affected by the interaction time between atoms and light, and that is usually limited by the collision between atoms and the transition lifetime. One way to reduce the collision between atoms and thus increase the interaction time with light is to cool the atoms with laser radiation. There are two systems capable of increasing the interaction time between atoms

and light. One such experiment was carried out using a single ion with a sufficiently long transition lifetime that was trapped by electric fields and then laser cooled [5]. Another system uses laser cooled atoms confined in optical lattices, again with a long transition lifetime [6].

Figure 1 also shows the applications of high-resolution spectroscopy. Frequency-stabilized lasers have long been used to realize the definition of the “meter” [7, 8]. Recent developments on optical frequency standards have encouraged time applications and accelerated work on the redefinition of the second [9, 10]. In terms of fundamental physics, optical clocks can be used to demonstrate relativistic effects in daily life and to verify the constancy of fundamental constants [11, 12]. In practice, high-resolution laser spectroscopy can provide new tools for improving telecommunication systems [13-14] and medical diagnostics systems [15]. High performance optical clocks are expected to open the door to new research frontiers such as relativistic geodesy [16].

Figure 2 shows the standards and measurement tools involved in optical frequency metrology. With the laser cooling technique, the uncertainty of Cs atomic clocks has been reduced to the 10^{-16} level [17, 18]. On the other hand, state-of-the-art experiments have achieved an uncertainty of 10^{-18} with optical clocks [19-22]. Optical frequency combs are used to measure and compare optical and microwave clocks [23]. A frequency comb based on a Kerr-lens Ti:sapphire laser can cover more than one octave of optical frequencies from 500 to 1100 nm [3, 4]. Furthermore, a frequency comb based on an Er-doped mode-locked fibre laser [24-26] can cover more than one octave of optical frequencies from 1000 to 2200 nm. A detailed description of fibre-based optical frequency combs can be found elsewhere [27]. The frequency combs are able to link optical and microwave frequencies with uncertainties of 10^{-17} - 10^{-18} [28, 29] and to link optical frequencies with uncertainties of 10^{-19} - 10^{-21} [30, 31]. A microwave frequency can be transferred using either a global positioning system (GPS) link or a carrier-phase two-way satellite time and frequency transfer (TWSTFT) with uncertainties of 10^{-15} - 10^{-17} [32-35], while an optical frequency can be transferred using an optical fibre link with uncertainties of 10^{-18} - 10^{-21} [36-39]. A transportable optical clock can also be used to compare optical frequency standards located in different places

with an uncertainty limited by that of the transportable clock [40].

In Section 2, we present an overview of optical frequency standards recommended by the International Committee for Weights and Measures (CIPM) updated in 2015. We then review the recent development of secondary representations of the second, which constitutes part of the CIPM recommendations, in Section 3. In section 4, we present other CIPM recommended optical frequency standards together with some standards that have not been recommended by the CIPM but that are widely used. In Section 5, we introduce optical frequency ratio measurement and also advanced time and frequency transfer technologies, together with some discussion of the issues related to the redefinition of the second. Finally, section 6 describes the prospects for applications of optical frequency standards and optical frequency combs.

2. Overview of optical frequency standards recommended by the International Committee for Weights and Measures (CIPM)

Optical frequency standards are established by observing atomic or molecular transitions using high-resolution laser spectroscopy and stabilizing the laser frequency using the observed spectral lines. Following the equation $\lambda = c/\nu$, where λ is wavelength, c is the speed of light and ν is optical frequency, an optical frequency standard can be used as a wavelength standard for length measurements when its frequency is measured based on the Cs frequency standard. This was the main application of optical frequency standards in the last century. With improved accuracy, optical frequency standards are now used as both a wavelength standard and a time standard (time period $T = 1/\nu$). For those optical frequency standards with lower uncertainties compared with the Cs frequency standard, they have a potential to provide a new definition of the second.

Table 1 shows a list of optical frequency standards drawn from the CIPM recommended standard frequencies for applications including the practical realization of the metre and secondary representations of the second [41]. This list only includes the optical frequency standards that are being actively studied

or are in practical use. To make the applications clear, in this article we divide the list into three parts:

- 1) Secondary representations of the second (SRSs);
- 2) Standards that are mainly aimed at establishing a time standard (time);
- 3) Standards that are used for length and other applications.

The list is in order of wavelength and contains information about the atomic or molecular transition, the spectroscopic method, and the frequency and the uncertainty of each standard. The frequency and uncertainty values were where necessary updated in 2015 and will be further updated in the future based on new research results.

In principle, all the standards in the list may be used for the practical realization of the metre. This is performed by using an optical frequency standard (as a wavelength standard) in the interferometric measurement for determining the length of a practical length standard, for example a gauge block. SRSs, listed in the first part of Table 1, are candidates for the future redefinition of the second and can already be used as time standards for the calibration of the International Atomic Time (TAI). Standards that are mainly intended to become time standards but that have not yet qualified as an SRS are given in the second part of the list. In the third part of the list, iodine- or Rb-stabilized lasers are shown as practical wavelength standards for length applications. The 1.54- μm acetylene-stabilized laser is used as a wavelength standard for telecommunication applications [13, 14, 42]. Since the light source of the 778-nm ^{85}Rb -stabilized laser can be either a direct 778-nm diode laser or a frequency-doubled laser using a 1.56- μm diode laser, this optical frequency standard can be used for either length applications at 778 nm or telecommunication applications at 1.56 μm . The 3.39- μm CH_4 -stabilized He-Ne laser played an important role historically and is still a significant standard in the mid infrared wavelength region.

All the standards for the SRSs are developed using either a trapped and laser-cooled single ion [5] or laser-cooled atoms in an optical lattice [6]. Other standards with the potential to become time standards also use the same methods except for the 243-nm ^1H and the 657-nm ^{40}Ca standards, where a cold atomic beam with two-photon absorption and cold atoms are used, respectively. Optical

frequency standards used for length and other applications are based on the saturation absorption method except for the 778-nm ^{85}Rb standard, where the two-photon absorption method is used to observe Doppler-free signals.

The recommended frequencies and uncertainties of the SRSs and other time standards (clocks) are calculated using the least-squares analysis of clock frequency comparison data including absolute frequencies (frequencies referenced to the SI second) and frequency ratios of different optical clocks [43]. The frequency uncertainties of the SRSs range from 10^{-15} to 10^{-16} , while the uncertainties of other time standards range from 10^{-13} to 10^{-16} . The Hg optical lattice clock with an uncertainty of 6×10^{-16} will be considered for adoption as an SRS in the near future. The uncertainties of standards for length and other applications range from 10^{-10} to 10^{-12} . Since the uncertainty of an interferometric measurement is at a level of about 10^{-9} and limited by the variation in the refractive index of the air, the uncertainties of the wavelength standards are usually good enough for length applications. In telecommunication applications, the required uncertainty of the standard is more relaxed.

3. Secondary representations of the second (SRSs)

The SRSs are recommended by the CIPM for research, comparison and discussion during the preparation of a future redefinition. They can be either optical or microwave frequency standards, provided that their uncertainty is well evaluated and is close to or smaller than the Cs uncertainty. In Table 1, seven optical frequency standards are listed as SRSs, while a total of eight SRSs are recommended by the CIPM. An SRS based on the ground-state hyperfine quantum transition of ^{87}Rb at 6.8 GHz is excluded because microwave frequency standards are not within the scope of this review. In this section, ^{87}Sr and ^{171}Yb optical lattice clocks are described with some experimental details, while others are introduced only with measurement results. This is because ^{87}Sr and ^{171}Yb optical lattice clocks are studied by a large number of groups worldwide and the experimental details could be of interest to a large number of readers.

1) ^{87}Sr optical lattice clock

An “optical lattice clock” was proposed with the aim of realizing high stability and accuracy simultaneously [44]. The scheme uses millions of neutral atoms trapped in an optical lattice, where the light field perturbation is cancelled out by properly designing the light shift potentials [45, 46]. The light shift cancellation is realized by setting the lattice laser at a particular wavelength (magic wavelength) so that the upper and lower states of the clock transition provide an equal light shift [6, 47]. The subwavelength localization of the atoms in each lattice site suppresses the first-order Doppler-shift [48] and collisional shift, while it provides a long interrogation time.

Figure 3 shows the relevant energy level diagram of ^{87}Sr . The $5s^2\ ^1S_0(F=9/2)$ - $5s5p\ ^3P_0(F=9/2)$ transition at 698 nm has a natural linewidth of 1 mHz and is used as a clock transition. The ultracold atoms for confinement in an optical lattice can be prepared by using first- and second-stage magneto-optical traps (MOTs). Firstly, by using the strong dipole-allowed transition ($^1S_0 - ^1P_1$; 461 nm, natural linewidth of 30 MHz), we can decelerate ^{87}Sr atoms in an atomic beam (to ~ 1 K) with a Zeeman slower. The atoms are then loaded into the first stage MOT using the same transition. A compact light source at 461 nm can be developed using a single-pass periodically poled LiNbO_3 waveguide for second-harmonic generation from a 922-nm extended cavity diode laser [49]. The atoms are then cooled further in the second stage MOT using a spin-forbidden transition ($^1S_0 - ^3P_1$; 689 nm, natural linewidth of 7.6 kHz). The Sr atoms were cooled to mK and μK levels, in the first and second stage MOTs, respectively. The ultranarrow $^1S_0 - ^3P_0$ clock transition of ^{87}Sr was first observed in 2003 by groups in Paris and Tokyo [50, 51]. The Paris group (SYRTE) [50] investigated the transition by employing saturated absorption spectroscopy in free space, while the Tokyo group (UT) [51] observed the transition with a linewidth of 700 Hz in an optical lattice tuned to the magic wavelength. In 2005, a Sr optical lattice clock was established and its absolute frequency was measured at UT in cooperation with NMIJ [6, 33]. In 2006, three groups (UT-NMIJ [34], JILA [52] and SYRTE [53]) measured the absolute frequency of their Sr optical lattice clocks independently. The weighted average of the frequencies measured by the three groups gave an average frequency with a standard deviation of 3.2 Hz (relatively

7.5×10^{-15}), which indicates good agreement between the measurement results of the three groups. Based on these results, in 2006 the CIPM decided to adopt the Sr lattice clock as an SRS [54].

Figure 4 shows absolute frequency measurement results and the CIPM recommendation value of the $^1S_0 - ^3P_0$ transition in ^{87}Sr . The absolute frequency results include the measurements made by SYRTE in 2008 [55], JILA in 2008 [56] and UT-NMIJ in 2009 [57] that contributed to the CIPM recommendations in 2009, 2013 and 2015; the measurements of PTB in 2011 [58] and NICT in 2012 [59] that contributed to the CIPM recommendations in 2013 and 2015; the updated results from SYRTE in 2013 [60], PTB in 2014 [61], NMIJ in 2014 [62], NIM in 2015 [63], NICT in 2015 [64], and NMIJ in 2015 [65] that contributed to the CIPM recommendations in 2015; and one new PTB result [66] that was published after the CIPM recommendation in 2015. The vertical solid line and the dashed lines, respectively, indicate the absolute frequency and the uncertainty (one sigma in each side) of the CIPM recommendation in 2015. The absolute frequency results agree with each other and also the recommended frequency. Since the frequency uncertainty of a Sr optical lattice clock is usually much smaller than that of microwave standards, the uncertainty of the absolute frequency results reflects that of the SI second realized by each institute.

Sr optical lattice clocks are being developed in many institutes and are the most investigated of all the optical clocks. The uncertainty of the CIPM recommended frequency (5×10^{-16}) is the lowest among all the optical clocks and is limited by the SI second. The frequency uncertainty of a Sr optical lattice clock can be as low as 10^{-18} , and is limited by the lattice light shift and the density shift [19, 20]. The low uncertainty of the Sr optical lattice clock was validated by using cryogenic optical lattice clocks in the same laboratory [19]. An international comparison of two Sr optical lattice clocks at PTB and SYRTE was undertaken using a 1415-km-long optical fibre network to confirm that the two clocks agreed at an uncertainty of 5×10^{-17} [39].

2) ^{171}Yb optical lattice clock

^{171}Yb has attracted considerable attention because it has a reasonable natural abundance of 14% and a simple $F=1/2$ spin system, which means we could avoid

the need for an extra optical pumping process in the experiment. Figure 5 shows the relevant energy level diagram of ^{171}Yb . The $6s^2\ ^1\text{S}_0$ ($F = 1/2$) - $6s6p\ ^3\text{P}_0$ ($F = 1/2$) transition has a natural linewidth of 44 mHz and is used as a clock transition. A clock laser at 578 nm can be generated using the sum-frequency generation of a 1319-nm laser and a 1030-nm laser [67, 68], or the second-harmonics generation (SHG) of a 1156-nm diode laser [69], with a PPLN waveguide device [70]. ^{171}Yb can be trapped and cooled by using first- and second-stage MOTs. The first-stage MOT uses a strong dipole-allowed transition ($^1\text{S}_0$ - $^3\text{P}_0$; 399 nm, natural linewidth of 29 MHz), while the second-stage MOT uses the spin-forbidden transition ($^1\text{S}_0$ - $^3\text{P}_1$; 556 nm, natural linewidth of 182 kHz). The Yb atoms were cooled to mK and 10 μK levels, in the first- and second-stage MOTs, respectively. The 399 and 556-nm light sources can be generated by using the SHG of a 798-nm diode laser [71] and a 1112-nm fibre laser [72], respectively, with a PPLN waveguide device.

An Yb optical lattice clock was established and its absolute frequency was measured at NMIJ [73]. The measurement results were reported to the CIPM in June 2009 for a discussion of new frequency standards. This has led to the first recommendation of a ^{171}Yb optical lattice clock by the CIPM. Figure 6 shows the frequency measurement results and the CIPM recommendation value for the $^1\text{S}_0$ - $^3\text{P}_0$ transition in ^{171}Yb . The results include the measurements obtained by NMIJ in 2009 [73], which contributed to the CIPM recommendations in 2009, 2013 and 2015; the measurements reported by NIST in 2009 [74] and by NMIJ in 2012 [75], which contributed to the CIPM recommendations in 2013 and 2015; the updated results from KRISS in 2013 [76], from NMIJ in 2014 [77, 78], and from RIKEN/UT in 2015 [79], which contributed to the CIPM recommendation in 2015; and one new INRIM result [80] that was published after the CIPM recommendation in 2015. Since the frequency uncertainty of an Yb optical lattice clock is usually smaller than that of the microwave standards, the uncertainty of the absolute frequency results (NIST09, NMIJ12, KRISS13 and INRIM16) reflects that of the SI second realized in each institute. The absolute frequency results provided by NMIJ14 and RIKEN/UT15 were calculated from frequency ratio measurements of Yb and Sr optical lattice clocks.

