

# Perspective for Using the Optical Frequency Standards in Realization of the Second

Karol Radecki

**Abstract**—The second is currently defined by the microwave transition in cesium atoms. Optical clocks offer the prospects of stabilities and reproducibilities that exceed those of cesium. This paper reviews the progress in frequency standards based on optical transitions, recommended by International Committee for Weights and Measures, as a secondary representation of the second. The operation of these standards is briefly described and factors affecting stability and accuracy of these and some new optical clocks are discussed.

**Keywords**—atomic clocks, atomic time scale, optical frequency standards.

## 1. Introduction

The best realization of the SI second today is served by cesium fountain primary frequency standards. The frequency accuracy of atomic time scale TAI realized by these standards is less than  $10^{-15}$  [1]–[3].

Commercial cesium clocks installed in time laboratories realize the second with accuracy and long term stability of  $10^{-14}$ . In laboratories, the active hydrogen masers are also installed, with short term instability better than  $10^{-15}$ . Commercial cesium and hydrogen masers standards contribute to the reliability and frequency stability of the atomic time scale, but they do not contribute to the realization of the second.

Over the past decade metrologists at various time and frequency standards laboratories have investigate the so-called forbidden optical transitions in cold trapped atoms and single ions. As clock transitions they have two major advantages: their frequencies are five orders of magnitude higher than the cesium frequency and natural linewidths are in the region of 1 Hz. This leads to high quality factors of these lines. However, the observed linewidths are larger, in the range up to few hundred Hz. Because the instability of optical clock is inversely proportional to the quality factor of the observed spectral line, it could be possible to achieve the short term stability of a few orders of magnitude better, assuming the number of atoms and transition interrogation time is the same.

Optical frequencies can be measured precisely by the femtosecond comb [4] and compared to cesium frequency with high accuracy.

Optical clocks offer the prospects of stabilities and reproducibilities that exceeds those of cesium. Today some optical clocks, based on  $^{88}\text{Sr}^+$ ,  $^{199}\text{Hg}^+$ ,  $^{171}\text{Yb}^+$ , may be used

as a secondary representation of the second [5], [6]. Recently, two optical clocks, based on  $^{27}\text{Al}^+$  ions and neutral  $^{87}\text{Sr}$  atoms, demonstrated systematic uncertainties which significantly exceed the current best evaluations of cesium primary standards. The progress in optical clocks is so rapid that in the near future the redefinition of the second will be most probably required.

## 2. Requirements for Optical Clock Transition

The main requirements for optical clock transition are a narrow natural line (linewidth less than 1 Hz) and the ability of their observation with the highest possible resolution. Transition frequency should also be unaffected by external electric and magnetic fields.

The clock transitions observed in number of laboratories worldwide are the weak, forbidden optical transitions in a single cold ion or cold atoms cloud.

In 2006 the International Committee for Weights and Measures (CIPM) recommended four optical transitions, which may be used as secondary representation of the second (Table 1) [5].

Table 1  
Recommended optical clock transitions (2006)

Atom/ion	Transition	Frequency of transition/ uncertainty
$^{87}\text{Sr}$	$5s^2\ ^1S_0 - 5s5p\ ^3P_0$	429 228 004 229 877 Hz/ $1.5 \cdot 10^{-14}$
$^{88}\text{Sr}^+$	$5s\ ^2S_{1/2} - 4d\ ^2D_{5/2}$	444 779 044 095 484 Hz/ $7 \cdot 10^{-15}$
$^{171}\text{Yb}^+$	$6s\ ^2S_{1/2}(F=0) -$ $5d\ ^2D_{3/2}(F=2)$	688 358 979 309 308 Hz/ $9 \cdot 10^{-15}$
$^{191}\text{Hg}^+$	$5d^{10}6s\ ^2S_{1/2}(F=0) -$ $5d^96s^2\ ^2D_{5/2}(F=2)$	1 064 721 609 899 145 Hz/ $3 \cdot 10^{-15}$

The CIPM has established the Working Group to review and discuss the uncertainty budget for possible optical candidates. It is required, that the selected frequency must have evaluated and documented uncertainty to the same level as it is required for primary standards contributing to international atomic time. In addition it is required, that

this uncertainty should be not worse than 10 times value that is for the best primary frequency standard.

