

3 Optical Frequency Standards

3-1 Recent Activity of Optical Frequency Standards

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In order to obtain the definition of the second based on optical radiation, optical frequency standards are extensively studied in every national standards laboratories including NICT. In this article, three main components of optical frequency standards, namely narrow-line cw lasers, precision atomic spectroscopy, and frequency dividers to microwave regime, are briefly introduced with current status of the activities in foreign and domestic laboratories.

Keywords

Optical frequency standard, Optical cavity, Optical atomic clock, Optical frequency comb, Redefinition of the second

1 Frequency standards reaching a historic milestone

Among various physical standards, overwhelmingly high accuracy can be achieved for frequency standards. Therefore, their development is a challenge to the proposition: “to what extent can human measure nature in minute detail?” and can be an indicator of the development in science and technology. Thus, studies are being extensively carried out at national standards institutes of each country. Also, the fact that lots of breakthroughs related to frequency standards were previously awarded the Nobel Prize almost once a decade, symbolizes how broadly this progress has impacted the science and technology. The current definition of the second according to the International System (SI) of Units was determined by the energy difference between the hyperfine levels of cesium in 1967. Studies of frequency standards over following 40 years fall roughly into two categories: “studies for more accurate realization of the current definition by cesium” and “studies for frequency standards that are newer and better than the cesium”. The final goal of

the latter is the redefinition of the SI second, which needs to prove undoubtedly superb accuracy and stability compared with cesium, since it will have a huge impact on all science and technology. Thus, it has been recognized that it would be achieved not in the microwave region, but by an optical transition with higher frequencies.

Why then are optical transitions with higher frequencies more advantageous than microwave transitions? The reasons for this can be summarized as following two points: in principle,

- When atomic energy levels are perturbed to a certain amount, using high-frequency transitions have fractionally smaller influence.
- Measuring frequencies by using higher frequency standards enables accurate measurement in shorter time.

From a phenomenological perspective, it can be pointed out that there are many quantum transitions of atoms in the visible and ultraviolet light regions, and among them, there are many forbidden transitions preferred for fre-

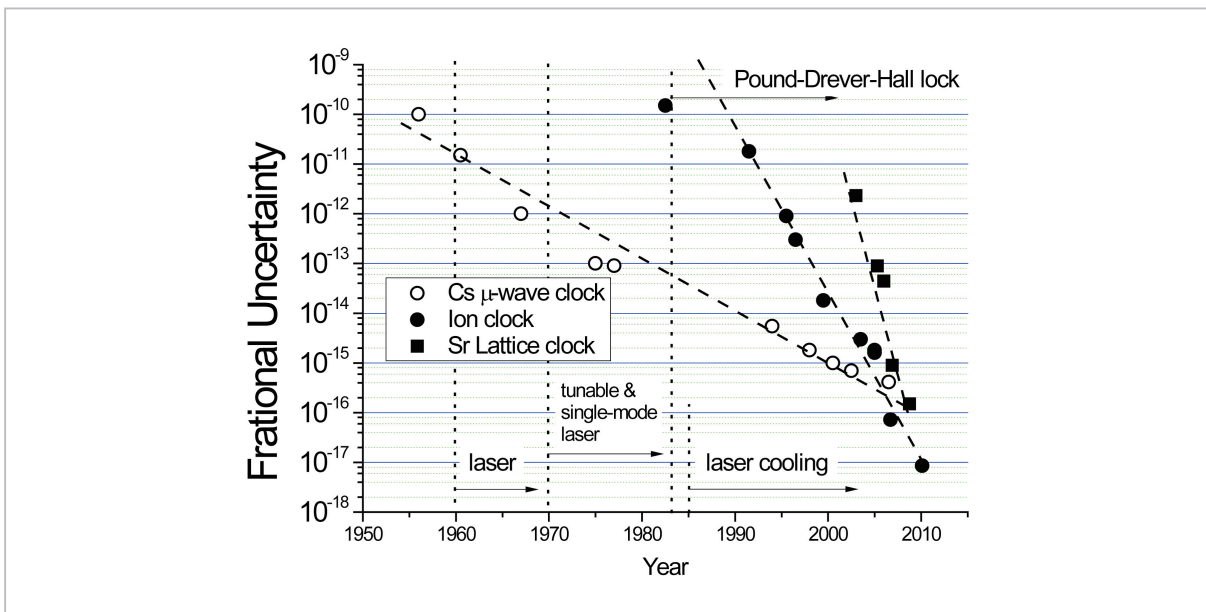


Fig.1 History and accuracy of atomic clocks development

quency standards due to their long coherence time.