The Yb optical lattice clock was recommended by the CIPM as an SRS in 2013

[81]. Yb optical lattice clocks have been developed in five institutes and are being actively investigated in the optical clock community. The uncertainty of the CIPM recommended frequency (2×10^{-15}) is slightly large compared with other SRSs and comes from the discrepancy in the frequency ratio measurement of Yb and Sr optical lattice clocks [77-79]. We will come back to this point in section 5. The frequency uncertainty of an Yb optical lattice clock can be as low as 10^{-17} , and is presently limited by the lattice light shift [82]. The low uncertainty of the Yb optical lattice clock can be validated by comparing two Yb optical lattice clocks in the same laboratory or remotely by using optical fibre links. An alternative validation method is to measure and compare the frequency ratio of Yb and Sr optical lattice clocks in different laboratories.

3) $^{171}\text{Yb}^+$ single-ion clock (quadrupole transition)

$^{171}\text{Yb}^+$ single-ion clocks using the $6s\ ^2S_{1/2}$ ($F=0$) – $5d\ ^2D_{3/2}$ ($F=2$) quadrupole transition are being developed by PTB [83] and NPL [84]. The CIPM recommended the $^{171}\text{Yb}^+$ single-ion clock (quadrupole transition) as an SRS in 2006 [54]. Figure 7 shows the frequency measurement results and the CIPM recommendation value of the $^2S_{1/2} - ^2D_{3/2}$ transition in $^{171}\text{Yb}^+$. The results include measurements obtained by PTB in 2005 [85] and 2009 [86] and by NPL in 2010 [87] that contributed to the CIPM recommendations in 2013 and 2015; the updated results from NPL in 2014 [84] and PTB in 2014 [83] that contributed to the CIPM recommendation in 2015. The absolute frequency results agree with each other and also the recommended frequency. The uncertainty of the CIPM recommended frequency is 6×10^{-16} , which is the second smallest among SRSs. The frequency uncertainty of a $^{171}\text{Yb}^+$ single-ion clock (quadrupole transition) can be as low as 1×10^{-16} , and is presently limited by the blackbody radiation shift and the residual quadrupole shift [83, 84].

4) $^{171}\text{Yb}^+$ single-ion clock (octupole transition)

$^{171}\text{Yb}^+$ single-ion clocks using the $6s\ ^2S_{1/2}$ ($F=0$) – $4f^{13}6s^2\ ^2F_{7/2}$ ($F=3$) octupole transition are being developed by NPL [84] and PTB [88]. An important feature of the $^{171}\text{Yb}^+$ single-ion clock is that the quadrupole and octupole transitions have an exceptionally large differential sensitivity to time variations of the fine structure constant, which makes it possible to undertake important tests in fundamental

physics [84, 88]. The CIPM recommended the $^{171}\text{Yb}^+$ single-ion clock (octupole transition) as an SRS in 2013 [81]. Figure 8 shows the frequency measurement results and the CIPM recommendation value of the $^2\text{S}_{1/2} - ^2\text{F}_{7/2}$ transition in $^{171}\text{Yb}^+$. The results include the measurements obtained by NPL in 2009 [89] and 2012 [90] and by NPL in 2012 [91] that contributed to the CIPM recommendations in 2013 and 2015; the updated results from NPL in 2014 [84] and PTB in 2014 [88] that contributed to the CIPM recommendation in 2015. The absolute frequency results agree with each other and also the recommended frequency. The uncertainty of the CIPM recommended frequency is 6×10^{-16} , which is also the second smallest one in SRSs. The frequency uncertainty of a $^{171}\text{Yb}^+$ single-ion clock (octupole transition) can be as low as 3.2×10^{-18} , and is presently limited by the quadratic Stark shift induced by thermal radiation at room temperature [21].

5) $^{88}\text{Sr}^+$ single-ion clock

$^{88}\text{Sr}^+$ single-ion clocks using the $5s \ ^2\text{S}_{1/2} - 4d \ ^2\text{D}_{5/2}$ transition are being developed by NPL [92] and NRC [93, 94]. The CIPM recommended the $^{88}\text{Sr}^+$ single-ion clock as an SRS in 2006 [54]. Figure 9 shows the frequency measurement results and the CIPM recommendation value of the $^2\text{S}_{1/2} - ^2\text{D}_{5/2}$ transition in $^{88}\text{Sr}^+$. The results include the measurements reported by NPL in 2004 [95] and by NRC in 2005 [96] and 2013 [92, 94] that contributed to the CIPM recommendations in 2013 and 2015; the updated results from NPL in 2014 [92] that contributed to the CIPM recommendation in 2015. The absolute frequency results agree with each other and also the recommended frequency. The uncertainty of the CIPM recommended frequency is 1.6×10^{-15} , which was enlarged by a factor of 3 [41], by the CIPM, taking into account that the uncertainty obtained by least-squares analysis [43] is dominated by one result [92] with a much lower uncertainty than any of the others. The frequency uncertainty of an $^{88}\text{Sr}^+$ single-ion clock can be as low as 5×10^{-17} , and is currently limited by the blackbody radiation shift [92].

6) $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ single-ion clocks

A $^{199}\text{Hg}^+$ single-ion clock using the $5d^{10}6s \ ^2\text{S}_{1/2} (F=0) - 5d^96s^2 \ ^2\text{D}_{5/2} (F=2)$ transition and a $^{27}\text{Al}^+$ single-ion clock using the $3s^2 \ ^1\text{S}_0 - 3s3p \ ^3\text{P}_0$ transition are being developed by NIST [11, 22, 97]. The $^{27}\text{Al}^+$ single-ion clock is realized using

the quantum logic spectroscopy (QLS) method [98], where an auxiliary ion (for example, $^9\text{Be}^+$) takes over the laser cooling and state detection requirements for $^{27}\text{Al}^+$. The CIPM recommended $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ single-ion clocks as SRSs in 2006 [54] and 2013 [81], respectively. The current CIPM recommendations for $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ single-ion clocks were updated in 2013 using the measurement results obtained for $^{199}\text{Hg}^+$ in 2007 [99] and for $^{27}\text{Al}^+$ in 2007 [100] and 2008 [97], respectively. The $^{27}\text{Al}^+$ measurement in 2008 was a frequency ratio measurement between $^{27}\text{Al}^+$ and $^{199}\text{Hg}^+$ clocks with a much smaller uncertainty than that of the Cs clock [97]. The uncertainty of the CIPM recommended absolute frequencies for the $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ single-ion clocks are 1.9×10^{-15} , which was enlarged by a factor of 3 [41] taking into account that the uncertainty obtained by least-squares analysis [43] is dominated by one result (absolute frequency of $^{199}\text{Hg}^+$ measured in 2007) [99] without validation from other laboratories. The frequency uncertainty of $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ single-ion clocks can be as low as 1.9×10^{-17} [97] and 8.6×10^{-18} [22], respectively, and is presently limited by a residual quadrupole shift [97] and excess micromotion [22], respectively.

4. Non-SRS optical frequency standards

4.1 Standards mainly designed for time standard applications

In Table 1, there are six optical frequency standards that are non-SRS that are mainly designed for time standard applications. Here we focus on three standards (^{199}Hg , $^{40}\text{Ca}^+$, ^1H) that are being actively studied and that were updated in 2015 and we describe their measurement results. $^{115}\text{In}^+$, ^{40}Ca and ^{88}Sr were updated in 2003, 2005 and 2009, respectively. Although the last update of the $^{115}\text{In}^+$ single-ion clock was in 2003, some new research activities have been reported toward the quantum logic spectroscopy [101] and a multiple-ion clock [102] of In^+ .

1) ^{199}Hg optical lattice clock

^{199}Hg optical lattice clocks using the $5s^2 \ ^1\text{S}_0 - 5s5p \ ^3\text{P}_0$ transition have been developed by RIKEN/UT [103] and SYRTE [104]. Figure 10 shows frequency measurement results for a ^{199}Hg optical lattice clock. The results include the

measurements reported by SYRTE in 2012 [105, 106] and by RIKEN/UT in 2015 [103], that contributed to the CIPM recommendation in 2015; and one new result from SYRTE [104] that was published after the CIPM recommendation in 2015. The absolute frequency results agree with each other and also the recommended frequency. The uncertainty of the CIPM recommended frequency is 6×10^{-16} , which is much lower than that of some of the SRSs. The ^{199}Hg optical lattice clock should be considered for adoption as an SRS in the near future. The frequency uncertainty of the ^{199}Hg optical lattice clock can be as low as 7.2×10^{-17} , and is presently limited by the lattice light shift [103].

2) $^{40}\text{Ca}^+$ single-ion clock

$^{40}\text{Ca}^+$ single-ion clocks using the $4s \ ^2S_{1/2} - 3d \ ^2D_{5/2}$ transition have been developed by Innsbruck [107], NICT [108] and WIPM [109]. Figure 11 shows frequency measurement results for a $^{40}\text{Ca}^+$ single-ion clock. The results include the measurements reported by Innsbruck in 2009 [107], WIPM in 2012 [110] and NICT in 2012 that contributed to the CIPM recommendations in 2013 and 2015 (except for the 2012 WIPM); the updated results reported by WIPM in 2016 [109] that contributed to the CIPM recommendation in 2015. The 2012 WIPM result has been withdrawn by the authors due to an underestimation of the uncertainty contributed by ion micromotion [41]. As shown in Fig. 11, despite the withdrawal of WIPM12, there is still a discrepancy between the measurement results. The uncertainty of the CIPM recommended frequency is 1.2×10^{-14} , which was increased by a factor of 10 [41], by the CIPM, taking into account that the uncertainty obtained by least-squares analysis [43] is calculated using inconsistent data from different institutes. The frequency uncertainty of a $^{40}\text{Ca}^+$ single-ion clock can be as low as 5×10^{-17} , and is presently limited by the shifts of excess micromotion and blackbody radiation [109]. We expect further research to clarify the cause of the discrepancy of the measurement results from different institutes.

3) ^1H

The ^1H optical frequency standard using the $1S - 2S$ 2-photon transition was developed by MPQ [111]. A cold atomic beam of ^1H with two-photon absorption spectroscopy was employed in the experiment. A recent frequency measurement

result [111] has an uncertainty of 4.5×10^{-15} , and is mainly limited by the second order Doppler effect in the spectroscopy. The uncertainty of the CIPM recommended frequency is 9×10^{-15} , which is based on two previous measurements reported by MPQ [111, 112] that include a slight drift in the frequency values.

4.2 Standards used for length and other applications

In Table 1, there are eight optical frequency standards that can be used for length and other applications. Here we concentrate on two standards that have been newly added to the 2015 CIPM recommendations, namely the 531-nm diode laser stabilized to $^{127}\text{I}_2$ and the 780-nm diode laser stabilized to ^{87}Rb , and describe their measurement results. Other standards were updated until 2007, because they are quite mature and there is no new measurement reported. Among them, the 633-nm iodine-stabilized He-Ne laser is characterized as the most popular standard for length applications, and the 3.99- μm CH_4 -stabilized laser is being studied using new light sources [113, 114].

1) 531-nm iodine-stabilized diode laser

The frequency of a 531-nm iodine-stabilized diode laser was newly recommended by the CIPM in 2015. Iodine absorption lines near 532 nm [115, 116] are much stronger than those near 633 nm. With high-resolution laser spectroscopy [117], iodine-stabilized Nd:YAG lasers were developed with excellent laser frequency stability [2] and reproducibility [118]. Accurate hyperfine constants of molecular iodine near 532 nm were obtained based on detailed measurements of the hyperfine structures of molecular iodine [115, 119-125]. For practical length applications, the 532-nm iodine-stabilized Nd:YAG laser was expected to replace the 543-nm iodine-stabilized He-Ne laser since the former produces a much higher output power at the green wavelength region than the latter. As a new wavelength standard, the frequency reproducibility of the laser was validated through international comparisons [126-129]. The 532-nm iodine-stabilized Nd:YAG laser turned out to be a perfect laser source for length applications except that it is very expensive for practical applications. The recommendation of the 531-nm iodine-stabilized diode laser will make up for the shortcomings of the Nd:YAG laser.

The 531-nm iodine-stabilized diode laser uses a low-cost coin-sized light source consisting of a 1062-nm distributed-feedback diode laser and a frequency-doubling element that provide 20 mW of green output power. To perform Doppler-free spectroscopy of molecular iodine using the 531-nm laser module, a simple and compact iodine spectrometer that employs saturated absorption was developed using a short iodine cell operated at room temperature and without using any expensive optical components such as an electro-optic modulator [130]. A frequency stability at the 10^{-12} level and a frequency uncertainty at the 10^{-11} level have been achieved with the compact iodine-stabilized diode laser. The uncertainty of the CIPM recommended frequency is 1×10^{-10} , which was larger by a factor of 7 than the experimentally measured uncertainty [130]. This was due to the fact that the iodine vapour pressure used was much higher than with other iodine-stabilized lasers, and also that the result comes from a single laboratory without validation from other laboratories [41]. Recently, a 531-nm iodine-stabilized diode laser has been successfully applied to long gauge block measurements [131].

2) 780-nm Rb-stabilized laser

The frequency of a 780-nm Rb-stabilized laser was newly recommended by the CIPM in 2015. This is to meet a request from the length community, although the stabilized laser was not newly developed. This laser is attractive for length applications because it uses a commercially available external-cavity diode laser and a simple and compact spectroscopic configuration, and has a high optical power output [132]. The laser frequency is stabilized to the Doppler-free Rb D₂ line $5S_{1/2} - 5P_{3/2}$ ($F''=2 - F'=2/F''=2 - F'=3$ crossover line) by using a third-harmonic technique. The relative frequency uncertainty was 4.3×10^{-10} for a 0.01-s averaging time. The uncertainty of the CIPM recommended frequency is 5×10^{-10} , which is based on two previous measurements reported by JILA [133] and NMIJ [129].