Table 1 gives the values of recommended by CPIM 2006 unperturbed ground-state hyperfine the frequency transitions and estimated relative standard uncertainties. At present, due to progress in the optical clocks and the measurements systems, these parameters are evaluated more accurately.

The instability of the frequency standard that is operated in the interrogation cycles of duration  $T$  can be written as [7], [8]:

$$\sigma_y(\tau) \approx \frac{C}{SNR \cdot Q} \sqrt{\frac{T}{N\tau}}, \quad (1)$$

where:  $C$  is the constant that depends on the interrogation scheme,  $Q$  is the resonance quality factor  $Q = f_0/\Delta f$ ,  $\Delta f$  is the linewidth of resonance line centered at frequency  $f_0$ ,  $SNR$  is signal to noise ratio ( $SNR \approx 1$  if limited by quantum projection noise),  $T$  is the interrogation time (it should not be significantly larger than  $1/\Delta f$  because of the stability degradation),  $N$  is the total number of atoms/ions.

If we assume quantum limited operation of the  $^{199}\text{Hg}^+$  clock,  $\Delta f = 10$  Hz,  $N = 1$  ion and Rabi excitation pulse of  $T = 100$  ms, then the expected instability is  $\sigma_y(\tau) \approx 3 \cdot 10^{-15} \tau^{-1/2}$ . Similarly for the  $^{87}\text{Sr}$  optical lattice clock and  $N = 10^4$  atoms, the instability is about  $\sigma_y(\tau) \approx 7 \cdot 10^{-17} \tau^{-1/2}$ . For comparison the instability of  $^{133}\text{Cs}$  fountain clock with  $\Delta f = 1$  Hz,  $T = 1$  s and  $N = 10^6$  atoms is expected to be  $\sigma_y(\tau) \approx 5 \cdot 10^{-14} \tau^{-1/2}$ .

### 3. Look into Possible Optical Time and Frequency Standards

The main requirement for optical frequency standard is the need for highly stable laser which is disciplined by the clock transition in the trapped cold ion or neutral atoms. This is the so-called forbidden transition with natural linewidth of 1 Hz or less. The wideband femtosecond comb [4] is applied for precise comparison of optical frequency of the resonance line with cesium microwave frequency.

Ion or atomic trap works in cycles. The measurement cycle comprises laser cooling, state preparation, excitation of the clock transition and detection. Clock transitions are excited in a weak external magnetic field using Rabi or Ramsey pulse technique.

#### 3.1. Optical Clocks Based on Single Ions

Recommended by CIPM 2006 clock transitions are: the  $5s \ ^2S_{1/2} \leftrightarrow 4d \ ^2D_{5/2}$  in  $^{88}\text{Sr}^+$ , the  $5d^{10}6s \ ^2S_{1/2}(F=0, m_F=0) \leftrightarrow 5d^96s^2 \ ^2D_{5/2}(F=2, m_F=0)$  in  $^{199}\text{Hg}^+$  and the  $6s \ ^2S_{1/2}(F=0, m_F=0) \leftrightarrow 5d \ ^2D_{3/2}(F=2, m_F=0)$  in  $^{171}\text{Yb}^+$ . There are quadrupole transitions with natural linewidths of 0.4 Hz, 1.1 Hz and 3.1 Hz, respectively.

Ions are confined in RF traps and laser cooled to the so-called Lamb-Dicke limit. This greatly reduces the Doppler

broadening and frequency shift associated with ions motion relative to the excitation clock radiation.

Partial energy levels schemes of  $^{199}\text{Hg}^+$  and  $^{171}\text{Yb}^+$ , are very similar (Figs. 1 and 2). Clock transitions are excited in a small external magnetic field ( $\sim 1 \mu\text{T}$ ) between  $m_F = 0$  sublevels with no first order Zeeman shift.

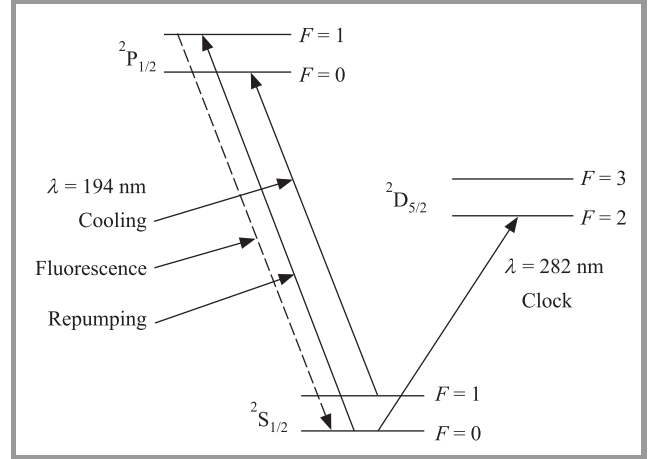


Fig. 1. Partial energy levels scheme of  $^{199}\text{Hg}^+$ .

