Recent progress on the $^{27}$Al$^+$ ion optical clock

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Abstract. An aluminium ion optical clock is under development at Huazhong University of Science and Technology. The $^{25}$Mg$^+$ ion is chosen as logic ion to sympathetically cool an Al$^+$ ion and to detect its states. The $^{25}$Mg$^+$ ion is cooled to the motional ground state through Raman sideband cooling as the first step for quantum logic spectroscopy. Ultra-stable lasers for the interrogation of the clock transition are developed. The instability of the laser beat frequency is $1.2 \times 10^{-15}$ at 1 s, which is close to the thermal noise limit of the reference cavity.

1. Introduction

The $^1S_0-^3P_0$ transition in Al$^+$ at 267.4 nm, which has a narrow natural linewidth of 8 mHz, has long been recognized as a good potential clock transition [1]. Its electric quadrupole shift is zero ($J=0$), therefore the transition frequency is not influenced by the gradient of the electric field in the ion trap. Furthermore, the blackbody radiation contribution to the fractional frequency uncertainty of the Al$^+$ clock is as low as $4 \times 10^{-19}$ [2] at room temperature (300 K), which is the smallest among atomic species currently under consideration for optical clocks.

Since the Doppler cooling for Al$^+$ ions of 167 nm cannot be experimentally realized currently, the method of quantum logic spectroscopy (QLS) [3] is proposed to cool the ion and detect the clock transition. We choose $^{25}$Mg$^+$ as logic ion. Cooling the logic ion to ground state [4] is the first step to use QLS. In this paper we will report our work on Raman sideband cooling.

On the other hand, to detect the narrow clock transition we need an ultra-stable clock laser. We lock the laser to a very stable Fabry-Perot (FP) cavity with the Pound-Drever-Hall (PDH) technique [5]. There are many perturbation sources that affect the length of the cavity. Thermal noise limit is one of the fundamental noise sources due to the Brownian motion of the spacer, mirror substrate, and mirror coating [6]. Temperature fluctuations also affect the cavity length, so a very low coefficient of thermal expansion (CTE) material and a very good temperature control are needed. We investigate the noise contributions due to temperature fluctuations, optical power fluctuations and residual amplitude modulation to identify the limiting factors. We achieve two sub-Hz linewidth ultra-stable lasers. The instability of the lasers’ beat frequency is $1.2 \times 10^{-15}$ at 1 s, which is approaching the thermal noise limit of the two reference cavities.

2. Ion trap system

We use a linear Paul trap [7] to trap aluminium and magnesium ions. The linear trap consists of four blade electrodes and two end-cap tip electrodes. All the electrodes are made of titanium to reduce the influence of magnetic field. A helical resonator with a Q value of 200 is used to amplify the RF voltage to several hundred Volts, and its resonance frequency is about 23.8 MHz. The Q-factor is
defined by the amount of power stored in the resonator divided by the energy losses per cycle. The fluorescence of ions is collected by an imaging system into a PMT. The fluorescence collection efficiency is about 0.2 %. Mg and Al atoms are photo-ionized by a 285 nm laser and a 395 nm laser, respectively.

Figure 1. Partial energy levels of $^{25}\text{Mg}^+$ ion and used relevant lasers.

In our setup, two 280 nm frequency-quadruple laser systems are used for Raman sideband cooling [8]. One laser provides the two Raman beams and repumping light with the help of a 9.2 GHz EOM. Doppler cooling light is produced by a second 280 nm laser. So that during the detection process the background scattering rate can be reduced to 1 kHz. The relevant laser beams used are shown in figure 1. All the lasers are independently controlled with AOMs through RF switches. These RF switches are controlled by TTL signals generated by an NI 7851R FPGA board.

To determine the transition frequency between $|F=2, m=2\rangle$, i.e. $|\uparrow\rangle$, and $|F=3, m=2\rangle$, i.e. $|\downarrow\rangle$, states which will be used to repump $|\uparrow\rangle$ state back to $|F=3, m=3\rangle$, i.e. $|\downarrow\rangle$, state during Raman sideband cooling, we scan the spectrum of the transition. We first pump the ion to the $|\uparrow\rangle$ state, and then apply a microwave pulse followed by Doppler cooling light to detect the states of the Mg ion. The resonance frequency is 1.777830 GHz at about $5\times10^{-4}$ T magnetic field, and the Rabi frequency is 20 kHz. This Rabi frequency is still not high enough when performing quantum logic operation. Currently the antenna for the microwave is a homemade $\lambda/4$ antenna made by copper wire and mounted outside the vacuum chamber. We are considering two possible ways to increase the Rabi frequency. One is to shorten the distance between the antenna and the ion. Another is to impedance match the antenna with the microwave source.