4.3 Other CIPM-recommended standards

CIPM-recommended optical frequency standards also include an unstabilized He-Ne laser, an OsO₄-stabilized CO₂ laser, spectral lamps and iodine-stabilized

lasers operating at other wavelengths that are not listed in Table 1. The lamps and the iodine-stabilized lasers were summarized in 2001 in category 2 of the CIPM recommendation [7]. They are ^{86}Kr , ^{198}Hg and ^{114}Cd spectral lamps and iodine stabilized lasers operating at 515, 576, 612 and 640 nm. Although they are not currently being actively studied, some of them are still in use. Of these only the frequency of the 515-nm iodine stabilized laser was updated by the CIPM recommendation in 2003 [8].

4.4 Other optical frequency standards

There are some optical frequency standards that have not been recommended by the CIPM but that are widely used for various applications. For example, HCN-stabilized lasers emitting at $1.5\ \mu\text{m}$ [134] are used as a wavelength standard for telecom applications as well as the acetylene-stabilized laser. Moreover, a single-mode optical-fibre-coupled absorption cell containing acetylene gas is used as a simple wavelength standard based on Doppler-limited frequency references of acetylene [135]. Iodine-stabilized lasers are investigated across visible wavelengths including 502 nm [136], 578 nm [67, 137] and 660 nm [138]. In addition to Rb-stabilized lasers, Cs-stabilized lasers are also used as optical frequency standards in the near infrared region [139]. Furthermore, the absorption spectra of $^{130}\text{Te}_2$ provide suitable wavelength references at blue and green regions [140, 141]. For astronomical spectroscopy calibration, Th-Ar hollow cathode lamps [142, 143] are widely used as a wavelength standard in observatories.

5. Optical frequency ratio measurement and advanced frequency links

Optical clocks are now approaching uncertainties of 10^{-18} , which are orders of magnitude lower than the SI second. Consequently, optical frequency ratios between optical clocks give much more detailed frequency descriptions than absolute frequency measurements. The validation of clock uncertainties based on frequency ratio measurement can confirm clock consistency and are an essential step towards the redefinition of the second [9, 10]. Furthermore,

accurate frequency ratio measurement plays an important role in fundamental physics for verifying the constancy of fundamental constants [11, 12]. On the other hand, optical frequency links using optical fibre networks and transportable optical clocks can assist with progress on frequency ratio measurements by establishing a clock network between laboratories on a continental and even inter-continental scale. The validation of clock consistency between different laboratories is especially important in terms of the redefinition of the second. Furthermore, accurate comparisons of optical clocks in different places provide direct information about geopotential measurements [15]. Finally, we conclude this section by discussing certain issues related to the redefinition of the second.

5.1 Optical frequency ratio measurements

Figure 12 shows the relationship between the optical frequency standards and the SI second. All the optical frequency standards have been measured with reference to the SI second, except for the $^{27}\text{Al}^+$ single-ion clock. Optical frequency ratio measurements have been performed between an ^{87}Sr optical lattice clock and several other clocks, $^{27}\text{Al}^+$ and $^{199}\text{Hg}^+$ single-ion clocks, and $^{171}\text{Yb}^+$ (quadrupole) and $^{171}\text{Yb}^+$ (octupole) single-ion clocks.

1) Frequency ratio of $^{27}\text{Al}^+$ and $^{199}\text{Hg}^+$ single-ion clocks

The frequency ratio of the $^{27}\text{Al}^+$ and $^{199}\text{Hg}^+$ single-ion clocks was measured at NIST in 2008 [97]. The uncertainty of the measured ratio was 5.2×10^{-17} , which comprises a statistical measurement uncertainty of 4.3×10^{-17} , and systematic uncertainties of 1.9×10^{-17} and 2.3×10^{-17} in $^{199}\text{Hg}^+$ and $^{27}\text{Al}^+$ clocks, respectively. This was the first frequency ratio measurement between optical clocks to go beyond the Cs limit. The frequency ratio was used to determine the absolute frequency of a $^{27}\text{Al}^+$ clock using the absolute frequency of a $^{199}\text{Hg}^+$ clock [99]. Repeated measurements over more than one year yielded a preliminary constraint on the temporal variation of the fine-structure constant $\alpha'/\alpha = -1.6(2.3) \times 10^{-17}/\text{year}$ [99].

2) Frequency ratio of $^{171}\text{Yb}^+$ (quadrupole) and $^{171}\text{Yb}^+$ (octupole) single-ion clocks

The frequency ratio of $^{171}\text{Yb}^+$ (quadrupole) and $^{171}\text{Yb}^+$ (octupole) single-ion clocks was measured at NPL in 2014 [84]. The uncertainty of the measured ratio

was 3.3×10^{-16} , which was mostly limited by the uncertainty of the $^{171}\text{Yb}^+$ (quadrupole) clock. In this experiment, both the frequency ratio and the absolute frequency of each $^{171}\text{Yb}^+$ clock were measured simultaneously based on the Cs fountain clock at NPL. These results provided an important self-consistency check. The measurements have led to a threefold improvement in the constraint on the time variation of the proton-to-electron mass ratio $\mu'/\mu = 0.2(1.1) \times 10^{-16}/\text{year}$, along with an improved constraint on the temporal variation of the fine-structure constant $\alpha'/\alpha = -0.7(2.1) \times 10^{-17}/\text{year}$.

3) Frequency ratio of ^{171}Yb and ^{87}Sr optical lattice clocks

The frequency ratio of ^{171}Yb and ^{87}Sr optical lattice clocks is being measured at NMIJ [77, 78] and RIKEN/UT [79, 82]. Figure 13 shows the frequency ratio measurements obtained by NMIJ in 2014 [77, 78] and RIKEN/UT in 2014 [79] and 2016 [82]. The measurements RIKEN/UT14 and RIKEN/UT16 agree with each other within their measurement uncertainties, but disagree with the measurement result NMIJ14. The difference between NMIJ14 and RIKEN/UT16 is 2.9×10^{-15} , while their individual uncertainties are 1.4×10^{-15} and 4.6×10^{-17} , respectively. The recent absolute frequency measurement of the ^{171}Yb optical lattice clock with an uncertainty of 5.4×10^{-16} together with the CIPM recommendation of the ^{87}Sr optical lattice clock provide additional information on the $^{171}\text{Yb}/^{87}\text{Sr}$ ratio as shown in Fig. 13. We expect further research to clarify the cause of this discrepancy.

4) Frequency ratio of ^{199}Hg and ^{87}Sr optical lattice clocks

The frequency ratio of ^{199}Hg and ^{87}Sr optical lattice clocks is being measured at RIKEN/UT [103] and SYRTE [104]. Excellent agreement has been obtained between the two measurements. The difference between the two measurements is 1.7×10^{-16} , while their individual uncertainties are 8.4×10^{-17} (RIKEN/UT) and 1.8×10^{-16} (SYRTE). This is the first result comparing frequency ratios that has an agreement better than the Cs clock accuracy. With this kind of frequency ratio comparison, it is possible to validate the consistencies of optical clocks in different laboratories beyond the Cs limit, especially when we can close the loop of the ratio measurement (for example, by measuring the ratios of $^{171}\text{Yb}/^{87}\text{Sr}$, $^{87}\text{Sr}/^{199}\text{Hg}$ and $^{199}\text{Hg}/^{171}\text{Yb}$, and multiply the ratios to obtain the result of 1 for checking the

consistency of the measurements).

5) Frequency ratio of $^{40}\text{Ca}^+$ single ion and ^{87}Sr optical lattice clocks

The frequency ratio of the $^{40}\text{Ca}^+$ single ion and ^{87}Sr optical lattice clocks was measured at NICT in 2012 [108]. The uncertainty of the measured ratio was 2.3×10^{-15} , which was mostly limited by the uncertainty of the $^{40}\text{Ca}^+$ clock. This experiment was performed mainly to check the consistency with the absolute frequency measurement of the $^{40}\text{Ca}^+$ clock, since there were two published values [107, 110] that are inconsistent with the NICT's measurement.

6) Frequency difference between ^{88}Sr and ^{87}Sr optical lattice clocks

The frequency difference between the ^{88}Sr and ^{87}Sr optical lattice clocks was measured at the University of Tokyo in 2008 [144]. The frequency difference was measured instead of the frequency ratio because the frequency difference between the two clocks is only about 62 MHz, and it is much easier to deal with the frequency difference in this case. The uncertainty of the measured frequency difference was 3×10^{-15} , which was mostly limited by the uncertainty of the ^{88}Sr clock.

5.2 Advanced frequency links

The precision of the conventional frequency transfer using microwave signals needs to be improved so as to meet the high stability requirement of optical clocks. The ubiquitous fibre optic network enables us to transfer frequencies with an extremely low phase noise [145, 146]. There are two methods for transferring frequencies using optical fibres: 1) microwave frequencies transmitted over a modulated optical signal; 2) an optical carrier signal with the optical phase information to achieve stability that is several orders of magnitude higher than that of microwave modulation signals. In both cases, active phase noise cancellation is necessary because of the additional phase noise resulting from the optical length fluctuations of the delivering fibres caused by mechanical or temperature variations.

Results have been reported for tests on coherent optical frequency transfer with residual frequency instabilities of 1×10^{-17} through a 32-km fibre [36] and of 2×10^{-16} through a 251-km fibre [37] at 1 s. The precision frequency measurement

of a Sr optical lattice clock was demonstrated on the basis of an optical carrier transfer over a 120-km long-haul fibre network composed of installed rural and urban telecom fibres [38, 57]. Stable optical fibre links were used to compare two optical clocks without degrading their stability and accuracy [147, 148]. A 920-km-long optical fibre link for frequency metrology has been established between Braunschweig and München in Germany with uncertainties of 5×10^{-17} 1s and 4×10^{-19} for long integration times [149]. A 642-km-long optical fibre link was established in Italy to link atomic clocks in INRIM, Turin and LENS [150]. Recently, a 1415-km telecom fibre link has been established for comparing optical clocks in Paris and Braunschweig [39]. The link reached an uncertainty of 3×10^{-17} after an averaging time of 1,000 s, which is ten times better and more than four orders of magnitude faster than previous long-distance clock comparisons. It has been confirmed that the two ^{87}Sr optical lattice clocks at SYRTE and PTB agree with each other at an uncertainty of 5×10^{-17} , limited by the instability and uncertainty of the strontium lattice clocks themselves. This is the first international long-distance optical clock comparison to validate clock uncertainty at a level well below the Cs limit. On the other hand, a recent short-distance clock comparison using a telecom fibre has achieved geopotential measurements with an uncertainty of 5 cm (5×10^{-18}) [16].

Earth tide is a displacement of the earth's solid surface caused by the gravity of the Moon and other effects including the Sun. The main components of a tide are a period of about 12 hours and an amplitude of about 40 cm. This means that all the clocks on the earth experience this effect and their frequencies are modulated at a level of about 4×10^{-17} . When we compare clocks at the same location, the tide effect is eliminated. However, when we compare clocks separated by a long distance, we will need to confirm and remove the variation in the signal caused by the tide effect to observe other smaller signals of the gravitational potential. Consequently, optical clocks linked with fibres may serve as a direct sensor for gravitational potential with dynamic responses at a cm uncertainty level.

Other advanced time and frequency transfer technologies include a transportable optical clock [40], the Atomic Clock Ensemble in Space (ACES)

project [151] and carrier-phase two-way satellite frequency transfer [35]. In 2015, the Consultative Committee for Time and Frequency (CCTF), one of the consultative committees under the CIPM, recommended that national metrology institutes and other relevant international bodies vigorously support the research and development of time and frequency transfer techniques matching the stability and uncertainty of the most advanced optical clocks [152].

5.3 Issues related to the redefinition of the second

Optical clocks have demonstrated orders of magnitude smaller uncertainty and better reproducibility than the current best Cs fountain clocks. One of these clocks may finally lead to a new definition of the second. Discussions on the roadmap for the redefinition of the second are ongoing at CCTF and CIPM [9, 10]. We need to consider the next issues.

1) Evaluating the uncertainty of optical clocks is important. When providing a new definition of the second, an optical clock must have a smaller uncertainty than a Cs clock. At present, the ^{87}Sr optical lattice clock [19, 20], the $^{171}\text{Yb}^+$ single-ion clock (octupole transition) [21], and the $^{27}\text{Al}^+$ single-ion clock [22] have been evaluated as having an uncertainty at the 10^{-18} level, which is about two orders of magnitude better than that of the current best Cs fountain clocks.

2) It is better if multiple national institutes work on the same type of clock. This is important as regards confirming the equality of the clocks and also their reliability. Currently, a number of national institutes are investing their research resources into ^{87}Sr optical lattice clocks. Fibre links have been established to confirm the agreement between two ^{87}Sr optical lattice clocks at PTB and SRYTE at a level of 10^{-17} [39] and two ^{87}Sr optical lattice clocks at RIKEN and UT at a level of 10^{-18} [19]. A transportable ^{87}Sr optical lattice clock with an uncertainty of $< 10^{-17}$ is under development at PTB [40].

3) The frequency ratio measurement of optical clocks with an uncertainty smaller than the SI second would also be useful for validating the reproducibility of the optical clocks. Furthermore, after the redefinition of the second, information on the frequency ratio could be used to realize a new SI second by using other optical clocks. The frequency ratios of $^{27}\text{Al}^+ / ^{199}\text{Hg}^+$ [97], $^{171}\text{Yb} / ^{87}\text{Sr}$ [82], and $^{199}\text{Hg} / ^{87}\text{Sr}$

[103, 104] have already been measured at the 10^{-17} level, with the ratios of $^{199}\text{Hg}/^{87}\text{Sr}$ also being confirmed at the 10^{-17} level by two independent institutes [103, 104]. The $^{171}\text{Yb}/^{87}\text{Sr}$ frequency ratio at the lowest uncertainty level [82] should be able to be confirmed by NMIJ or at Boulder by NIST and JILA. The $^{171}\text{Yb}^+(\text{quadrupole})/^{171}\text{Yb}^+(\text{octupole})$ frequency ratio is also promising with respect to reaching the 10^{-17} uncertainty level.

4) We need to consider the contribution of the secondary representations to TAI. This will take the form of a report to BIPM detailing the experimental results obtained in a particular reporting period including the uncertainty budget and references. As an SRS, the Rb fountain clock has already started to contribute to TAI. On the other hand, reliable frequency measurement results of optical clocks based on Cs fountain clocks are still necessary if we are to obtain the best link between the new and the present definitions.

6. Outlook

Both the uncertainty and the reliability of an optical clock are very important in terms of redefining the second. It has been demonstrated recently at PTB that an ^{87}Sr optical lattice clock, with a time coverage of 46 % over 25 days, is now able to maintain a local timescale with a time error of less than 200 ps, which is superior to one based on even the best Cs fountain clock [153]. It has also been recently demonstrated at SYRTE that an ^{87}Sr optical lattice clock can be reliably operated over time periods of several weeks, with a time coverage exceeding 80 % [154]. Research along this line is increasing rapidly.