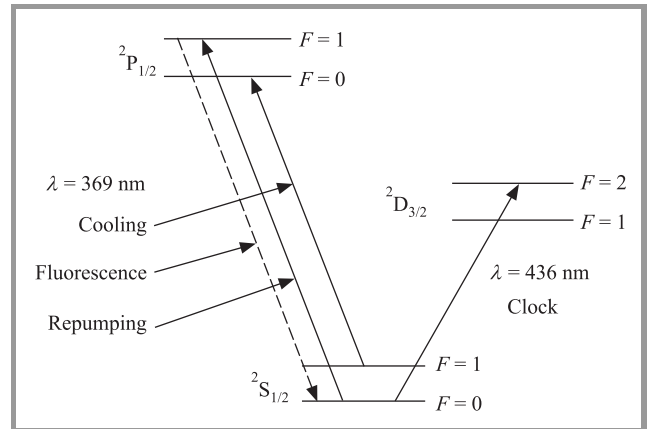


Fig. 2. Partial energy levels scheme of  $^{171}\text{Yb}^+$ .

A single trapped  $^{199}\text{Hg}^+$  ion is laser cooling using  $\lambda = 194$  nm. After cooling, ion is prepared in  $^2S_{1/2}(F=0, m_F=0)$ . To probe the transition, the  $\lambda = 282$  nm laser radiation is used. The clock transition is observed on  $\lambda = 194$  nm using quantum jumps technique. The line shape is measured from the statistics of many quantum jumps during discrete laser frequency sweeping across the clock transition. The measured linewidth of 6.5 Hz has been demonstrated with Rabi excitation pulse 120 ms long [9], [10].

Frequency instability of  $5 \cdot 10^{-15}$  at  $\tau = 1$  s was measured. In comparison with the  $^{27}\text{Al}^+$  standard, the systematic fractional uncertainty of  $\text{Hg}^+$  standard was estimated to be less than  $3 \cdot 10^{-17}$  [11]. The systematic uncertainty through comparison with Cs NIST-F1 frequency standard is estimated at  $7 \cdot 10^{-16}$  [12]. Optical cryogenic clock based

on  $^{199}\text{Hg}^+$  are now investigated at National Institute of Standard and Technology (NIST), USA.

In the  $^{171}\text{Yb}^+$  clock, the  $\lambda = 369$  nm laser radiation with repumper sideband is used for cooling ion (Fig. 2). After cooling, the ion is prepared in  $^2\text{S}_{1/2}(F = 0, m_F = 0)$ . To probe the transition, the  $\lambda = 436$  nm laser radiation is used. The clock transition is observed on  $\lambda = 369$  nm using quantum jumps technique.

The measured linewidth of 30 Hz has been demonstrated with Rabi excitation pulse 30 ms long. The systematic uncertainty through comparison with PTB Cs standards is  $1.5 \cdot 10^{-15}$  [13]. Optical clocks based on  $^{171}\text{Yb}^+$  are investigated at Physikalisch Technische Bundesanstalt, Germany (PTB) and British National Physics Laboratory (NPL).

The partial energy levels scheme for the  $^{88}\text{Sr}^+$  ion is shown in Fig. 3. The ion is laser cooled using radiations at both  $\lambda = 422$  nm and  $\lambda = 1092$  nm.

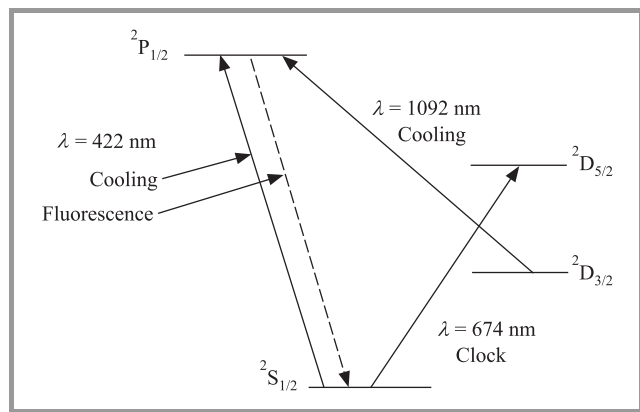


Fig. 3. Partial energy levels scheme of  $^{88}\text{Sr}^+$ .