Figure 1 summarizes the history of the last 30 years or so of the accuracy of clocks with cesium and with optical transitions and the related developed technologies. It shows that optical frequency standards finally surpassed microwave standards in the late 2000's. Consequently, the redefinition of the second began to be discussed in the community of time and frequency specialists such as the Bureau International des Poids et Mesures, where the list of atomic transitions that have performances similar to cesium atomic clocks is declared as "secondary representations of a second". In 2009, at the Consultative Committee for Time and Frequency (CCTF), possibility to redefine the second by an optical atomic transition was discussed. The committee currently makes reservation that the advancement should be saturated at certain point and technologies of international comparisons of optical frequency standards should be established. Note that Cs atomic clocks had been ahead of optical standards by improving the scheme of clock operation from magnetic state detection to laser excitation state detection and then fountain clocks. Cs clocks have utilized state-of-the-art techniques of optical physics such as tunable con-

tinuous-wave lasers and laser cooling. However, it can be understood that the advancement of the laser physics later than 2000 did not lead to the improvement of microwave clocks, and then finally the microwave clocks gave away the lead to optical frequency standards. The laser was invented a half century ago and the remarkable development of related optical technologies have replaced conventional methods with those using light in various fields such as of memory disks, information and communications technology, which is now becoming real also for frequency standards.

2 Structure of optical atomic clocks

Schematic diagram of optical atomic clocks is shown in Fig. 2. Optical atomic clocks are composed of following three parts:

- A) Narrow-linewidth tunable laser as a local oscillator
- B) Precision atomic spectroscopy
- C) Optical to microwave down-converter

The basic idea is that the frequency shift of the laser from the atomic resonance is first detected and then the result is fed back to compensate the difference. In addition, even if frequency standards can be obtained in the optical fre-

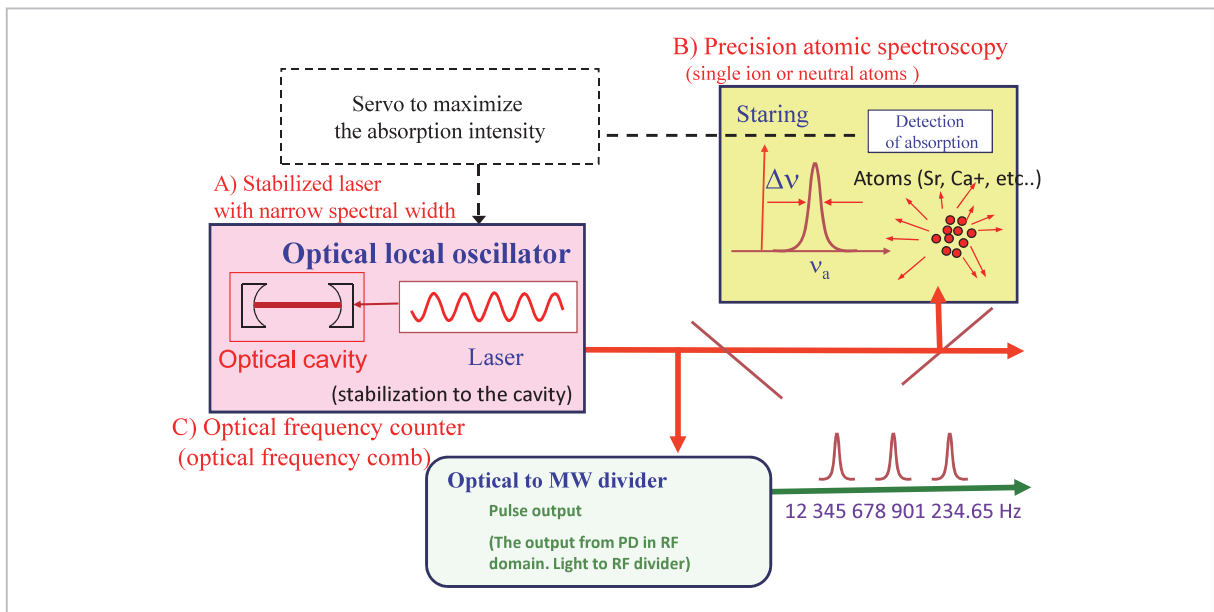


Fig.2 Three components of an optical atomic clock

quency domain, the current demand for high-accuracy frequency standards lies mainly in the microwave region. Furthermore, in order to make the existing SI traceable, it is necessary to divide the frequency of several hundreds of THz of a narrow-linewidth laser down to microwaves. Speaking of optical frequency standards in general, one tends to pay attention only to B) precision atomic spectroscopy and so does only to which transition of which atomic species is more advantageous to utilize. However, all of these three parts are indispensable. As a result, the performance of the entire optical frequency standards is restricted by what has the least performance among A), B), and C). Therefore, in other words, once a breakthrough has occurred in one with the lowest performance, either of the other two provides the limitation to the performance of whole optical frequency standards. Thus, the study of that field is accelerated, causing another breakthrough. Such a positive feedback has been procuring the development of the optical frequency standards.