Efforts are still being made to further reduce the systematic uncertainty of optical clocks to $< 1 \times 10^{-18}$, either by improving the current optical clocks [155] or by starting new clock schemes using a nuclear clock transition in ^{229}Th [156, 157] or highly charged ions [158, 159]. But it may take a long time since some of these experiments are very complicated. Furthermore, the geoid of the earth is not well defined at the 10^{-18} level. Fibre-linked and transportable optical clocks may improve our knowledge regarding geoids, but the beat of the earth will finally limit the reproducibility of optical clocks on earth. An optical clock in space may

experience more uniform and stable gravitational potential compared to one on the earth, and thus have a higher clock stability and reproducibility. A space-based gravitational wave detector consisting of optical lattice atomic clocks has recently been proposed [160]. Also, there has been a proposal to search for dark matter using optical clocks [161]. Again, this will take a long time to realize, and we need to consider how significantly we can improve measurement on earth by using clocks in space. Therefore, it may not be long before we decide to change the definition of the second and proceed to applications.

It is very important to find killer applications for optical clocks. At present, an optical clock as a direct sensor of gravitational potential with dynamic responses appears quite promising for applications to geology, dynamic ocean topography, and seismology. We may expect more applications for optical clocks in the future, such as GPS navigation systems using microwave clocks.

The development of optical clocks is strongly linked with that of optical frequency combs. For example, the realization of a narrow linewidth fibre comb [162-164] has contributed to the frequency ratio measurement of optical lattice clocks [103]. Here, it is worth noting two research fields related to optical frequency combs that are being rapidly developed. Instead of using one comb component as in frequency measurement and link experiments, both approaches employ a large number of comb components simultaneously in an experiment. Astro-combs are being introduced into observatories to calibrate astronomical spectrographs as part of the search for extrasolar planets and for the direct measurement of the universe's expansion history [165, 166]. Dual comb spectroscopy [167, 168] is performed to observe the spectra of atoms and molecules across a 140-THz-wide spectrum from 1.0 to 1.9 μm [169]. This method is helpful for both basic spectroscopic research, such as understanding the molecular line profile [170-172], and applications such as the multicomponent analysis and investigation of the relaxation associated with several vibrational states [173].

It is now an exciting period for researchers working on optical frequency metrology. Rapid developments on optical clocks, frequency combs, and advanced frequency links will open up new possibilities for research ranging from

fundamental physics, and astrophysics, to aspects of engineering such as space clocks and the remote sensing of the beat of our earth.

Acknowledgments

We are grateful to K. Hosaka and D. Akamatsu for helpful discussions. Part of this work was supported by the Japan Society for the Promotion of Science (JSPS) KAKENHI (15H02028).

References

- [1] Udem T, Holzwarth R and Hänsch T W 2002 Optical frequency metrology *Nature* 416 233-7
- [2] Hall J L, Ma L S, Taubman M, Tiemann B, Hong F L, Pfister O and Ye J 1999 Stabilization and frequency measurement of the I₂-stabilized Nd : YAG laser *IEEE Trans. Instrum. Meas.* 48 583-6
- [3] Udem Th, Reichert J, Holzwarth R, and Hänsch T W 1999 Absolute optical frequency measurement of the cesium D₁ line with a mode-locked laser *Phys. Rev. Lett.* 82 3568-71
- [4] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis *Science* 288 635-9
- [5] Dehmelt H G 1982 Mono-ion oscillator as potential ultimate laser frequency standard *IEEE Trans. Instrum. Meas.* 31 83-7
- [6] Takamoto M, Hong F L, Higashi R and Katori H 2005 An optical lattice clock *Nature* 435 321-4
- [7] Quinn T J 2003 Practical realization of the definition of the metre, including recommended radiations of other optical frequency standards (2001) *Metrologia* 40 103-33
- [8] Felder R 2005 Practical realization of the definition of the metre, including recommended radiations of other optical frequency standards (2003) *Metrologia* 42 323-5
- [9] Riehle F 2015 Towards a re-definition of the second based on optical atomic clocks *C. R. Physique* 16 506-15
- [10] Gill P 2015 When should we change the definition of the second? *Phil. Trans. R. Soc. A* 369 4109-37
- [11] Chou C W, Hume D B, Rosenband T and Wineland D J 2010 Optical Clocks and Relativity *Science* 329 1630-3
- [12] Peik E, Lipphardt B, Schnatz H, Schneider T, Tamm C and Karshenboim S G 2004 Limit on the present temporal variation of the fine structure constant *Phys. Rev. Lett.* 93 170801
- [13] Nakagawa K, de Labachellerie M, Awaji Y and Kouroggi M 1996 Accurate optical frequency atlas of the 1.5- μ m bands of acetylene *J. Opt. Soc. Am. B* 13 2708-14
- [14] Hong F L, Onae A, Jiang J, Guo R, Inaba H, Minoshima K, Schibli T R, Matsumoto H and Nakagawa K, 2003 Absolute frequency measurement of an acetylene-stabilized laser at 1542 nm *Opt. Lett.* 28 2324-6
- [15] Thorpe M J, Balslev-Clausen D, Kirchner M S and Ye J 2008 Cavity-enhanced optical frequency comb spectroscopy: application to human breath analysis *Opt. Express* 16, 2387-97
- [16] Takano T, Takamoto M, Ushijima I, Ohmae N, Akatsuka T, Yamaguchi A, Kuroishi Y,

- Munekane H, Miyahara B and Katori H 2016 Geopotential measurements with synchronously linked optical lattice clocks *Nat. Photonics* 10 662-6
- [17] Guéna J, Abgrall M, Rovera D, Laurent P, Chupin B, Lours M, Santarelli G, Rosenbusch P, Tobar M, Li R, Gibble K, Clairon A and Bize S 2012 Progress in atomic fountains at LNE-SYRTE *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 59 391-410
- [18] Szymaniec K, Lea S N and Liu K 2014 An evaluation of the frequency shift caused by collisions with background gas in the primary frequency standard NPL-CsF2 *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 61 203-6
- [19] Ushijima I, Takamoto M, Das M, Ohkubo T and Katori H 2015 Cryogenic optical lattice clocks *Nat. Photonics* 9 185-9
- [20] Nicholson T L, Campbell S L, Hutson R B, Marti G E, Bloom B J, McNally R L, Zhang W, Barrett M D, Safronova M S, Strouse G F, Tew W L and Ye J 2015 Systematic evaluation of an atomic clock at 2×10^{-18} total uncertainty *Nature Communications* 6 6896
- [21] Huntemann N, Sanner C, Lipphardt B, Tamm Chr and Peik E 2016 Single-Ion Atomic Clock with 3×10^{-18} Systematic Uncertainty *Phys. Rev. Lett.* 116 063001
- [22] Chou C W, Hume D B, Koelemeij J C J, Wineland D J and Rosenband T 2010 Frequency comparison of two high-accuracy Al^+ optical Clocks *Phys. Rev. Lett.* 104 070802
- [23] Hong F L and Katori H 2010 Frequency metrology with optical lattice clocks *Jpn. J. Appl. Phys.* 49 080001
- [24] Hong F L, Minoshima K, Onae A, Inaba H, Takada H, Hirai A, Matsumoto H, Sugiura T and Yoshida M 2003 Broad-spectrum frequency comb generation and carrier-envelope offset frequency measurement by second-harmonic generation of a mode-locked fiber laser *Opt. Lett.* 28 1516-8
- [25] Washburn B R, Diddams S A, Newbury N R, Nicholson J W, Yan M F and Jorgensen C G 2004 Phase-locked, erbium-fiber-laser-based frequency comb in the near infrared *Opt. Lett.* 29 250-2
- [26] Schibli T R, Minoshima K, Hong F L, Inaba H, Onae A, Matsumoto H, Hartl I and Fermann M E 2004 Frequency metrology with a turnkey all-fiber system *Opt. Lett.* 29 2467-9
- [27] Xia W and Chen X 2016 Recent developments in fiber-based optical frequency comb and its applications *Meas. Sci. Technol.* 27 041001
- [28] Bartels A, Diddams S A, Oates C W, Wilpers G, Bergquist J C, Oskay W H and Hollberg L 2005 Femtosecond-laser-based synthesis of ultrastable microwave signals from optical frequency references *Opt. Lett.* 30 667-9
- [29] Millo J, Boudot R, Lours M, Bourgeois P Y, Luiten A N, Le Coq Y, Kersale Y and Santarelli G 2009 Ultra-low-noise microwave extraction from fiber-based optical frequency comb *Opt. Lett.* 34 3707-9

- [30] Ma L S, Bi Z, Bartels A, Robertsson L, Zucco M, Windeler R, Wilpers G, Oates C, Hollberg L and Diddams S A 2004 Optical frequency synthesis and comparison with uncertainty at the 10^{-19} Level *Science* 303 1843-5
- [31] Inaba H, Nakajima Y, Iwakuni K, Hosaka K, Onae A, Yasuda M, Akamatsu D and Hong F L 2012 Toward an optical frequency comb with relative frequency uncertainty at 10^{-21} -level *Conf. on Lasers and Electro-Optics. Quantum Electronics and Laser Science Conf.*
- [32] Larson K M and Levine J 1999 Carrier-phase time transfer *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 46 1001-12
- [33] Hong F L, Takamoto M, Higashi R, Fukuyama Y, Jiang J and Katori H 2005 Frequency measurement of a Sr lattice clock using an SI-second-referenced optical frequency comb linked by a global positioning system (GPS) *Opt. Express* 13 5253-62
- [34] Takamoto M, Hong F L, Higashi R, Fujii Y, Imae M and Katori H 2006 Improved Frequency measurement of a one-dimensional optical lattice clock with a spin-polarized fermionic ^{87}Sr isotope *J. Phys. Soc. Jpn.* 75 104302
- [35] Fujieda M, Piester D, Gotoh T, Becker J, Aida M, Bauch A 2014 Carrier-phase two-way satellite frequency transfer over a very long baseline *Metrologia* 51 253-62
- [36] Foreman S M, Ludlow A D, de Miranda M H G, Stalnaker J E, Diddams S A and Ye J 2007 Coherent optical phase transfer over a 32-km fiber with 1 s instability at 10^{-17} *Phys. Rev. Lett.* 99 153601
- [37] Newbury N R, Williams P A and Swann W C 2007 Coherent transfer of an optical carrier over 251 km *Opt. Lett.* 32 3056-8
- [38] Musha M, Hong F L, Nakagawa K and Ueda K 2008 Coherent optical frequency transfer over 50-km physical distance using a 120-km-long installed telecom fiber network *Opt. Express* 16 16459-66
- [39] Lisdat C, Grosche G, Quintin N, Shi C, Raupach S M F, Grebing C, Nicolodi D, Stefani F, Al-Masoudi A, Dörscher S, Häfner S, Robyr J L, Chiodo N, Bilicki S, Bookjans E, Koczwara A, Koke S, Kuhl A, Wiotte F, Meynadier F, Camisard E, Abgrall M, Lours M, Legero T, Schnatz H, Sterr U, Denker H, Chardonnet C, Le Coq Y, Santarelli G, Amy-Klein A, Le Targat R, Lodewyck J, Lopez O and Pottie P E 2016 A clock network for geodesy and fundamental science *Nature Communications* 7 12443
- [40] Poli N, Schioppo M, Vogt S, Falke S, Sterr U, Lisda, C, Tino G M 2014 A transportable strontium optical lattice clock *Appl. Phys. B* 117 1107-16
- [41] <http://www.bipm.fr/en/publications/mep.html>
- [42] Jiang J, Onae A, Matsumoto H and Hong F L 2005 Frequency measurement of acetylene-stabilized lasers using a femtosecond optical comb without carrier-envelope offset frequency control *Opt. Express* 13 1958-65
- [43] Margolis H S and Gill P 2015 Least-squares analysis of clock frequency comparison data

- to deduce optimized frequency and frequency ratio values *Metrologia* 52 628-34
- [44] Katori H 2002 *Proc. 6th Symp. Frequency Standards and Metrology* ed. P. Gill (World Scientific, Singapore, 2002) p. 323.
- [45] Katori H, Ido T and Kuwata-Gonokami M 1999 Optimal design of dipole potentials for efficient loading of Sr atoms *J. Phys. Soc. Jpn.* 68 2479-82
- [46] Ido T and Katori H 2003 Recoil-free spectroscopy of neutral Sr atoms in the Lamb-Dicke regime *Phys. Rev. Lett.* 91 053001
- [47] Katori H, Takamoto M, Pal'chikov V G and Ovsiannikov V D 2003 Ultrastable optical clock with neutral atoms in an engineered light shift trap *Phys. Rev. Lett.* 91 173005
- [48] Dicke R H 1953 The effect of collisions upon the Doppler width of spectral lines *Phys. Rev.* 89 472-3
- [49] Akamatsu D, Yasuda M, Kohno T, Onae A and Hong F L 2011 A compact light source at 461 nm using a periodically poled LiNbO₃ waveguide for strontium magneto-optical trapping *Opt. Express* 19 20-51
- [50] Courty I, Quessada A, Kovacich R P, Bruschi A, Kolker D, Zondy J J, Rovera G D and Lemonde P 2003 Clock transition for a future optical frequency standard with trapped atoms *Phys. Rev. A* 68 030501
- [51] Takamoto M and Katori H 2003 Spectroscopy of the $^1S_0 - ^3P_0$ clock transition of ^{87}Sr in an optical lattice *Phys. Rev. Lett.* 91 223001
- [52] Ludlow A D, Boyd M M, Zelevinsky T, Foreman S M, Blatt S, Notcutt M, Ido T and Ye J 2006 Systematic study of the ^{87}Sr clock transition in an optical lattice *Phys. Rev. Lett.* 96 033003
- [53] Le Targat R, Baillard X, Fouche M, Bruschi A, Tcherbakoff O, Rovera G D, and Lemonde P 2006 Accurate optical lattice clock with ^{87}Sr atoms *Phys. Rev. Lett.* 97 130801
- [54] International Committee for Weights and Measures (CIPM) Report of the 95th meeting (2006) <http://www.bipm.org/utis/en/pdf/CIPM/CIPM2006-EN.pdf>
- [55] Baillard X, Fouché M, Targat R L, Westergaard P G, Lecallier A, Chapelet F, Abgrall M, Rovera G D, Laurent P, Rosenbusch P, Bize S, Santarelli G, Clairon A, Lemonde P, Grosche G, Lipphardt B and Schnatz H 2008 An optical lattice clock with spin-polarized ^{87}Sr atoms *Eur. Phys. J. D* 48 11-17
- [56] Campbell G K, Ludlow A D, Blatt S, Thomsen J W, Martin M J, de Miranda M H G, Zelevinsky T, Boyd M M, Ye J, Diddams S A, Heavner T P, Parker T E and Jefferts S R 2008 The absolute frequency of the ^{87}Sr optical clock transition *Metrologia* 45 539-48
- [57] Hong F L, Musha M, Takamoto M, Inaba H, Yanagimachi S, Takamizawa A, Watabe K, Ikegami T, Imae M, Fujii Y, Amemiya M, Nakagawa K, Ueda K and Katori H 2009 Measuring the frequency of a Sr optical lattice clock using a 120 km coherent optical transfer *Opt. Lett.* 34 692-4