In contrast to  $^{199}\text{Hg}^+$  and  $^{171}\text{Yb}^+$  ions the  $^{88}\text{Sr}^+$  ion has the linear Zeeman sensitivity to magnetic external field. This field split the clock transition into five pairs of Zeeman components, symmetrically located about the line centre. By probing one pair of components, cancellation of linear shift is achieved. The clock operates by stabilizing the interrogation laser to the mean transition frequency of the pair  $m_J = \pm 1/2, \Delta m_J = 0$  clock transitions.

The resonance linewidth is 9 Hz with Rabi excitation pulse 100 ms long [14]. The fractional systematic uncertainty through comparison with NPL primary Cs standard is estimated at  $3 \cdot 10^{-15}$  [15].

Optical standards with  $^{88}\text{Sr}^+$  are investigated at the NPL and National Research Council (NRC), Canada.

Recently, the optical clock transition  $^1\text{S}_0 - ^3\text{P}_0$  (with natural linewidth of 8 mHz) has been observed in  $^{27}\text{Al}^+$  ion, which cannot be directly laser cooled ( $\lambda = 167$  nm). The group at NIST [16] solved that problem by using sympathetic laser cooling of  $^{27}\text{Al}^+$  through the  $^9\text{Be}^+$  ion medium. Both ions are coupled together in the ion trap (by Coulomb interaction) and can be cooled using  $\lambda = 313$  nm radiation in  $^9\text{Be}^+$ . The  $^{27}\text{Al}^+$  ion is probed at  $\lambda = 267.4$  nm clock transition.

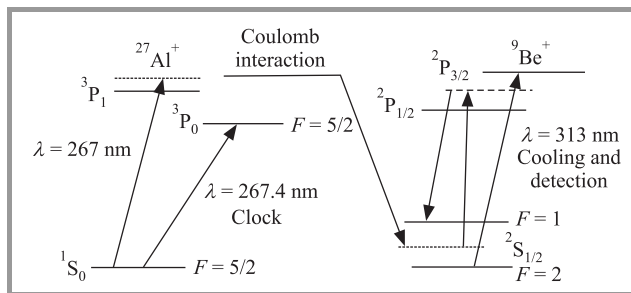


Fig. 4. Transfer of the  $^{27}\text{Al}^+$  clock state to detectable states in  $^9\text{Be}^+$  [16].

Clock transition information is sent to  $^9\text{Be}^+$  using quantum logic technique (Fig. 4). Pulse sequence maps the  $^{27}\text{Al}^+$  clock state  $^1\text{S}_0$  to detectable states ( $^2\text{S}_{1/2} F = 1$ ) in logic  $^9\text{Be}^+$  ion through the ions motional state, using  $^1\text{S}_0 - ^3\text{P}_1$  vibrational excitation  $\lambda = 267$  nm and Raman transition  $\lambda = 313$  nm. Fluorescence photons on  $\lambda = 313$  nm are counted if  $^{27}\text{Al}^+$  ion is in  $^3\text{P}_0$  state. The clock operates by stabilizing the interrogation laser to the mean transition frequency of the pair  $m_F = \pm 5/2, \Delta m_F = 0$  clock transitions. The systematic uncertainty through the comparison with  $^{199}\text{Hg}^+$  frequency standard is estimated at  $2.3 \cdot 10^{-17}$  [11].

### 3.2. Optical Clocks Based on Neutral Atoms

Recommended clock transition  $5s^2\ ^1\text{S}_0 - 5s5p\ ^3\text{P}_0$  ( $\lambda = 698$  nm) in neutral  $^{87}\text{Sr}$  atoms has the natural linewidth of 1 mHz. Partial energy levels scheme for  $^{87}\text{Sr}$  atom are shown in Fig. 5.