This article takes look back over the previous developmental situations of each of these three fields, and summarizes bottlenecks for the ongoing development as well as research at NICT and international institutes that are trying

to overcome them.

3 Local oscillators

Local oscillators for optical frequency standards are the continuous-wave lasers. They are also called clock lasers and need to be wavelength-tunable in order to finally stabilize frequencies for clock transitions. Although tunable lasers were first realized by dye lasers that had been broadly used until the 90's, semiconductor lasers or fiber lasers with remarkably advanced manufacturing technologies are currently used from the viewpoint of maintenance. While the clock laser is irradiated to atoms and until we obtain the relative frequency of the clock laser against the atomic resonance, the performance of the whole clock is solely determined by clock lasers. Thus, narrow linewidth and small frequency drift are required for clock lasers. For these requirements, the technique that stabilizes laser frequencies to an external high-Q optical cavity made of low-loss dielectric mirrors was established (Pound-Drever-Hall technique, i.e. PDH technique) in the early 80's[1]. Furthermore, in the late 80's, by assembling this optical cavity from zero thermal expansion glass, the drift of the cavity-length was considerably reduced, and currently a sub-Hertz

linewidth and a drift rate less than 0.1Hz/s for the laser frequency have been realized[2]. Note that this PDH technique for stabilizing laser frequencies was applied not only to optical cavities, but also to the stabilization to the atomic resonance obtained from vapor cells. Thus, the technique played an indispensable role for realization of laser cooling (Fig. 1) as well.

The key for spectrum narrowing in clock lasers lies in not “how to stabilize a laser to an optical cavity,” but “how to keep the cavity length stable”. This is evidenced by the reality that residual error signals in the PDH method are currently determined by shot noise of the photodetection. On the other hand, the resultant stability is worse than the shot noise by a few digits. With respect to an optical cavity, due to technical progress in recent years of the low loss dielectric mirrors, a narrow resonance linewidth of several kHz is achieved even in the case of high Free Spectral Range (FSR) such as 1GHz. Furthermore, the progress of optical table and cavity-supporting strategy has allowed the cavity stabilized to the so-called thermal noise limited regime, where the instability is determined by the Brownian motion of the mirror materials. Formerly, as the substrate of a dielectric mirror, the zero thermal expansion glass such as ULE of Corning Incorporated was used so as to prevent differences in the thermal expansion rate from a spacer. Still, since the zero thermal expansion glass is an admixture of multiple glass materials, its mechanical Q-value is low. Therefore, the change of the cavity length due to thermal noise are larger than that using fused-silica substrates by a few digits and consequently restrict the stability in the 10cm cavity by 1×10^{-15} . Then, most recently, at SYRTE, creating cavity mirrors with fused-silica substrates, the stability on the level of 10^{-16} was realized. Furthermore, in order to suppress the thermal noise limit, there are two approaches: 1) to cool to low temperature, or 2) to extend cavity length to relatively diminish an impact of thermal noise on cavity length, and related studies are currently being planned and conducted at national stan-

dards laboratories worldwide. Regarding 1), since thermal noise is proportional to the square root of temperature, in order to expect an improvement of one order of magnitude over that in room temperature, it is necessary to cool down to liquid helium temperature. Thus, there is concern that the zero thermal expansion rate and temperature stability of the spacer at ultralow temperatures can be realized and the performance could be restricted by the vibration of a refrigerator. In regard to the problem of zero expansion rate, a plan to create cavity spacers and mirror substrates with silicon crystal to use at zero thermal expansion temperature (approx. 120K and 18K) was formed, and in this respect, JILA and PTB are promoting collaborative research though reports have not yet become available. Meanwhile, regarding a cavity with a long cavity length, there is the problem that a homogeneous and massive quantity of ULE, the most-used zero thermal expansion material at present, is difficult to obtain. However, NIST acquired it and uses a cavity that has an inter-mirror distance of approximately 25cm for an aluminum ion clock and a Yb optical lattice clock, both of which have obtained a stability of the lower half of $\sim 10^{-16}$. At NICT, using AZ glass that is a material similar to ULE, an optical cavity that has a quite new shape with a cavity length of 30cm was designed[4] and is presently in the process of evaluation. In general, the longer the cavity length becomes, the larger the vibration susceptibility becomes. Thus, vibration control is the most crucial point in the present design, and new shapes and supporting methods of the cavity to constrain vibration sensitivity have been proposed.