- [58] Falke S, Schnatz H, Vellore Winfred J S R, Middelmann T, Vogt S, Weyers S, Lipphardt B, Grosche G, Riehle F, Sterr U and Lisdat C 2011 The ^{87}Sr optical frequency standard at PTB *Metrologia* 48, 399-407
- [59] Yamaguchi A, Shiga N, Nagano S, Li Y, Ishijima H, Hachisu H, Kumagai M and Ido T 2012 Stability transfer between two clock lasers operating at different wavelengths for absolute frequency measurement of clock transition in ^{87}Sr *Appl. Phys. Express* 5 022701
- [60] Le Targat R, Lorini L, Le Coq Y, Zawada M, Guéna J, Abgrall M, Gurov M, Rosenbusch P, Rovera D G, Nagórny B, Gartman R, Westergaard P G, Tobar M E, Lours M, Santarelli G, Clairon A, Bize S, Laurent P, Lemonde P and Lodewyck J 2013 Experimental realization of an optical second with strontium lattice clocks *Nature Com.* 4 2109
- [61] Falke S, Lemke N, Grebing C, Lipphardt B, Weyers S, Gerginov V, Huntemann N, Hagemann C, Al-Masoudi A, Häfner S, Vogt S, Sterr U and Lisdat C 2014 A strontium lattice clock with 3 times 10^{-17} inaccuracy and its frequency *New J. Phys.* 16 073023
- [62] Akamatsu D, Inaba H, Hosaka K, Yasuda M, Onae A, Suzuyama T, Amemiya M and Hong F L 2014 Spectroscopy and frequency measurement of the ^{87}Sr clock transition by laser linewidth transfer using an optical frequency comb *Appl. Phys. Express* 7 012401
- [63] Lin Y G, Wang Q, Li Y, Meng F, Lin B K, Zang E J, Sun Z, Fang F, Li T C and Fang Z J 2015 First evaluation and frequency measurement of the strontium optical lattice clock at NIM *Chin. Phys. Lett.* 32 090601
- [64] Hachisu H and Ido T 2015 Distributed optical frequency measurements to reduce the dead time uncertainty of frequency link *Jpn. J. Appl. Phys.* 54 112401
- [65] Tanabe T, Akamatsu D, Kobayashi T, Takamizawa A, Yanagimachi S, Ikegami T, Suzuyama T, Inaba H, Okubo S, Yasuda M, Hong F L, Onae A and Hosaka K 2015 Improved frequency measurement of the $^1\text{S}_0$ - $^3\text{P}_0$ clock transition in ^{87}Sr using the Cs fountain clock at NMIJ as a transfer oscillator *J. Phys. Soc. Jpn.* 84 115002
- [66] Grebing C, Al-Masoudi A, Dörscher S, Häfner S, Gerginov V, Weyers S, Lipphardt B, Riehle F, Sterr U and Lisdat C 2016 Realization of a timescale with an accurate optical lattice clock *Optica* 3 563-9
- [67] Hong F L, Inaba H, Hosaka K, Yasuda M, and Onae A 2009 Doppler-free spectroscopy of molecular iodine using a frequency-stable light source at 578 nm *Opt. Express* 17 1652-9
- [68] Hosaka K, Inaba H, Nakajima Y, Yasuda M, Kohno T, Onae A, and Hong F L 2010 Evaluation of the clock laser for an Yb lattice clock using an optical fibre comb *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 57 606-12
- [69] Kim E B, Lee W K, Park C Y, Yu D H and Park S E 2010 Narrow linewidth 578 nm light generation using frequency-doubling with a waveguide PPLN pumped by an optical injection-locked diode laser *Opt. Express* 18 10308-14
- [70] Nishikawa T, Ozawa A, Nishida Y, Asobe M, Hong F L and Hänsch T W 2009 Efficient

- 494 mW sum-frequency generation of sodium resonance radiation at 589 nm by using a periodically poled Zn:LiNbO₃ ridge waveguide *Opt. Express* 17 17792-800
- [71] Kobayashi T, Akamatsu D, Nishida Y, Tanabe T, Yasuda M, Hong F L and Hosaka K 2016 Second harmonic generation at 399 nm resonant on the $^1S_0-^1P_1$ transition of ytterbium using a periodically poled LiNbO₃ waveguide *Opt. Express* 24 12142-50
- [72] Yasuda M, Kohno T, Inaba H, Nakajima Y, Hosaka K, Onae A and Hong F L 2010 Fiber-comb-stabilized light source at 556 nm for magneto-optical trapping of ytterbium *J. Opt. Soc. Am. B* 27 1388-93
- [73] Kohno T, Yasuda M, Hosaka K, Inaba H, Nakajima Y and Hong F L 2009 One-dimensional optical lattice clock with a fermionic ^{171}Yb isotope *Appl. Phys. Express* 2 072501
- [74] Lemke N D, Ludlow A D, Barber Z W, Fortier T M, Diddams S A, Jiang Y, Jefferts S R, Heavner T P, Parker T E and Oates C W 2009 Spin-1/2 optical lattice clock *Phys. Rev. Lett.* 103 063001
- [75] Yasuda M, Inaba H, Kohno T, Tanabe T, Nakajima Y, Hosaka K, Akamatsu D, Onae A, Suzuyama T, Amemiya M and Hong F L 2012 Improved absolute frequency measurement of the ^{171}Yb optical lattice clock towards a candidate for the redefinition of the second *Appl. Phys. Express* 5 102401
- [76] Park C Y, Yu D H, Lee W K, Park S E, Kim E B, Lee S K, Cho J W, Yoon T H, Mun J, Park S J, Kwon T Y and Lee S B 2013 Absolute frequency measurement of $^1S_0(F=1/2)-^3P_0(F=1/2)$ transition of ^{171}Yb atoms in a one-dimensional optical lattice at KRISS *Metrologia* 50 119-28
- [77] Akamatsu D, Yasuda M, Inaba H, Hosaka K, Tanabe T, Onae A and Hong F L 2014 Frequency ratio measurement of ^{171}Yb and ^{87}Sr optical lattice clocks *Opt. Express* 22 7898-905
- [78] Akamatsu D, Yasuda M, Inaba H, Hosaka K, Tanabe T, Onae A and Hong F L 2014 Errata: Frequency ratio measurement of ^{171}Yb and ^{87}Sr optical lattice clocks *Opt. Express* 22 32199
- [79] Takamoto M, Ushijima I, Das M, Nemitz N, Ohkubo T, Yamanaka K, Ohmae N, Takano T, Akatsuka T, Yamaguchi A and Katori H 2015 Frequency ratios of Sr, Yb, and Hg based optical lattice clocks and their applications *C. R. Physique* 16 489-98
- [80] Pizzocaro M, Thoumany P, Rauf B, Bregolin F, Milani G, Clivati C, Costanzo G A, Levi F and Calonico D 2016 Absolute frequency measurement of the $^1S_0 - ^3P_0$ transition of ^{171}Yb arXiv:1609.01610
- [81] International Committee for Weights and Measures (CIPM) Report of the 102nd meeting (2013) <http://www.bipm.org/utis/en/pdf/CIPM/CIPM2013-EN.pdf>
- [82] Nemitz N, Ohkubo T, Takamoto M, Ushijima I, Das M, Ohmae N and Katori H 2016

- Frequency ratio of Yb and Sr clocks with 5×10^{-17} uncertainty at 150 seconds averaging time *Nature Photonics* 10 258-61
- [83] Tamm C, Huntemann N, Lipphardt B, Gerginov V, Nemitz N, Kazda M, Weyers S and Peik E 2014 A Cs-based optical frequency measurement using cross-linked optical and microwave oscillators *Phys. Rev. A* 89 023820
- [84] Godun R M, Nisbet-Jones P B R, Jones J M, King S A, Johnson L A M, Margolis H S, Szymaniec K, Lea S N, Bongs K and Gill P 2014 Frequency ratio of two optical clock transitions in $^{171}\text{Yb}^+$ and constraints on the time-variation of fundamental constants *Phys. Rev. Lett.* 113 210801
- [85] Working document: CCL-CCTF/06-11 (2006).
http://www.bipm.org/wg/CCL/CCL-CCTF/Allowed/2006/JWG_CCL-CCTF2006_questionnaire_PTb.pdf
- [86] Tamm Chr, Weyers S, Lipphardt B and Peik E 2009 Stray-field-induced quadrupole shift and absolute frequency of the 688-THz $^{171}\text{Yb}^+$ single-ion optical frequency standard *Phys. Rev. A* 80 043403.
- [87] Webster S, Godun R, King S, Huang G, Walton B, Tsaturian V, Margolis H, Lea S and Gill P 2010 Frequency measurement of the $^2\text{S}_{1/2}$ - $^2\text{D}_{3/2}$ electric quadrupole transition in a single $^{171}\text{Yb}^+$ ion *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* 57 592-9
- [88] Huntemann N, Lipphardt B, Chr. Tamm, Gerginov V, Weyers S and Peik E 2014 Improved limit on a temporal variation of m_p/m_e from comparisons of Yb⁺ and Cs atomic clocks *Phys. Rev. Lett.* 113 210802
- [89] Hosaka K, Webster S A, Stannard A, Walton B R, Margolis H S and Gill P 2009 Frequency measurement of the $^2\text{S}_{1/2}$ - $^2\text{F}_{7/2}$ electric octupole transition in a single $^{171}\text{Yb}^+$ ion *Phys. Rev. A* 79 033403
- [90] King S A, Godun R M, Webster S A, Margolis H S, Johnson L A M, Szymaniec K, Baird P E G and Gill P 2012 Absolute frequency measurement of the $^2\text{S}_{1/2}$ - $^2\text{F}_{7/2}$ electric octupole transition in a single ion of $^{171}\text{Yb}^+$ with 10^{-15} fractional uncertainty *New J. Phys.* 14 013045
- [91] Huntemann N, Okhapkin M, Lipphardt B, Weyers S, Tamm Chr and Peik E 2012 High-accuracy optical clock based on the octupole transition in $^{171}\text{Yb}^+$ *Phys. Rev. Lett.* 108 090801
- [92] Barwood G P, Huang G, Klein H A, Johnson L A M, King S A, Margolis H S, Szymaniec K and Gill P 2014 Agreement between two $^{88}\text{Sr}^+$ optical clocks to 4 parts in 10^{17} *Phys. Rev. A* 89 050501(R)
- [93] Madej A A, Dubé P, Zhou Z, Bernard J E and Gertszvolf M 2012 $^{88}\text{Sr}^+$ 445-THz single ion reference at the 10^{-17} level via control and cancellation of systematic uncertainties and its measurement against the SI second *Phys. Rev. Lett.* 109 203002
- [94] Dubé P, Madej A A, Zhou Z and Bernard J E 2013 Evaluation of systematic shifts of the

- $^{88}\text{Sr}^+$ single-ion optical frequency standard at the 10^{-17} level *Phys. Rev. A* 87 023806
- [95] Margolis H S, Barwood G P, Huang G, Klein H A, Lea S N, Szymaniec K and Gill P 2004 Hertz-level measurement of the optical clock frequency in a single $^{88}\text{Sr}^+$ ion *Science* 306 1355-8
- [96] Dubé P, Madej A A, Bernard J E, Marmet L, Boulanger J S and Cundy S 2005 Electric quadrupole shift cancellation in single-ion optical frequency standards *Phys. Rev. Lett.* 95 033001
- [97] Rosenband T, Hume D B, Schmidt P O, Chou C W, Brusch A, Lorini L, Oskay W H, Drullinger R E, Fortier T M, Stalnaker J E, Diddams S A, Swann W C, Newbury N R, Itano W M, Wineland D J and Bergquist J C 2008 Frequency ratio of Al^+ and Hg^+ single-ion optical clocks; Metrology at the 17th decimal place *Science* 319 1808-12
- [98] Schmidt P O, Rosenband T, Langer C, Itano W M, Bergquist J C and Wineland D J 2005 Spectroscopy Using Quantum Logic *Science* 309 749-52
- [99] Stalnaker J E, Diddams S A, Fortier T M, Kim K, Hollberg L, Bergquist J C, Itano W M, Delany M J, Lorini L, Oskay W H, Heavner T P, Jefferts S R, Levi F, Parker T E and Shirley J 2007 Optical-to-microwave frequency comparison with fractional uncertainty of 10^{-15} *Appl. Phys. B* 89 167-76
- [100] Rosenband T, Schmidt P O, Hume D B, Itano W M, Fortier T M, Stalnaker J E, Kim K, Diddams S A, Koelemeij J C J, Bergquist J C and Wineland D J 2007 Observation of the $^1\text{S}_0 \rightarrow ^3\text{P}_0$ clock transition in $^{27}\text{Al}^+$ *Phys. Rev. Lett.* 98 220801
- [101] Tanaka U, Kitanaka T, Hayasaka K and Urabe S 2015 Sideband cooling of a $\text{Ca}^+\text{-In}^+$ ion chain toward the quantum logic spectroscopy of In^+ *Appl. Phys. B* 121 147-53
- [102] Pyka K, Herschbach N, Keller J, Tanja E and Mehlstäubler T E 2014 A high-precision segmented Paul trap with minimized micromotion for an optical multiple-ion clock *Appl. Phys. B* 114 231-41
- [103] Yamanaka K, Ohmae N, Ushijima I, Takamoto M and Katori H 2015 Frequency ratio of ^{199}Hg and ^{87}Sr optical lattice clocks beyond the SI limit *Phys. Rev. Lett.* 114 230801
- [104] Tyumenov R, Favier M, Bilicki S, Bookjans E, Le Targat R, Lodewyck J, Nicolodi D, Le Coq Y, Abgrall M, Guéna J, De Sarlo L and Bize S 2016 Comparing a mercury optical lattice clock with microwave and optical frequency standards arXiv:1603.02026v1
- [105] McFerran J J, Yi L, Mejri S, Di Manno S, Zhang W, Guéna J, Le Coq Y and Bize S 2012 Neutral atom frequency reference in the deep ultraviolet with fractional uncertainty = 5.7×10^{-15} *Phys. Rev. Lett.* 108 183004
- [106] McFerran J J, Yi L, Mejri S, Di Manno S, Zhang W, Guéna J, Le Coq Y and Bize S 2015 Erratum: Neutral atom frequency reference in the deep ultraviolet with fractional uncertainty = 5.7×10^{-15} [*Phys. Rev. Lett.* 108, 183004 (2012)] *Phys. Rev. Lett.* 115 219901
- [107] Chwalla M, Benhelm J, Kim K, Kirchmair G, Monz T, Riebe M, Schindler P, Villar A S,

