Progress in optical frequency standards: ultracold Thulium, ions, and passive resonators

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Abstract. We report on different types of optical clocks and passive frequency references which are under development in our laboratories: optical lattice clock based on the inner-shell transition in the Tm atom at $\lambda = 1.14 \mu m$, optical ion clock on single $^{27}$Al$^+$ ion, and a family of lasers referenced to ultra-stable ULE and cryogenic silicon cavities.

1. Introduction

Frequency standards play an important role in a broad area of physics and many technical applications. For example, extremely stable and accurate frequency references are required for precision spectroscopy, tests of fundamental theories [1], measurements of fundamental constants [2] and provide a new approach to gravity sensing [3]. Progress in telecommunication [4], precision global positioning systems [5] and some other technologies are also strongly depend on performance of frequency standards. In the course of history fractional accuracy and stability of clocks were improving with increase in their operation frequency and the last revolution in the field happened with passage from microwave to the optical frequency domain in the beginning of the millenium. Active development of optical frequency standards began with advent of femtosecond frequency combs [6] which made it possible to transfer accuracy and stability from optical to radiofrequency domain. Also important role played advances in development of laser sources with sub-Hz spectral linewidths [7].

Narrowing of the "clock" laser spectral linewidth improves precision of the frequency standards and reduces integration time. For achieving sub-Hz performance, laser frequency is usually stabilized to an external high-finesse cavity. To minimize temperature influence, the body of the resonator is usually produced from materials with low temperature extension coefficients. In previous works we have reported on a family of laser systems based on diode lasers stabilized to ULE cavities [8] which show the fractional frequency instability of $1.5 \times 10^{-15}$ in 1-100 s time interval approaching the thermal noise limit. Lasers are currently used as clock lasers for Sr, Tm and Al$^+$ optical clocks at P.N. Lebedev Institute and VNIIFTIRI. To reach better performance we are currently working at cryogenic Si cavities [9] with GaAs/InGaAs mirrors for $\lambda = 1.5 \mu m$. The thermal noise will be lowered to $10^{-16}$ level which opens new perspectives both...
for applications in current optical frequency standards, and for relatively compact and simple ultrastable passive frequency references.

There are two main approaches to the development of an optical frequency standard: to use a narrow transition in a cloud of neutral atoms or in a single ion as a reference. Both ways have their own advantages and disadvantages.

In the first case a large number of interrogated atoms results in a high signal-to-noise ratio for the readout signal. Another advantage of these optical clocks is that the majority of atoms used for this purpose (Ca, Sr, Mg, H) [10] have "clock" and cooling transitions in visible region where they can be accessed with diode lasers. Drawbacks are relatively low lifetime of atoms in the cloud and strong trapping fields which cause shifts in energy structure of atoms and decreases performance of the clocks. The latter problem can be solved by trapping atoms in an optical lattice formed by laser fields at so-called "magic" wavelength [10]. Such lattice shifts energy levels of upper and lower clock levels equally leaving the transition frequency unperturbed. The best characteristics of accuracy and stability for neutral atoms clocks were achieved for Sr lattice frequency standards [11]. Their inaccuracy is on the level of $2.1 \times 10^{-18}$ and instability reaches $2 \times 10^{-18}$ at a few hours of averaging time.

The second possibility is to use a single ion in a radio-frequency trap. Single ions captured in Paul traps can be stored there for several hours or even months [12], subjected to weak trapping fields and do not interact with another particles. This enables one to build optical clocks with extremely high accuracy [13]. However signal-to-noise ratio for such clocks is inferior to lattice clocks which reduces stability and causes long integration times to reach the ultimate accuracy. Another disadvantage of single-ion optical clocks is that ions usually have cooling transitions in ultraviolet spectral domain and often can not be accessed by existing laser sources. In this case implementation of usual methods of direct laser cooling and electron shelving [14] for state readout becomes difficult or even impossible.

The method allowing to overcome these obstacles was demonstrated at NIST. Instead of direct laser cooling it was suggested to use sympathetic cooling [15] using a different auxiliary ion which has a convenient cooling transition. For state readout it was proposed to use quantum logic spectroscopy method [16] which enables one to map state of “clock” transition in the clock ion onto another metastable transition in auxiliary ion exploiting Coulomb interaction between particles. The state of auxiliary ion can be read with conventional electron-shelving method. Another advantage of quantum logic method is that after readout process reference ion remains particles. The state readout becomes difficult or even impossible.