While the linewidth of clock lasers has been so far obtained in sub-Hz, the natural linewidth of clock transitions in atoms and ions that will be discussed below is on the level of mHz, and the short-term stability of optical frequency standards is limited by the linewidth of the clock laser, i.e. the stability of the optical cavity. Therefore, innovative narrow-linewidth light sources such as active lasers[5] that utilize the improvement of optical cavities or narrow-

linewidth transitions are currently required.

4 High-resolution atomic spectroscopy

The performance of an atomic clock is given in terms of its accuracy and stability. Accuracy is the uncertainty of the frequency standard generated by the clock. The resonant frequency of an atom is shifted due to perturbations and errors to set up various experimental parameters. There is a limit to predict or measure shifts, which causes uncertainty of frequencies; this is called accuracy. When the performance of an atomic clock is expressed by a number such as 5×10^{-15} , it generally refers to its accuracy. On the other hand, there is an index denoting fluctuations in frequencies, i.e. stability (Allan variance), which is expressed as a function of averaging time t . While readers should refer to the article by Kajita et al. in the present Special issue to know the detail[6], it would be sufficient to comprehend it as an index for how much difference in frequencies between two consecutive measurements arise when measuring average frequencies for time t repeatedly. Therefore, when discussing the stability of optical clocks, it is necessary to specify a stability for how many seconds on average; stabilities at approximately $t = 1$ second is generally called short-term stability, and values at roughly more than $t = 100$ seconds is called long-term stability. Most of actual oscillators show the characteristics of either:

- a) Less stability in the short-term but high stability in the long-term,

or:

- b) High stability in the short-term but bad stability in the long-term because of frequency drifts, etc.

In the microwave region, a) corresponds to Cs atomic clocks and b) to hydrogen masers or cryogenic sapphire oscillators.

The prerequisite for high-resolution spectroscopy in atomic spectroscopy is to tightly bind atoms in real space. When atoms exist in free space, they suffer recoil momentum by absorption and emission of photons to lead to

variation of kinetic energy. Therefore, when free atoms absorb photons, absorption occurs at off resonant frequencies by compensating the variation of kinetic energy. This is so-called the Doppler effect. Thus, it is crucial in the atomic spectroscopy for an optical frequency standard to fix atoms in a region that is duly smaller than the transition wavelength so that they don't suffer the photon recoil (Lamb-Dicke regime)[7]. As to confine in this Lamb-Dicke regime, ion clocks have been extensively studied since the 80's, and it was realized also in neutral atom systems by an optical lattice clock proposed by Prof. Katori, the University of Tokyo, in 2001. And, at NICT, a clock using Ca ions as a single-ion clock and a clock using Sr as an optical lattice clock have been developed. Readers should refer to the article by Matsubara et al. regarding Ca ion clocks[8], and to the article by Yamaguchi et al. regarding Sr optical lattice clocks[9].

4.1 Single-ion optical frequency standard

For single-ion optical frequency standards, a single ion localized in a region that is smaller than the transition wavelength due to laser cooling in ion traps is used as a frequency standard. Its prototype was proposed by H. Dehmelt in the 1980's[10]. So far, optical frequency measurements with six ionic species of $^{191}\text{Hg}^+$, $^{171}\text{Yb}^+$, $^{88}\text{Sr}^+$, $^{40}\text{Ca}^+$, $^{115}\text{In}^+$, and $^{27}\text{Al}^+$ have been reported, among which Hg^+ , Yb^+ , and Sr^+ are adopted as secondary representations of the second. An uncertainty of 8.6×10^{-18} with comparison between Al^+ ions has been reported, and thus the highest accuracy as a frequency standard is currently realized with the single-ion optical frequency standard. Since the single-ion optical frequency standard is based on one single ion standing still in a Paul trap, it excels optical lattice clocks in accuracy owing to simpler management and evaluation of frequency shifts but underperforms in stability due to its weak signal strength. Table 1 shows ionic species whose clock frequencies have been so far reported.

For $^{191}\text{Hg}^+$ and $^{171}\text{Yb}^+$ that are odd-mass iso-

Table 1 Major ionic species being studied as optical frequency standards

Ionic Species	Transitions	Wavelength [nm]	Natural Width [Hz]	Uncertainty	Major Institutions
$^{40}\text{Ca}^+$	$^2\text{S}_{1/2}\text{-}^2\text{D}_{5/2}$	729	0.14	2.4×10^{-15} [15]	NICT, University of Innsbruck
$^{88}\text{Sr}^+$	$^2\text{S}_{1/2}\text{-}^2\text{D}_{5/2}$	674	0.4	3.8×10^{-15} [13]	NPL, NRC
$^{171}\text{Yb}^+$	$^2\text{S}_{1/2}\text{-}^2\text{D}_{3/2}$	436	3.1	3.8×10^{-16} [11]	PTB, NPL
$^{171}\text{Yb}^+$	$^2\text{S}_{1/2}\text{-}^2\text{F}_{7/2}$	467	10^{-9}	2×10^{-14} [12]	NPL, PTB
$^{199}\text{Hg}^+$	$^2\text{S}_{1/2}\text{-}^2\text{D}_{5/2}$	282	1.8	1.9×10^{-17} [14]	NIST
$^{27}\text{Al}^+$	$^1\text{S}_0\text{-}^3\text{P}_0$	267	0.008	8.6×10^{-18} [19]	NIST, PTB
$^{115}\text{In}^+$	$^1\text{S}_0\text{-}^3\text{P}_0$	237	0.8	1.8×10^{-13} [17]	MPQ, NICT, PTB