- Hänsel W, Roos C F, Blatt R, Abgrall M, Santarelli G, Rovera G D and Laurent Ph 2009 Absolute frequency measurement of the $^{40}\text{Ca}^+ 4s\ ^2\text{S}_{1/2}-3d\ ^2\text{D}_{5/2}$ clock transition *Phys. Rev. Lett.* 102 023002
- [108] Matsubara K, Hachisu H, Li Y, Nagano S, Locke C, Nogami A, Kajita M, Hayasaka K, Ido T and Hosokawa M 2012 Direct comparison of a Ca^+ single-ion clock against a Sr lattice clock to verify the absolute frequency measurement *Opt. Express* 20 22034-41
- [109] Huang Y, Guan H, Liu P, Bian W, Ma L, Liang K, Li T and Gao K 2016 Frequency comparison of two $^{40}\text{Ca}^+$ optical clocks with an uncertainty at the 10^{-17} Level *Phys. Rev. Lett.* 116 013001
- [110] Huang Y, Cao J, Liu P, Liang K, Ou B, Guan H, Huang X, Li T and Gao K 2012 Hertz-level measurement of the $^{40}\text{Ca}^+ 4s\ ^2\text{S}_{1/2}-3d\ ^2\text{D}_{5/2}$ clock transition frequency with respect to the SI second through the global positioning system *Phys. Rev. A* 85 030503(R)
- [111] Matveev A, Parthey C G, Predehl K, Alnis J, Beyer A, Holzwarth R, Udem T, Wilken T, Kolachevsky N, Abgrall M, Rovera D, Salomon C, Laurent P, Grosche G, Terra O, Legero T, Schnatz H, Weyers S, Altschul B and Hänsch T W 2013 Precision measurement of the hydrogen $^1\text{S} - ^2\text{S}$ frequency via a 920-km fiber link *Phys. Rev. Lett.* 110 230801
- [112] Parthey C G, Matveev A, Alnis J, Bernhardt B, Beyer A, Holzwarth R, Maistrou A, Pohl R, Predehl K, Udem T, Wilken T, Kolachevsky N, Abgrall M, Rovera D, Salomon C, Laurent P and Hänsch T W 2011 Improved measurement of the hydrogen $^1\text{S}-^2\text{S}$ transition frequency *Phys. Rev. Lett.* 107 203001
- [113] Takahata K, Kobayashi T, Sasada H, Nakajima Y, Inaba H and Hong F L 2009 Absolute frequency measurement of sub-Doppler molecular lines using a $3.4\text{-}\mu\text{m}$ difference-frequency-generation spectrometer and a fiber-based frequency comb *Phys. Rev. A* 80 032518
- [114] Sera H, Abe M, Iwakuni K, Okubo S, Inaba H, Hong F L and Sasada H 2015 Sub-Doppler resolution mid-infrared spectroscopy using a difference-frequency-generation source spectrally narrowed by laser linewidth transfer *Opt. Lett.* 40 5467-70
- [115] Ye J, Robertsson L, Picard S, Ma L S and Hall J L 1999 Absolute frequency atlas of I_2 molecular lines at 532 nm *IEEE Trans. Instrum. Meas.* 48 544-8
- [116] Zhang Y, Ishikawa J and Hong F L 2001 Accurate frequency atlas of molecular iodine near 532 nm measured by an optical frequency comb generator *Opt. Commun.* 200 209-15
- [117] Eickhoff M L and Hall J L 1995 Optical frequency standard at 532 nm *IEEE Trans. Instrum. Meas.* 44 155-8
- [118] Hong F L, Ishikawa J, Zhang Y, Guo R X, Onae A and Matsumoto H 2004 Frequency reproducibility of an iodine-stabilized Nd:YAG laser at 532 nm *Opt. Commun.* 235 377-85
- [119] Hong F L, Ye J, Ma L S, Picard S, Borde Ch J and Hall J L 2001 Rotation dependence of electric quadrupole hyperfine interaction in the ground state of molecular iodine by

- high-resolution laser spectroscopy *J. Opt. Soc. Am. B* 18 379-87
- [120] Hong F L and Ishikawa J 2000 Hyperfine structures of the R(122)35-0 and P(84)33-0 transitions of $^{127}\text{I}_2$ near 532 nm *Opt. Commun.* 183 101-8
- [121] Hong F L, Ishikawa J, Onae A and Matsumoto H 2001 Rotation dependence of the excited-state electric quadrupole hyperfine interaction by high-resolution laser spectroscopy of $^{127}\text{I}_2$ *J. Opt. Soc. Am. B* 18 1416-22
- [122] Hong F L, Zhang Y, Ishikawa J, Onae A and Matsumoto H 2002 Vibration dependence of the tensor spin–spin and scalar spin–spin hyperfine interactions by precision measurement of hyperfine structures of $^{127}\text{I}_2$ near 532 nm *J. Opt. Soc. Am. B* 19 946-53
- [123] Hong F L, Zhang Y, Ishikawa J, Onae A and Matsumoto H 2002 Hyperfine structure and absolute frequency determination of the R(121)35-0 and P(142)37-0 transitions of $^{127}\text{I}_2$ near 532 nm *Opt. Commun.* 212 89-95
- [124] Hong F L, Diddams S A, Guo R, Bi Z Y, Onae A, Inaba H, Ishikawa J, Okumura K, Katsuragi D, Hirata J, Shimizu T, Kurosu T, Koga Y and Matsumoto H 2004 Frequency measurements and hyperfine structure of the R(85)33–0 transition of molecular iodine with a femtosecond optical comb *J. Opt. Soc. Am. B* 21 88-95
- [125] Chen L, Cheng W Y and Ye J 2004 Hyperfine interactions and perturbation effects in the $\text{B}0_u^+(3\Pi_u)$ state of $^{127}\text{I}_2$ *J. Opt. Soc. Am. B* 21 820-32
- [126] Hong F L, Ishikawa J, Yoda J, Ye J, Ma L S and Hall J L 1999 Frequency comparison of $^{127}\text{I}_2$ -stabilized Nd:YAG lasers *IEEE Trans. Instrum. Meas.* 48 532-6
- [127] Hong F L, Ishikawa J, Bi Z Y, Zhang J, Seta K, Onae A, Yoda J and Matsumoto H 2001 Portable I_2 -stabilized Nd:YAG laser for international comparisons *IEEE Trans. Instrum. Meas.* 50 486-9
- [128] Robertsson L, Picard S, Hong F L, Millerieux Y, Juncar P and Ma L S 2001 International comparison of $^{127}\text{I}_2$ -stabilized frequency-doubled Nd:YAG lasers between the BIPM, the NRLM and the BNM-INM, October 2000 *Metrologia* 38 567-72
- [129] Hong F L, Ishikawa J, Sugiyama K, Onae A, Matsumoto H, Ye J and Hall J L 2003 Comparison of independent optical frequency measurement using a portable iodine-stabilized Nd:YAG laser *IEEE Trans. Instrum. Meas.* 52 240-4
- [130] Kobayashi T, Akamatsu D, Hosaka K, Inaba H, Okubo S, Tanabe T, Yasuda M, Onae A and Hong F L 2015 Compact iodine-stabilized laser operating at 531 nm with stability at the 10^{-12} level and using a coin-sized laser module *Opt. Express* 23 20749-59
- [131] Bitou Y, Kobayashi T, and Hong F L 2016 Compact and inexpensive iodine-stabilized diode laser system with an output at 531 nm for gauge block interferometers *Precision Engineering* Article in press and available online: <http://dx.doi.org/10.1016/j.precisioneng.2016.07.008>
- [132] Bitou Y, Sasaki K, Inaba H, Hong F L and Onae A 2004 Rubidium stabilized diode laser

- for high precision interferometer *Opt. Eng.* 43 900-3
- [133] Ye J, Swartz S, Jungner P and Hall J L 1996 Hyperfine structure and absolute frequency of the ^{87}Rb $5P_{3/2}$ state *Opt. Lett.* 21 1280-2
- [134] Sasada H and Yamada K 1990 Calibration lines of HCN in the 1.5- μm region *Appl. Opt.* 29 3535-47
- [135] Gilbert S L, Swann W C 2001 Acetylene $^{12}\text{C}_2\text{H}_2$ absorption reference for 1510 nm to 1540 nm wavelength calibration—SRM 2517a *NIST Special Publication* 260-133 <http://www.nist.gov/srm/upload/SP260-133.PDF>
- [136] du Burck F, Daussy C, Amy-Klein A, Goncharov A N, Lopez O, Chardonnet C and Wallerand J P 2005 Frequency measurement of an Ar^+ laser stabilized on narrow lines of molecular iodine at 501.7 nm *IEEE Trans. Instrum. Meas.* 54 754-8
- [137] Kobayashi T, Akamatsu D, Hosaka K, Inaba H, Okubo S, Tanabe T, Yasuda M, Onae A and Hong F L 2016 Absolute frequency measurements and hyperfine structures of the molecular iodine transitions at 578 nm *J. Opt. Soc. Am. B* 33 725-34
- [138] Guo R, Hong F L, Onae A, Bi Z Y, Matsumoto H and Nakagawa K 2004 Frequency stabilization of a 1319-nm Nd:YAG laser by saturation spectroscopy of molecular iodine *Opt. Lett.* 29 1733-5
- [139] Rovera G D, Santarelli G, Clairon A 1994 A laser-diode system stabilized on the cesium D_2 line *Rev. Sci. Instrum.* 65 1502-5
- [140] Cariou J and Luc P 1980 Atlas du Spectre d’Absorption de la Molécule Tellure (Laboratoire Aime-Cotton, CNRS II, Orsay, France)
- [141] Gillaspay J D and Sansonetti C J 1991 Absolute wavelength determinations in molecular tellurium: new reference lines for precision laser spectroscopy *J. Opt. Soc. Am. B* 8 2414-9
- [142] Palmer B A and Engleman R Jr. 1983 Atlas of the thorium spectrum, Los Alamos Scientific Laboratory Rep. LA-9615 (Los Alamos National Laboratory, Los Alamos, N.M.)
- [143] Lovis C and Pepe F 2007 A new list of thorium and argon spectral lines in the visible *Astronomy and Astrophysics* 468 1115-21
- [144] Akatsuka T, Takamoto M and Katori H 2008 Optical lattice clocks with non-interacting bosons and fermions *Nature Physics* 4 954-9
- [145] Ma L S, Jungner P, Ye J and Hall J L 1994 Delivering the same optical frequency at two places: accurate cancellation of phase noise introduced by an optical fiber or other time-varying path *Opt. Lett.* 19 1777-9
- [146] Ye J, Peng J L, Jones R J, Holman K W, Hall J L, Jones D J, Diddams S A, Kitching J, Bize S, Bergquist J C, Hollberg L W, Robertsson L and Ma L S 2003 Delivery of high-stability optical and microwave frequency standards over an optical fiber network *J. Opt. Soc. Am. B* 20 1459-67
- [147] Ludlow A D, Zelevinsky T, Campbell G K, Blatt S, Boyd M M, de Miranda M H G,

- Martin M J, Thomsen J W, Foreman S M, Ye J, Fortier T M, Stalnaker J E, Diddams S A, Le Coq Y, Barber Z W, Poli N, Lemke N D, Beck K M and Oates C W 2008 Sr lattice clock at 1×10^{-16} fractional uncertainty by remote optical evaluation with a Ca clock *Science* 319 1805-8
- [148] Yamaguchi A, Fujieda M, Kumagai M, Hachisu H, Nagano S, Li Y, Ido T, Takano T, Takamoto M and Katori H 2011 Direct comparison of distant optical lattice clocks at the 10^{-16} uncertainty *Appl. Phys. Express* 4 082203
- [149] Predehl K, Grosche G, Raupach S M F, Droste S, Terra O, Alnis J, Legero Th, Hänsch T W, Udem Th, Holzwarth R and Schnatz H 2012 A 920-kilometer optical fiber link for frequency metrology at the 19th decimal place *Science* 336 441-4
- [150] Clivati C, Cappellini G, Livi L F, Poggiali F, de Cumis M S, Mancini M, Pagano G, Frittelli M, Mura A, Costanzo G A, Levi F, Calonico D, Fallani L, Catani J and Inguscio M 2016 Measuring absolute frequencies beyond the GPS limit via long-haul optical frequency dissemination *Opt. Express* 24 11865-75
- [151] Laurent P, Massonnet D, Cacciapuoti L and Salomon C 2015 The ACES/PHARAO space mission *C. R. Physique* 16 540-52
- [152] Consultative Committee for Time and Frequency (CCTF) Report of the 20th meeting (2015) <http://www.bipm.org/utils/common/pdf/CC/CCTF/CCTF20.pdf>
- [153] Grebing C, Al-Masoudi A, Dörscher S, Häfner S, Gerginov V, Weyers S, Lipphardt B, Riehle F, Sterr U and Lisdat C 2016 Realization of a timescale with an accurate optical lattice clock *Optica* 3 563-9
- [154] Lodewyck J, Bilicki S, Bookjans E, Robyr J-L, Shi C, Vallet G, Targat R L, Nicolodi D, Coq Y L, Guéna J, Abgrall M, Rosenbusch P and Bize S 2016 Optical to microwave clock frequency ratios with a nearly continuous strontium optical lattice clock *Metrologia* 53 1123-30
- [155] Katori H, Ovsiannikov V D, Marmo S I and Palchikov V G 2015 Strategies for reducing the light shift in atomic clocks *Phys. Rev. A* 91 052503
- [156] Peik E and Tamm Chr 2003 Nuclear laser spectroscopy of the 3.5 eV transition in Th-229 *Europhys. Lett.* 61 181-6
- [157] von der Wense L, Seiferle B, Laatiaoui M, Neumayr J B, Maier H J, Wirth H F, Mokry C, Runke J, Eberhardt K, Düllmann C E, Trautmann N G, Thirolf Peter G 2016 Direct detection of the ^{229}Th nuclear clock transition *Nature* 533 47-51
- [158] Derevianko A, Dzuba V A and V. V. Flambaum 2012 Highly Charged ions as a basis of optical atomic clockwork of exceptional accuracy *Phys. Rev. Lett.* 109 180801
- [159] VDzuba V A, Flambaum V V and Katori H 2015 Optical clock sensitive to variations of the fine-structure constant based on the Ho^{14+} ion *Phys. Rev. A* 91 022119
- [160] Kolkowitz S, Pikovski I, Langellier N, Lukin M D, Walsworth R L and Ye J 2016