To implement a lattice optical clock we work on an optical frequency standard based on the inner-shell transition [Xe]$4f^{13}6s^2$ ($J = 7/2, F = 4, m = 0$) $\rightarrow$ [Xe]$4f^{13}6s^2$ ($J = 5/2, F = 3, m = 2$) in the Tm atom at $\lambda = 1.14 \mu m$. It is considered as a candidate for an optical lattice clock. The transition wavelengths and probabilities for two clock levels $J = 7/2$ and $J = 5/2$ in the spectral range 250 – 1200 nm are calculated using the COWAN package which allows deducing of the differential dynamic polarizability and suggests that the magic wavelength is at around $6807$ nm with an attractive optical potential. The suggested clock transition demonstrates a low sensitivity to the BBR shift which provides a clock frequency instability at the low $10^{18}$ level competing with the best known optical clocks. We also evaluated other feasible contributions to clock performance (magnetic interactions, light shifts, van der Waals, and quadrupole shifts) which, after reasonable assumptions, can be lowered to the $10^{18}$ level. Together with the relative simplicity of laser cooling and trapping Tm atoms, our results demonstrate that Tm is a promising candidate for optical clock applications [21]. Experiments with direct excitation of the clock transition by spectrally narrow laser radiation at $\lambda = 1.14 \mu m$ set a lower limit for
the upper clock level lifetime of 112 ms which corresponds to the natural linewidth of < 1.4 Hz. Modulating the trap depth and analyzing the corresponding parametric resonances frequencies, we deduced the scalar polarizability of the Tm ground state at 532 nm which shows reasonable agreement with our calculations.

To apply the second approach with a single trapped ion we are developing a single-ion-based optical frequency standard on base of the $^1S_0 \rightarrow ^3P_0$ transition in Al$^+$ ion. This transition has a wavelength $\lambda = 267 \text{ nm}$ and the natural linewidth of $8 \text{ mHz}$ [22]. The transition also has the lowest BBR shift [23] among all ions and atoms used in optical clocks so far and has low sensitivity to external fields which makes it a promising candidate for a frequency standard. Since the cooling transition at 167 nm cannot be accessed by existing laser sources we plan to implement methods of sympathetic cooling and quantum logic spectroscopy with $^{25}\text{Mg}^+$ ion.

In this paper we show our progress on trapping $^{24}\text{Mg}^+$ ions in a linear Paul trap and their Doppler laser cooling. This is a first step towards creation of an optical frequency standard based on $^{27}\text{Al}^+$ reference ion and $^{25}\text{Mg}^+$ as an auxiliary ion for sympathetic cooling and state readout.

2. Experimental setup

The main part of our setup is a linear quadrupole Paul trap [24] with four cylindrical tungsten electrodes. Diameter of those electrodes is $d_{el} = 2 \text{ mm}$, the distance from the electrode surface to the trap centre of $r_0 = 1.475 \text{ mm}$. Using cylindrical geometry instead of hyperbolically blade caused by better optical access. Radial confinement is realised by applying RF potential with frequency $\omega = 2\pi \times 18.25 \text{ MHz}$ and $V_{ac} = 600 \text{ V}$ peak to peak amplitude to the diagonal pair of cylindrical electrodes. Constant part of electrodes potential $U_{dc}$ is set to zero. Axial confinement is provided by applying a DC voltage of $1000 \text{ V}$ to four ring electrodes. Two inner rings have diameter of 12.5 mm two others are of 8 mm diameter. For $^{24}\text{Mg}^+$ axial frequency with described parameters is about $\omega_{ax} = 2\pi \times 80 \text{ kHz}$, $q$ Mathieu parameter is equal to 0.084, and radial secular frequency is $\omega_{rad} = 2\pi \times 1.54 \text{ MHz}$.

Ions are loaded by electron impact ionisation of neutral Mg atoms directly in the trap, while trapping potentials are on. Mg atoms are produced by a resistive heating of a stainless-steel tube oven. The electron beam is provided by electron gun EGA-1012. Electron energy is about 500 eV, and current is 5 $\mu$A. We use such high electron energy rates due to electrons with lower energies are deflected by RF trap field and do not achieve its center. The number of stored ions can be measured with channeltron, situated on the trap axis.