Names of research institutions are as follows; NPL: National Physical Laboratory (UK), NRC: National Research Council of Canada (Canada), PTB: Physikalisch-Technische Bundesanstalt (Germany), NIST: National Institute of Standards and Technology (USA), MPQ: Max-Planck Institute of Quantum Optics (Germany)

topes among ionic species having the electron configuration of alkali metals, the total angular momentum (F) becomes integer. When setting the inter-level transitions with magnetic field components being zero ($m_F = 0$) among Zeeman sublevels as the clock transition, there are no frequency fluctuations to the variations of magnetic field due to the first-order Zeeman shift, and especially, perturbations by the alternating magnetic field from power-supply lines can be conveniently avoided. Research and development of $^{171}\text{Yb}^+$ has been conducted mainly at PTB and NPL that have reported an uncertainty of 3.8×10^{-16} using $^2\text{S}_{1/2}\text{-}^2\text{D}_{3/2}$ transition (436 nm)[11] and 2×10^{-14} with $^2\text{S}_{1/2}\text{-}^2\text{F}_{7/2}$ transition (467nm)[12]. In case of $^{199}\text{Hg}^+$ optical clock at NIST an ion trap cooled with liquid helium is employed in order to avoid collisional shifts by residual gas. This cryogenic system has the advantage that it enables reduction not only of collisional shifts but also of black-body radiation shifts due to the low temperature. An uncertainty of 1.9×10^{-17} has been reported by comparison with the Al^+ optical frequency standard that will be described later[14].

In contrast to ions like $^{199}\text{Hg}^+$ and $^{171}\text{Yb}^+$ with a small value of nuclear angular momentum $I = 1/2$, the Zeeman sublevel structure is excessively complicated in ions with large values of I , such as in $^{87}\text{Sr}^+$ ($I = 9/2$) and $^{43}\text{Ca}^+$ ($I = 7/2$). In these ions the excitation of transition between $m_F = 0$ levels is difficult. Transitions between $m_F \neq 0$ levels of even isotopes like

$^{88}\text{Sr}^+$ and $^{40}\text{Ca}^+$ are used instead due to their simple level structures. Since these transition are not free from the first-order Zeeman shift, the clock frequencies are evaluated as the average value of two transitions between $m_F = \pm 1/2$ and $m_F = \pm 1/2$ levels, which have the minimum positive and negative frequency shifts. Using this method, an uncertainty of 3.8×10^{-15} with $^{88}\text{Sr}^+$ has been reported by NPL[13], and 2.4×10^{-15} and 4.4×10^{-14} with $^{40}\text{Ca}^+$ by the University of Innsbruck[15] and by NICT[16] respectively. The accuracy improvement and details of $^{40}\text{Ca}^+$ optical frequency standard at NICT are described in the article by Matsubara et al. in this special issue[8].

In the above-mentioned ionic species having the electron configuration of alkali metals, the uncertainty is significantly restricted by electric quadrupole shift and black-body radiation shift. The electric quadrupole shift is usually corrected by averaging three pairs of transitions between magnetic sublevels of the Zeeman energy structure[14]. Although the black-body radiation shift could be estimated by the precise evaluation of the temperature distribution surrounding ions, the uncertainty is limited to the upper end of 10^{-16} level due to difficulty in the precise evaluation of the electrode temperature. Since the black-body radiation shift is proportional to the fourth power of temperature, cooling at extremely low temperatures such as in $^{199}\text{Hg}^+$ case is considered the most effective method to minimize the black-

body radiation shift.