Gravitational wave detection with optical lattice atomic clocks arXiv:1606.01859

- [161] Derevianko A and Pospelov M 2014 Hunting for topological dark matter with atomic clocks *Nature Phys.* 10 933-6
- [162] Nakajima Y, Inaba H, Hosaka K, Minoshima K, Onae A, Yasuda M, Kohno T, Kawato S, Kobayashi T, Katsuyama T and Hong F L 2010 A multi-branch, fiber-based frequency comb with millihertz-level relative linewidths using an intra-cavity electro-optic modulator *Opt. Express* 18 1667-76
- [163] Iwakuni K, Inaba H, Nakajima Y, Kobayashi T, Hosaka K, Onae A and Hong F L 2012 Narrow linewidth comb realized with a mode-locked fiber laser using an intra-cavity waveguide electro-optic modulator for high-speed control *Opt. Express* 20 13769-76
- [164] Inaba H, Hosaka K, Yasuda M, Nakajima Y, Iwakuni K, Akamatsu D, Okubo S, Kohno T, Onae A and Hong F L 2013 Spectroscopy of ^{171}Yb in an optical lattice based on laser linewidth transfer using a narrow linewidth frequency comb *Opt. Express* 21 7891-6
- [165] Steinmetz T, Wilken T, Araujo-Hauck C, Holzwarth R, Hänsch T W, Pasquini L, Manescau A, D'Odorico S, Murphy M T, Kentischer T, Schmidt W and Udem Th 2008 Laser Frequency Combs for Astronomical Observations *Science* 321 1335-7
- [166] Li C H, Benedick A J, Fendel P, Glenday A G, Kärtner F X, Phillips D F, Sasselov D, Szentgyorgyi A and Walsworth R L 2008 A laser frequency comb that enables radial velocity measurements with a precision of 1 cm s^{-1} *Nature* 452 610-2
- [167] Keilmann F, Gohle C and Holzwarth R 2004 Time-domain mid-infrared frequency-comb spectrometer *Opt. Lett.* 29 1542-4
- [168] Coddington I, Swann W C and Newbury N R 2008 Coherent multiheterodyne spectroscopy using stabilized optical frequency combs *Phys Rev Lett.* 100 013902
- [169] Okubo S, Iwakuni K, Inaba J, Hosaka K, Onae A, Sasada H and Hong F L 2015 Ultra-broadband dual-comb spectroscopy across $1.0\text{--}1.9 \mu\text{m}$ *Appl. Phys. Express* 8 082402
- [170] Iwakuni K, Okubo S, Yamada K M T, Inaba H, Onae A, Hong F L and Sasada H 2016 Ortho-para-dependent pressure effects observed in the near infrared band of acetylene by dual-comb spectroscopy *Phys. Rev. Lett.* 117 143902
- [171] Yamada K M T, Onae A, Hong F L, Inaba H, Matsumoto H, Nakajima Y, Ito F and Shimizu T 2008 High precision line profile measurements on ^{13}C acetylene using a near infrared frequency comb spectrometer *J. Mol. Spectrosc.* 249 95-9
- [172] Yamada K M T, Onae A, Hong F L, Inaba H and Shimizu T 2009 Precise determination of the Doppler width of a rovibrational absorption line using a comb-locked diode laser *C. R. Physique* 10 907-15
- [173] Hartland G V, Qin D and Dai H L 1994 Collisional deactivation of highly vibrationally excited NO_2 monitored by time-resolved Fourier transform infrared emission spectroscopy *J. Chem. Phys.* 100 7832-5

Table 1. Laser frequency standards recommended by the International Committee for Weights and Measures (CIPM) in 2015. SRS is a secondary representation of the second.

Application	λ	Laser and reference	Spectroscopy	Frequency	Uncertainty
SRS	267 nm	$^{27}\text{Al}^+$, $3s^2\ ^1\text{S}_0 - 3s3p\ ^3\text{P}_0$	Ion trap	1121015393207857.3 Hz	1.9×10^{-15}
	282 nm	$^{199}\text{Hg}^+$, $5d^{10}6s\ ^2\text{S}_{1/2} (F=0) - 5d^96s^2\ ^2\text{D}_{5/2} (F=2)$	Ion trap	1064721609899145.30 Hz	1.9×10^{-15}
	436 nm	$^{171}\text{Yb}^+$, $6s\ ^2\text{S}_{1/2} (F=0) - 5d\ ^2\text{D}_{3/2} (F=2)$	Ion trap	688358979309308.3 Hz	6×10^{-16}
	467 nm	$^{171}\text{Yb}^+$, $6s\ ^2\text{S}_{1/2} (F=0) - 4f^{13}6s^2\ ^2\text{F}_{7/2} (F=3)$	Ion trap	642121496772645.0 Hz	6×10^{-16}
	578 nm	^{171}Yb , $6s^2\ ^1\text{S}_0 (F=1/2) - 6s6p\ ^3\text{P}_0 (F=1/2)$	Optical lattice	518295836590864.0 Hz	2×10^{-15}
	674 nm	$^{88}\text{Sr}^+$, $5s\ ^2\text{S}_{1/2} - 4d\ ^2\text{D}_{5/2}$	Ion trap	444779044095486.6 Hz	1.6×10^{-15}
	698 nm	^{87}Sr , $5s^2\ ^1\text{S}_0 (F=9/2) - 5s5p\ ^3\text{P}_0 (F=9/2)$	Optical lattice	429228004229873.2 Hz	5×10^{-16}
Time	237 nm	$^{115}\text{In}^+$, $5s^2\ ^1\text{S}_0 - 5s5p\ ^3\text{P}_0$	Ion trap	1267402452899.92 kHz	3.6×10^{-13}
	243 nm	^1H , $1\text{S} - 2\text{S}$, 2 photon	Atomic beam, two photons	1233030706593514 Hz	9×10^{-15}
	266 nm	^{199}Hg , $6s^2\ ^1\text{S}_0 - 6s6p\ ^3\text{P}_0$	Optical lattice	1128575290808154.8 Hz	6×10^{-16}
	657 nm	^{40}Ca , $^1\text{S}_0 - ^3\text{P}_1$, $\Delta m_J = 0$	Cold atoms	455986240494140 Hz	1.8×10^{-14}
	698 nm	^{88}Sr , $5s^2\ ^1\text{S}_0 - 5s5p\ ^3\text{P}_0$	Optical lattice	429228066418012 Hz	1×10^{-14}
	729 nm	$^{40}\text{Ca}^+$, $4s\ ^2\text{S}_{1/2} - 3d\ ^2\text{D}_{5/2}$	Ion trap	411042129776398.4 Hz	1.2×10^{-14}
Length & others	531 nm	Diode laser, $^{127}\text{I}_2$, R(36)32-0:a ₁	Saturation absorption	564074632.42 MHz	1×10^{-10}
	532 nm	Nd:YAG laser, $^{127}\text{I}_2$, R(56)32-0:a ₁₀	Saturation absorption	563260223513 kHz	8.9×10^{-12}
	543 nm	He-Ne laser, $^{127}\text{I}_2$, R(106)28-8:b ₁₀	Saturation absorption	551580162400 kHz	4.5×10^{-11}
	633 nm	He-Ne laser, $^{127}\text{I}_2$, R(127)11-5:a ₁₆	Saturation absorption	473612353604 kHz	2.1×10^{-11}
	778 nm	^{85}Rb , $5\text{S}_{1/2}(F=3) - 5\text{D}_{5/2}(F=5)$, 2 photon	Two photons	385285142375 kHz	1.3×10^{-11}
	780 nm	^{87}Rb , $5\text{S}_{1/2} - 5\text{P}_{3/2}$, d/f crossover	Saturation absorption	384227981.9 MHz	5×10^{-10}
	1.54 μm	$^{13}\text{C}_2\text{H}_2$, P(16)($v_1 + v_3$)	Saturation absorption	194369569384 kHz	2.6×10^{-11}
	3.39 μm	He-Ne laser, CH_4 , n_3 , P(7), F ₂ ⁽²⁾	Saturation absorption	88376181600.18 kHz	3×10^{-12}

Figure captions

Fig.1 Technologies used in high-resolution laser spectroscopy and their applications.

Fig. 2 Standards and measurement tools involved in optical frequency metrology. When optical clocks are compared with microwave clocks or other optical clocks in the same laboratory, optical frequency combs are used to link different frequencies. When clocks in different laboratories are compared, optical fiber, satellites or transportable clocks are used for frequency link.

Fig. 3 Energy levels of ^{87}Sr . Wavelengths and natural linewidths are indicated for the relevant cooling, trapping, and clock transitions.

Fig. 4 Absolute frequency measurements and the CIPM recommendation value of the $5s^2\ ^1S_0$ ($F=9/2$) - $5s5p\ ^3P_0$ ($F=9/2$) transition in ^{87}Sr .

Fig. 5 Energy levels of ^{171}Yb . Wavelengths and natural linewidths are indicated for the relevant cooling, trapping, and clock transitions.

Fig. 6 Absolute frequency measurements and the CIPM recommendation value of the $6s^2\ ^1S_0$ ($F=1/2$) - $6s6p\ ^3P_0$ ($F=1/2$) transition in ^{171}Yb .

Fig. 7 Absolute frequency measurements and the CIPM recommendation value of the $6s\ ^2S_{1/2}$ ($F=0$) - $5d\ ^2D_{3/2}$ ($F=2$) quadrupole transition in $^{171}\text{Yb}^+$.

Fig. 8 Absolute frequency measurements and the CIPM recommendation value of the $6s\ ^2S_{1/2}$ ($F=0$) - $4f^{13}6s^2\ ^2F_{7/2}$ ($F=3$) octupole transition in $^{171}\text{Yb}^+$.

Fig. 9 Absolute frequency measurements and the CIPM recommendation value of the $5s\ ^2S_{1/2}$ - $4d\ ^2D_{5/2}$ transition in $^{88}\text{Sr}^+$.

Fig. 10 Absolute frequency measurements and the CIPM recommendation value of the $6s^2\ ^1S_0 - 6s6p\ ^3P_0$ transition in ^{199}Hg .

Fig. 11 Absolute frequency measurements and the CIPM recommendation value of the $4s\ ^2S_{1/2} - 3d\ ^2D_{5/2}$ transition in $^{40}\text{Ca}^+$.

Fig. 12 Relation between the optical frequency standards and the SI second. Solid lines indicate the absolute frequency measurement based on Cs atomic clocks, while dashed lines indicate the frequency ratio measurement between optical frequency standards.

Fig. 13 Frequency ratio of ^{171}Yb and ^{87}Sr optical lattice clocks.