All parts are mounted inside the stainless vacuum chamber with $P = 2 \times 10^{-10} \text{ mBar}$, pumped by ion-getter pump NexTorr D100-5. The scheme of the vacuum part of the setup is pictured in a figure 2

For Doppler cooling of $^{24}\text{Mg}^+$ we use cycling transition $^2S_{1/2} \rightarrow ^2P_{3/2}$ with the natural linewidth of $\Gamma = 2\pi \times 41.4 \text{ MHz}$ and the saturation intensity of about $I_{sat} \approx 2.5 \text{ mW/mm}^2$. This cycling transition requires single laser system at approximately 280 nm with stability and accuracy on a level of about 10 MHz. To meet this requirements we built a laser system which is shown on a figure 2. The ECDL laser with tapered amplifier Toptica TA pro provides 800 mW power at 1120 nm, which is converted to about 15 mW optical power at 280 nm with two stages of home-made bow-tie SHG cavities with LBO and BBO crystals respectively. Cooling laser frequency is locked to a wavelength meter WSU-10, which ensures long term stability and accuracy on required level.

Ion luminescence is collected by high aperture lens assembly made of UV grade fused silica with anti-reflection coating. The lens covers about 3.6% of $4\pi$ solid angle. Photons are detected with Raptor Photonics Falcon Blue EMCCD camera with quantum efficiency of 28% at 280 nm. Also fluorescence is measured by Hamamatsu H-8259 photomultiplier (PMT).
**Figure 1.** Sketch of the linear Paul ion trap, atomic source and detecting system. The inset shows the electric circuit of electrode connections. A – linear quadrupole Paul trap, B – channeltron, C – electron gun, Mg – magnesium atomic oven.

**Figure 2.** Laser system for Doppler cooling of $^{24}\text{Mg}^+$ ions.

3. $^{24}\text{Mg}^+$ ion cooling
To cool all degrees of freedom of ions we use two perpendicular laser beams. One of them propagates along the trap axis and another one is orthogonal to it. Due to very high initial ion temperatures beams are red detuned by $3\Gamma$ and have an intensity $15I_{\text{sat}}$. Propagation directions of cooling beams and scheme of fluorescence detection system are shown in the figure 3.

In the beginning of an experimental cycle trapping fields, cooling beams, both atomic and electron guns are switched on and ions are being trapped and laser cooled. After approximately
Figure 3. Orientation of cooling beams in the trap and fluorescence detection scheme. One of the laser beams propagates along trap axis, another one is orthogonal to it. Optical axis of the lens is perpendicular to both cooling beams. EMMCCD – Falcon Blue camera, PMT – Hamamatsu H-8259 photomultiplier, BS – beamsplitter.

Figure 4. Picture of trapped cloud of $^{24}$Mg$^+$ ions. White lines show boundaries of cooling laser beams.

Figure 5. Dependence of ions cloud fluorescence signal on storage time. Lifetime of ions in the trap equals to $\tau = 120$ s.

10 seconds the trap is loaded and guns are turned off. Then we measure dependence of fluorescence signal on storage time using the camera reading. Image of a trapped cloud of ions is shown in figure 4 and dependence of fluorescence rate on storage time pictured in figure 5. Exponential approximation shows that lifetime of trapped ions is about 120 s, which is two order of magnitude higher than without cooling [25]. This time is limited by collisions with the rest gas.

Micromotion inducted by stray fields prevents ion crystallisation. To facilitate crystallisation we currently work on photo-ionisation system and compensation system for stray fields. It will allow to reduce influence of stray fields, caused by charging of electrodes with electron beam.
4. Outlook

Doppler cooling of $^{24}\text{Mg}^+$ ion is a significant step towards development of single $\text{Al}^+$ ion frequency standard. In this work we demonstrate detection of fluorescence signal from $\text{Mg}^+$ ions, and the lifetime of ions in the trap up to 120 s. Now we are working on reducing excess micromotion caused by stray fields from charging of trapping electrodes with electron beam. Also we are developing third laser system for Raman resolved sideband cooling of $^{25}\text{Mg}^+$ to achieve ground state cooling.

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