Dehmelt's proposal assumes the use of ion species having electron configurations of alkaline earth metals such as B^+ , Al^+ , Ga^+ , In^+ , Tl^+ instead of those with alkali metal electron configurations described above. With these ionic species, the quadrupole shift does not exist for clock transitions and the black-body radiation shift is extremely small. However, since the transition used for cooling and detection (1S_0 – 1P_1) is located in the vacuum ultraviolet (VUV) region as is represented by the energy level structure of $^{115}In^+$ shown in Fig. 3, its generation is extremely difficult. Thus, the optical frequency standard faithful to the prototype proposed by Dehmelt has not been yet realized for over 30 years. Although its realization was attempted only with In^+ by using the 1S_0 – 3P_1 transition (230nm) as an alternative of the VUV transition for laser cooling and detection in 1990's, the reported uncertainty has remained at 1.8×10^{-13} [17]. Meanwhile, in the field of quantum information science that has been actively promoted based on the technique for single-ion optical frequency standards since the late 1990's, technical development has matured to result in the realization of small-scale quantum computers. Dehmelt's single-ion optical clock has been realized for the first time only recently as $^{27}Al^+$ optical frequency standard, which incorporates the Quantum Logic Spectroscopy (QLS) method based on the

small-scale quantum computer technique[14].

Al^+ optical frequency standard uses a single Al^+ ion localized in a linear trap, but not in the conventional Paul trap, as a frequency reference. The Al^+ is cooled sympathetically with one $^9Be^+$ ion trapped as "logic ion". The kinetic energy of Al^+ is coupled to the movement of Be^+ by coulomb force and is disposed of as the kinetic momentum of fluorescent photons by the laser cooling of Be^+ . In this manner, Al^+ is cooled to the ground state of quantized center of mass motion together with Be^+ . The detection of clock transitions is conducted by the fluorescence emitted from the $^2S_{1/2}$ – $^2P_{3/2}$ transition of Be^+ (313nm) after transferring the quantum-superposition state in Al^+ generated by the clock laser irradiation to the quantum state in Be^+ . In this process, the quantum information technology is used to exchange the internal quantum state of the ions and the quantized center-of-mass motion by means of laser pulse. In this respect the Al^+ frequency standard is an application of a quantum computer. After the first report of the QLS in 2005[18], an uncertainty of 2.4×10^{-17} was reported compared with Hg^+ in 2008[14]. Following this, an uncertainty of 8.6×10^{-18} , the most precise value of optical frequency measurements, has been reported in the recent experiment that uses Mg^+ as the logic ion[19]. The uncertainty is limited by the second Doppler shift due to residual vibrational motion, and thus it is supposed that uncertainty is further reduced by better cooling. Aiming at comparison with NIST, PTB has been attempting to develop a portable Al^+ optical frequency standard.

With the success of Al^+ , high accuracy realized with the ion species of alkaline earth electron configuration has again gained attention. However, the Al^+ optical frequency standard is still highly demanding compared to other frequency standards, since it requires high-end quantum information technologies such as sideband cooling, quantum gates, adaptive detection and the like. As an alternative, In^+ attracts much attention due to simpler access to high accuracy of an ion with the alkaline earth electron configuration. Three approaches to the

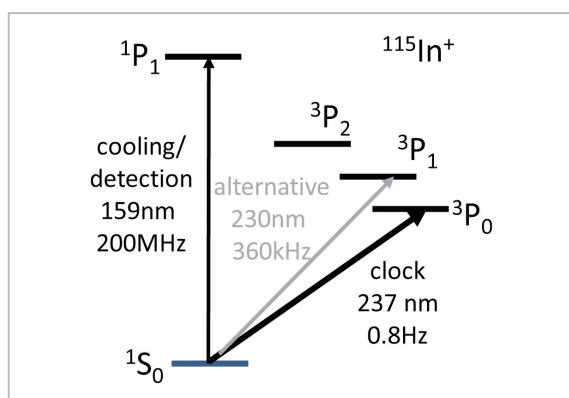


Fig.3 Energy levels of $^{115}In^+$

Functions, wavelengths and linewidths of transitions are shown

In⁺ optical frequency standards, namely QLS, excitation of the VUV transition and use of ¹S₀-³P₁ transition (230nm), might be implemented using an In⁺ cooled sympathetically with another ion. Relevant energy levels of In⁺ are shown in Fig. 3. Faster detection rate than that by QLS is expected by direct excitation of the ¹P₁ state with ultraviolet pulses prepared by high harmonic generation (HHG) under rapid development. Detection rate using the ¹S₀-³P₁ transition is quite small due to its small transition probability, which is roughly 1/100 of typical allowed transitions. This slow rate severely limits the stability of the optical clock, but the high accuracy of In⁺ might be fully exploited when the stability is compensated by use of a clock laser linked to the clock laser locked to a Sr optical lattice via optical frequency comb. This Sr-In⁺ *hybrid optical clock* owns the ultimate stability of the optical lattice clock and the ultimate accuracy of the single-ion optical clock. Uncertainty due to the blackbody radiation shift is a major limiting factor to the total uncertainty, but it is estimated in the lower end of 10⁻¹⁷ at 300K. Thus an inaccuracy in the order of 10⁻¹⁸ is reasonably expected with the In⁺ optical clock. Efforts to implement the three approaches are going on at NICT, while a miniature ion trap array for better signal noise ratio is under development at PTB.