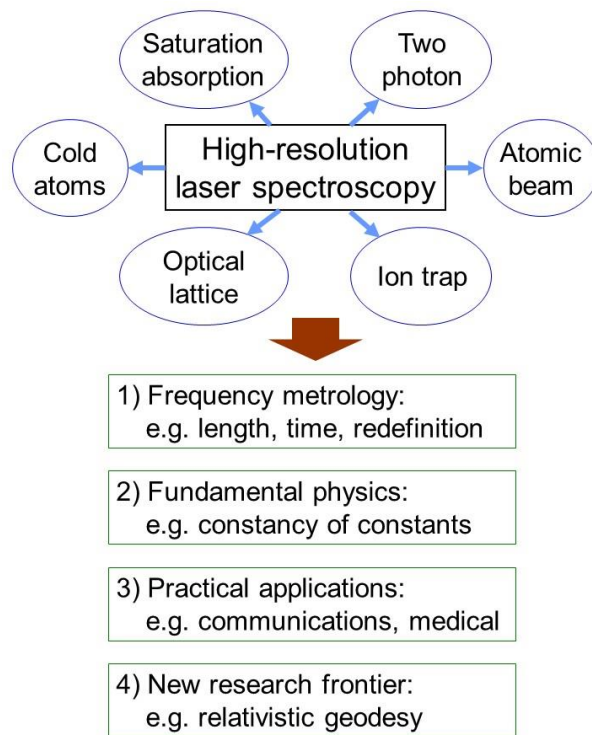


Fig. 1

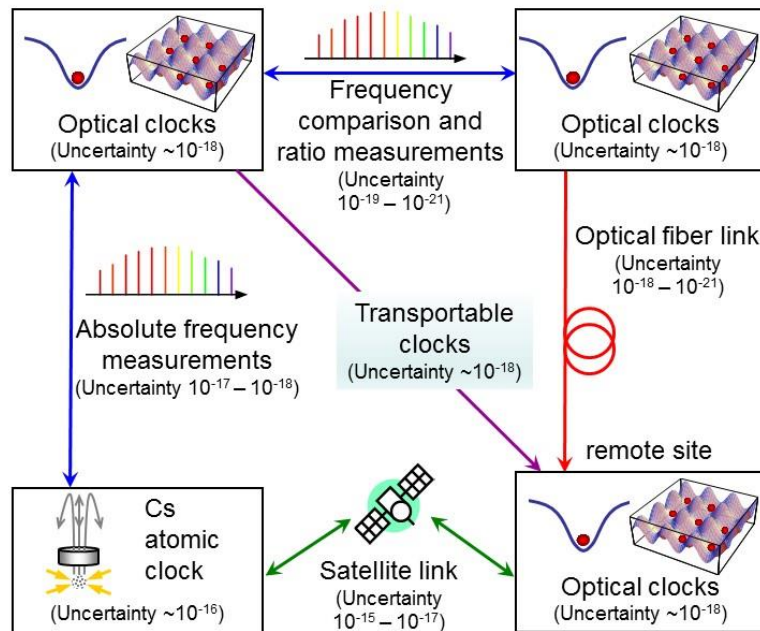


Fig. 2

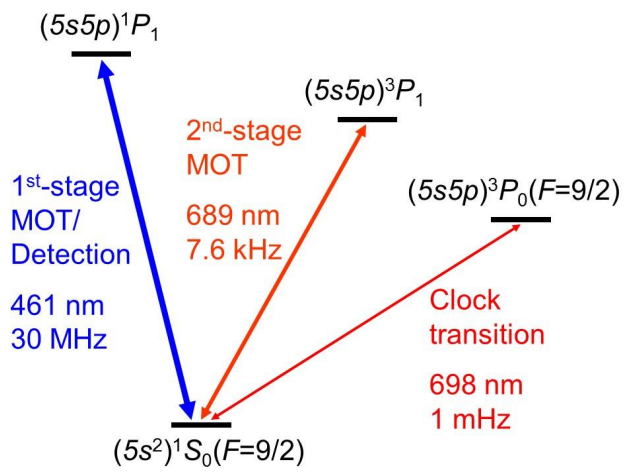


Fig. 3

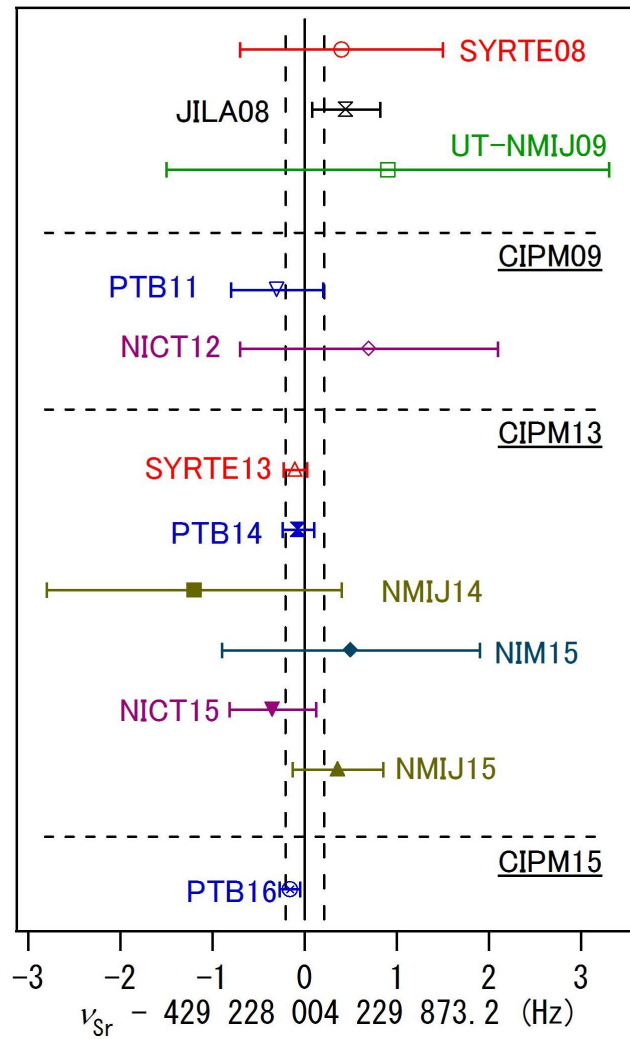


Fig. 4

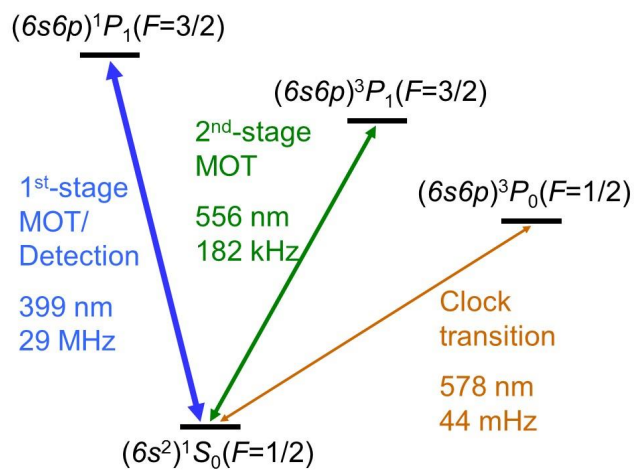


Fig. 5

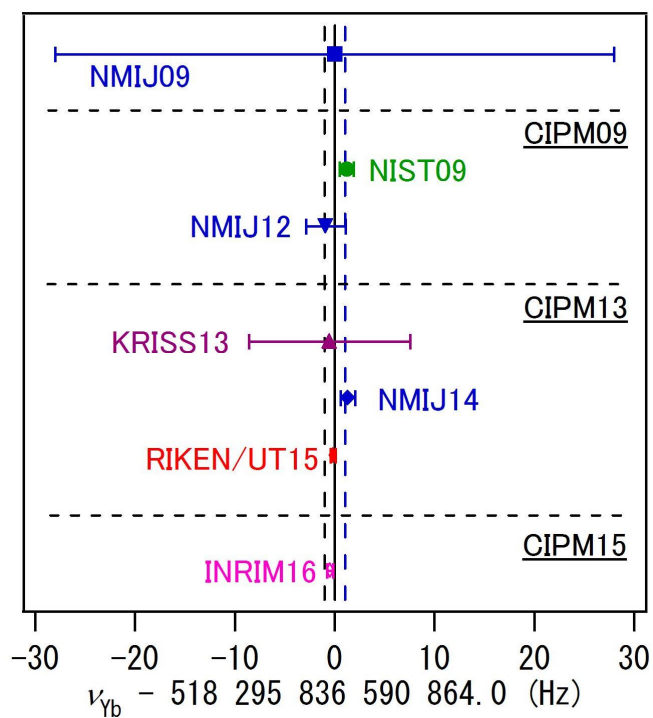


Fig. 6

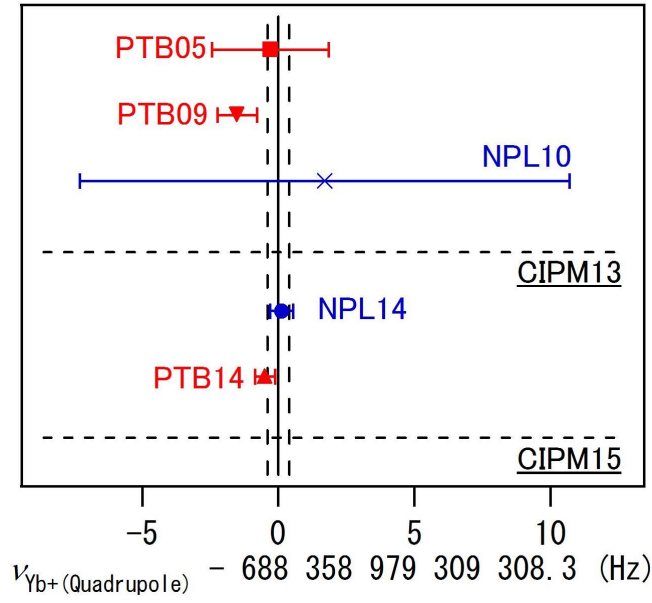


Fig. 7

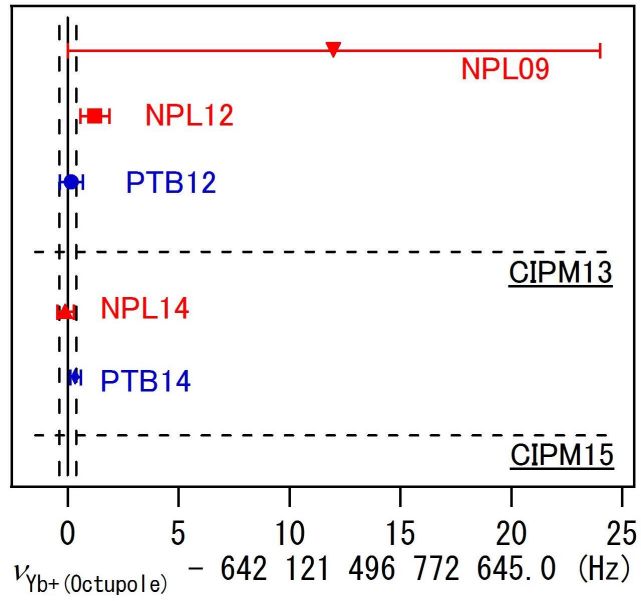


Fig. 8

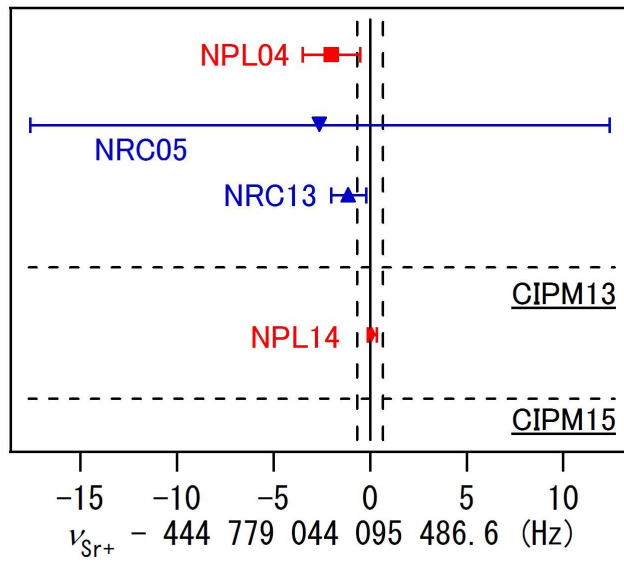


Fig. 9

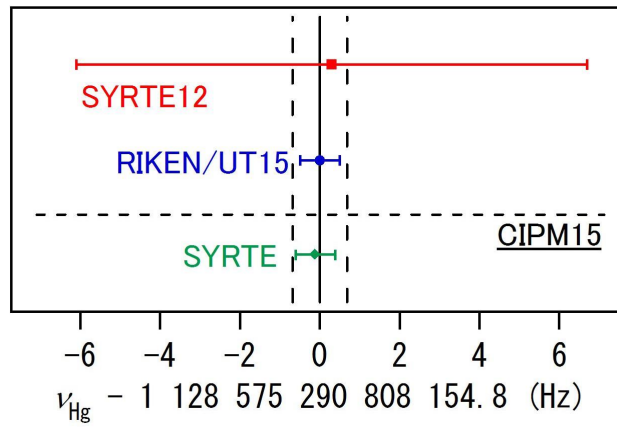


Fig. 10

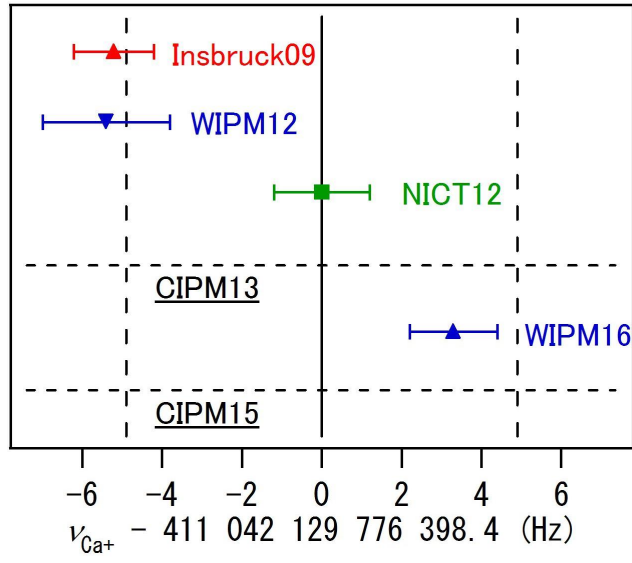


Fig. 11

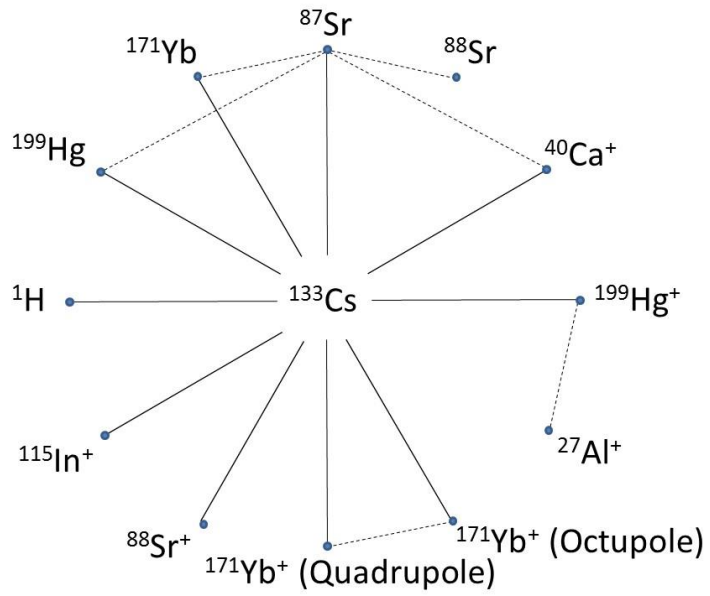


Fig. 12

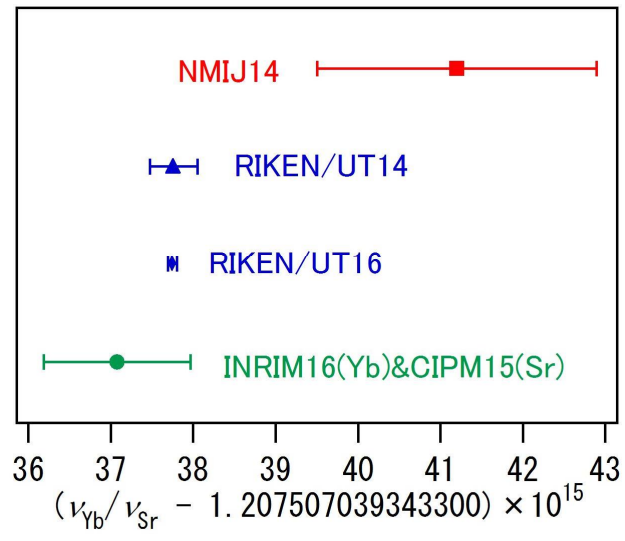


Fig. 13