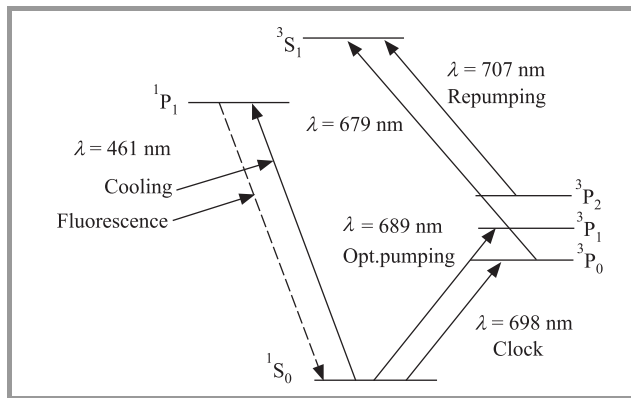


Fig. 5. Partial energy levels scheme of  $^{87}\text{Sr}$ .

Neutral atoms are trapped and cooled in magneto optical trap (MOT) operated on  $\lambda = 461$  nm transition. Two repumping lasers ( $\lambda = 707$  nm and  $\lambda = 679$  nm) are used to prevent atom loss into the  $^3\text{P}_2$  state. In a second stage of MOT, the atoms are cooled on  $\lambda = 689$  nm transition to a final temperature of  $2.5 \mu\text{K}$ . After cooling the atoms are loaded into optical lattice trap. Optical lattice greatly reduce the motional effects of atoms and allow for extension interrogation times of probing laser [17].

The clock operates at two transitions  $^1\text{S}_0(F = 9/2, m_F = \pm 9/2) \leftrightarrow ^3\text{P}_0(F = 9/2, m_F = \pm 9/2)$  excited on  $\lambda = 698$  nm and observed by measuring fluorescence on

$\lambda = 461$  nm. The clock centre frequency is found by taking the average frequency of both transition peaks. The resonance linewidth of 10 Hz with Rabi excitation pulse 80 ms long was observed [18].

The fractional systematic uncertainty of  $^{87}\text{Sr}$  clock through comparison with  $^{40}\text{Ca}$  NIST clock and NIST H-maser was evaluated at  $1.5 \cdot 10^{-16}$  [19], [20].

Optical standards with  $^{87}\text{Sr}$  are investigated at National Institute of Standard and Technology (NIST) USA, Laboratoire National de Métrologie et d'Essais (LNE-SYRTE), Physikalisch Technische Bundesanstalt Germany (PTB), National Metrology Institute of Japan (NMIJ) and University of Tokyo.

Optical clocks based on neutral  $^{40}\text{Ca}$ ,  $^{199}\text{Hg}$  and  $^{171}\text{Yb}$  atoms are also developed [21]–[24]. In contrast to  $^{87}\text{Sr}$  and  $^{171}\text{Yb}$  neutral mercury has low sensitivity to black body radiation and has the potential to achieve uncertainty at  $10^{-18}$  level [25].

### 3.3. Stability and Accuracy

Recently evaluated (2007/2008) systematic uncertainties and short term stabilities ( $\tau = 100$  s) for the optical clocks recommended by CIPM are summarized in Table 2. In the single ion frequency standards a significant uncertainty can arise from uncancelled electric quadrupole shift and quadratic Zeeman effect.

Table 2  
Systematic uncertainties and stabilities  
for various optical clocks

Optical clocks	$^{87}\text{Sr}/^{40}\text{Ca}$	$^{88}\text{Sr}^+ / ^{133}\text{Cs}$	$^{171}\text{Yb}^+ / ^{171}\text{Yb}$	$^{199}\text{Hg}^+ / ^{27}\text{Al}^+$	$^{27}\text{Al}^+ / ^{199}\text{Hg}^+$
$\sigma_y(\tau)$ 100 s	$6 \cdot 10^{-15}$	$3 \cdot 10^{-15}$	$10^{-15}$	$4 \cdot 10^{-16}$	$4 \cdot 10^{-16}$
$u_B$	$1.5 \cdot 10^{-16}$	$3 \cdot 10^{-15}$	$1.5 \cdot 10^{-15}$	$1.9 \cdot 10^{-17}$	$2.3 \cdot 10^{-17}$

The  $^{199}\text{Hg}^+$  and  $^{27}\text{Al}^+$  clock frequencies were measured relatively each other, and to the NIST-F1 cesium fountain [11], [16]. In both ion standards inaccuracies at  $2 - 3 \cdot 10^{-17}$  were evaluated. The dominant uncertainties in the  $^{199}\text{Hg}^+$  standard are due to the AC quadratic Zeeman effect and the magnetic field orientation, but in the  $^{27}\text{Al}^+$  the dominant components are due to the micromotion and secular 2nd order Doppler shifts. The black-body radiation shift for the  $^{199}\text{Hg}^+$  standard is negligible because the ion trap is operated at liquid helium temperature (4.2K). However, the black-body radiation shift for the  $^{27}\text{Al}^+$  standard is unusually small at the normal operating temperature ( $\sim 10^{-17}$  at 300K).