4.2 Optical lattice clocks

The laser cooling of neutral atoms realized in mid 80's has enabled us to reduce the Doppler width of atomic spectroscopy less than the natural linewidth of the transition (~MHz). Following this success, the possibility of constructing frequency standards with neutral atoms was pointed out[20]. While spectroscopy utilizing optical Ramsay resonance for free-falling atoms was conducted according to this prescription in the 90's, it was figured out that it is impossible to realize 15 digits of accuracy like Cs atomic fountains due to the restriction by residual Doppler effects caused by the curvature and non-uniformities of the probe light wavefront. However, the situation changed dramatically with the method of optical lattice

clocks proposed by Prof. Katori in 2001 that realized the Lamb-Dicke regime in the neutral atom systems[21]. Regarding how to confine neutral atoms, one can think of almost nothing but optical dipole traps in order to realize confinement in the region smaller than the optical wavelength. However, this usually causes large AC Stark shift. With such a huge systematic error, it cannot deserve a frequency standard. However, Prof. Katori focused attention on the fact that suitable choice of the wavelength of optical dipole traps when setting the ¹S₀-³P₀ transition of two-electron atoms such as those of alkaline earth atoms as a clock transition enables to create the same trap shape for ¹S₀ and ³P₀ each, to realize the Lamb-Dicke regime similar to ion traps and to lengthen interaction time. Then, he proposed optical lattice clocks using neutral atoms in 2001[21], conducted recoil-free spectroscopy as a principle verification in the ⁸⁸Sr: ¹S₀-³P₁ transition in 2003[22], and succeeded in the ¹S₀-³P₀ transition being an optical lattice transition in 2005[23]. An optical lattice clock is realized by deftly utilizing singlet and triplet systems. Experiments are being promoted with the atomic species such as strontium (Sr), ytterbium (Yb) and mercury (Hg); clock operations have been realized for Sr at the University of Tokyo, JILA, SYRTE, NICT and PTB, and for Yb at AIST (Japan) and NIST (US). For mercury (Hg), development has been promoted at the University of Tokyo and SYRTE, but not yet reached the spectroscopy of atoms in the optical lattice due to the difficulty that most of the used transitions are in the ultra-violet region. However, since uncertainties with black-body radiation shifts are much lower than others, accuracy is expected to be better than Sr when recoil-free spectroscopy is realized. The Sr optical lattice clock was adopted as the secondary representation of the second by Comité International des Poids et Mesures (CIPM) in 2006, and has been studied at national standards laboratories in Japan, US, France, Germany, UK and China; it has currently the least uncertainty among secondary representations of the second including single-ion clocks and is accordingly

a candidate for the re-definition of the second. The performance accounts for an accuracy of 10^{-16} level, which is restricted by the uncertainties of the black-body radiation shift (1×10^{-16}), of the collisional shift (5×10^{-17}) and of the AC Stark shift (5×10^{-17}). The stability has been reported as 2×10^{-16} @300s[24]. The accuracy of the optical lattice clock with Yb is evaluated as 3.4×10^{-16} [25]. The Sr optical lattice clock at NICT has lately started its clock operations. Readers should refer to Yamaguchi's article in this special issue for details[9]. Also, this optical lattice clock has enabled us to directly compare with the optical lattice clock at the University of Tokyo via fiber link, and thus demonstrated the fiber transfer of optical frequency at a stability of 14 digits per second on average. Regarding fiber link, readers should refer to the article by Fujieda et al. of this special issue[26].

The reason why most national laboratories are currently developing optical lattice clocks lies in the distinguished short-term stability that optical lattice clocks potentially possess. In general, instability $\sigma(\tau)$ of an atomic clock is limited in principle by quantum noise (quantum projection noise) that arises when conducting destructive measurement of the excited state population after irradiating the clock laser, which is expressed by:

$$\sigma(\tau) = \frac{\Delta\nu}{\nu} \sqrt{\frac{T}{N\tau}} \quad (1)$$

Here, $\Delta\nu/\nu$ stands for quality factor of high-resolution spectroscopy, T for the time required for one measurement, and N for the number of atoms. Therefore, it turns out that on the condition of the same spectral shape of high-resolution spectroscopy, optical lattice clocks utilizing a large number of neutral atoms (10^4 – 10^5) are overwhelmingly more advantageous than ionic clocks from the view of stability. So far in the microwave region, Cs has been used when aiming at accuracy, and hydrogen masers or CSO (Cryogenic Sapphire Oscillator) have been used when good short-term stability is important. Thus, in the scene of research and development aiming at an accuracy of more than

13 digits, hydrogen masers with smaller phase noise are used as standard signals for frequency combs and signal generators, etc. And, calibration is applied to Cs clocks and GPS signals, etc. Thus, it is not an exaggeration to say that pursuing frequency standards in university laboratory is difficult by the lack of the low phase noise reference oscillators like hydrogen masers. Thinking about an analogy with the microwave region, even if a theoretical or technical limit for optical lattice clocks would come into sight about their accuracy, optical lattice clocks deserve a means to provide in the optical region alternatives to masers or CSO in the current microwave region with superior stability. Therefore, optical lattice clocks are a necessary technique in prompting optical frequency standards, and the above-mentioned Sr–In⁺ hybrid frequency standards are also based on the strategy of taking advantage of the superior stability of optical lattice clocks.