Similarly low level uncertainty at  $10^{-16}$  level was evaluated for the  $^{87}\text{Sr}$  clock compared with  $^{40}\text{Ca}$  optical clock [18], [20]. The dominant systematic uncertainty arose from lattice laser field, the room temperature black body radiation and interatomic collisions.

The  $^{88}\text{Sr}^+$  and  $^{171}\text{Yb}^+$  optical clocks have been evaluated in comparison with Cs primary atomic clock. Experiments which allow for the tests of frequency stability and evaluation of systematic frequency shifts by comparing two identical clocks are currently underway.

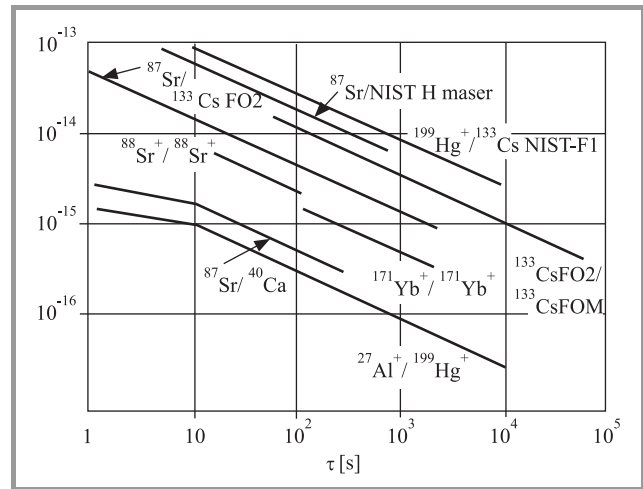


Fig. 6. Short term stability diagrams for optical clocks.

Figure 6 shows short term stability graphs for cesium fountains (FOM/FO2) and presently investigated optical clocks as a function of averaging time. The combined short term instability between FO2 and FOM (LNE-SYRTE) is  $8.4 \cdot 10^{-14} \tau^{-1/2}$ . Recently measured fractional frequency instability of the  $^{27}\text{Al}^+ / ^{199}\text{Hg}^+$  optical frequency comparison is  $\sigma_y(\tau) \approx 4 \cdot 10^{-15} \tau^{-1/2}$  for measurement duration  $\tau > 10$  s. Under assumption that both clocks contribute the same uncorrelated noise to the statistical measurement uncertainty, the short term stability of  $2.8 \cdot 10^{-15} \tau^{-1/2}$  for each clock is derived [11].

## 4. Summary

Narrow optical transitions observed in many atoms and ions are now promising candidates for next generation of high performance frequency standards. Recent advances in optical frequency measurements technique allow to achieve very high accuracy of remote optical clocks comparison over kilometer distances. Through this comparison, the uncertainty of optical clocks placed in different laboratories can be evaluated at the  $10^{-16}$  or at better level.

Optical clocks based on recommended by CIPM 2006 transitions are still in progress. To date optical standards based on  $^{199}\text{Hg}^+$  ion, neutral  $^{87}\text{Sr}$  atoms and new one based on  $^{27}\text{Al}^+$  ion, have demonstrated systematic uncertainties which significantly exceed (10 times) the current best evaluations of cesium primary standards.

Presently it is not clear what kind of clocks will be the best: single trapped ion or neutral atoms lattice clock [26]. Lattice clocks combine the advantages of trapped single ions and the large number of neutral atoms: long storage times and the good signal-to-noise ratio. These clocks require

precise and long term compensation of the large frequency shift associated with the lattice laser field. Promising candidate for reaching the ultimate performance of lattice clock is neutral mercury because of a low sensitivity to blackbody radiation (20 times smaller than Sr).

It seems that the optical clocks with instabilities and inaccuracies at  $10^{-18}$  level are expected in the time and frequency laboratories over the next several years. The progress in optical clocks is so rapid that in the near future the redefinition of the second will be most probably required.

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