5 Optical frequency divider (frequency comb)

Since the invention of the laser, while the realization of frequency standards in the optical region with higher carrier frequencies by 4 digits than microwaves was a huge goal for researchers of laser physics, there was more than a little skepticism about optical frequency standards among specialists in time and frequency standards from the practical standpoint. This is due to the reality that even though optical frequency standards are successfully established to enable us to obtain optical frequencies with overwhelmingly high stability and accuracy, the predictable demand for frequency standards lies mainly in the microwave region, and therefore, the precise frequency division technique of 4 digits from light to microwaves should be established. In relation to this issue, until the mid 90's, frequency chains had been developed at national standards laboratories, but they confronted the extremely tough problem as to how to accurately link phase of the boundary domain between microwave and light, i.e. the terahertz region. Around that time, the group

led by Hänsch and others at Max-Planck-Institute focused attention on the fact that the pulse laser has the equally-spaced comb frequency spectrum with the same interval as the repetition frequency. Then, they simply extracted this repetition frequency by a photodetector to prove it possible to easily divide from the optical region to the microwave region, which solved this issue at once. While the interval of comb spectrum is determined by the repetition frequency, the offset frequency is determined by the carrier envelope phase or CEP. The precise control of this carrier envelope phase had been unrealistic until the mid 90's because there was huge intensity and phase noise in pulse lasers. However, the titanium-sapphire laser with a huge gain bandwidth was obtained with pump lasers as the SHG of Nd: YVO laser. This pump laser has an intensity noise dramatically smaller than the conventional Ar⁺ laser, and the photonic crystal fiber that expands the spectrum width by more than one octave was invented, which realized the optical frequency comb with highly accurate spectrum. Then, it turned out that offset frequency can be precisely obtained by integrating $f-2f$ self-reference interferometer into the spectrum of more than one octave, which enabled to perfectly control the spectrum of frequency comb. Although, at first, the frequency comb was only the titanium-sapphire laser, the stabilization of CEP has become possible also with Ti: CrO₄ laser, CrLiSAF laser, Er⁺ fiber laser, Yb⁺ fiber laser, etc., to enable us to obtain the performance duly available for frequency standards since the early 2000's. Motivations for these developments lie in: for solid-state lasers, adopting inexpensive laser diode as a pump laser avoiding expensive 532nm lasers; for fiber-based lasers, long continuous operations are possible with little impact from air disturbance, etc. and stable realization can be achieved even with intense mode locking. Now that these have been realized, fiber lasers are expected to be a mainstream for the development of frequency combs. At NICT, two frequency combs based

on Ti: sapphire lasers are steadily operated at present, and Er⁺ fiber lasers are being introduced to the scene of development of optical frequency standards with the recent stabilization of CEP. For details, readers are requested to refer to the article by Nagano et al. (Special issue of this NICT Journal, 3-4)

Another direction for optical frequency combs is the expansion of the wavelength domain. Presently-available frequency combs are basically in a wavelength of approximately 2 μ m to 300nm, and development for the realization of frequency combs in the terahertz domain and the vacuum ultraviolet region is also advancing.

6 Concluding remarks

While optical atomic clocks are expected to contribute also to the international atomic time in the event of their establishment, it is impractical to operate optical lattice clocks and single-ion optical frequency standards 24 hours a day. This situation holds true for the present Cs atomic fountain clocks; their continuous operation is extremely difficult and thus they are usually operated intermittently and used for calibration of the international atomic time. Therefore, the use of optical atomic clocks, as an alternative for the current Cs atomic clock, for the calibration of the international atomic time is supposed to be the first step for the contribution to provide time. From the viewpoint of continuous and steady supply of time, robustness and redundancy that are obtained by operating many commercial Cs atomic clocks (5071A, etc. of Symmetricom, Inc.) are supreme. Now that optical atomic clocks are confirmed to exceed conventional atomic clocks in accuracy, there remain challenging tasks: further raising their precision in comparison between multiple optical atomic clocks, and at the same time, realizing comparable robustness and redundancy with clocks in the optical region which overwhelm commercial microwave clocks in stability and accuracy.

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