The Raman spectrum is obtained by the following time sequence (shown in Fig. 2). First, we simultaneously switch on Doppler cooling and repumping light for 300 $\mu$s. Then we apply the Raman beams on the ion for 10 $\mu$s. During Raman sideband cooling, Raman interaction will repeat for 15 times. In between, repumping light will open for 2 $\mu$s followed by a 20 $\mu$s microwave pulse. In order to make sure the ion is completely repumped back to the ground state, this repumping process is repeated three times. The Raman sideband cooling is followed by scanning the Raman spectroscopy. Finally Doppler cooling light is applied for 50 $\mu$s to detect the states of $^{25}\text{Mg}^+$. The whole time sequence takes about 4 ms. To obtain the transition probability, we repeat the cycle 2000 times to compile statistics for each point on the sideband spectroscopy. In our current sequence, the microwave interaction is the most time-consuming process.

Theoretically, the temperature of the ion after Doppler cooling is expected to reach the Doppler limit. The actual secular motion frequencies are $\omega_x=2.2$ MHz, $\omega_y=2.4$ MHz, and $\omega_z=1.5$ MHz,
respectively. For an Mg\(^+\) ion at 1.5 MHz axial mode frequency, this temperature corresponds to an average vibration quantum number of 10. The vibration quantum number can be measured by measuring the ratio between the 1\(^{\text{st}}\) red sideband (RSB) and the 1\(^{\text{st}}\) blue sideband (BSB) of the Raman transition. Typically, in our system the average quantum number of an Mg ion after Doppler cooling is 16. This may be caused by the laser detuning during Doppler cooling. A half-linewidth red detuning gives the smallest temperature, but we use a near resonant laser to implement Doppler cooling. This is because we use the same laser for both Doppler cooling and detection. In order to obtain high enough fluorescence rates we have to use a small detuning. The Rabi frequencies of transitions corresponded to the 1\(^{\text{st}}\) and the 2\(^{\text{nd}}\) order RSB vary with vibration quantum numbers. At certain points the Rabi frequency will be near zero. Therefore the ion could not be cooled to the ground state using only the 1\(^{\text{st}}\) order RSB cooling or the 2\(^{\text{nd}}\) order RSB cooling. To avoid this problem, the 2\(^{\text{nd}}\) order RSB are applied 15 times followed by 15 times 1\(^{\text{st}}\) order RSB, and the process is repeated for two times. Figure 3 shows the sidebands under different Raman sideband cooling sequences. At last the population of \(n=0\) is about 93 \% in figure 3.

![Figure 2: The time sequence for Raman sideband cooling.](image)

![Figure 3: Measurement of sidebands under different Raman sideband cooling sequences. (a) the 1\(^{\text{st}}\) BSB and RSB without Raman sideband cooling, \(n\approx16\); (b) the 1\(^{\text{st}}\) BSB and RSB when cooled with 15 times 1\(^{\text{st}}\) RSB and 15 times 2\(^{\text{nd}}\) RSB, \(n=5.3\); (c) the 1\(^{\text{st}}\) BSB and RSB when cooled with twice 15 times 1\(^{\text{st}}\) RSB and 15 times 2\(^{\text{nd}}\) RSB, \(n=0.24\); (d) the 1\(^{\text{st}}\) BSB and RSB when the last 1\(^{\text{st}}\) RSB](image)
cooling times is changed from 15 times to 30 times, \( n = 0.08(1) \), and the population of \( n = 0 \) is about 93%.

After Raman sideband cooling, we try to sympathetically cool \( ^{27}\text{Al}^+ \). Here we use \( ^{26}\text{Mg}^+ \) to sympathetically cool \( ^{27}\text{Al}^+ \) for demonstration purpose. At first, a single \( ^{26}\text{Mg}^+ \) is loaded. Then the Al oven is fired and the 395 nm ionization laser is switched on for several minutes. One can easily know whether \( ^{27}\text{Al}^+ \) is loaded by looking at the ion image on EMCCD, since the \( ^{26}\text{Mg}^+ \) ion will shift a little bit on the image plane due to the trapped dark \( ^{27}\text{Al}^+ \) ion. To confirm that the dark ion is \( ^{27}\text{Al}^+ \), mass spectroscopy experiments have been carried out. We first measure the secular motion frequency of two \( ^{26}\text{Mg}^+ \) ions, and then measure the secular motion frequency of a \( ^{26}\text{Mg}^+/^{27}\text{Al}^+ \) pair. As shown in Fig. 4, the secular motion frequency of \( ^{26}\text{Mg}^+/^{26}\text{Mg}^+ \) and \( ^{26}\text{Mg}^+/^{27}\text{Al}^+ \) are 434.4(7) kHz and 430.2(4) kHz, respectively. The results agree with the theoretical calculation, 434 kHz and 430 kHz, respectively.

![Figure 4. Secular motion frequency of Mg-Mg ion pair and Mg-Al ion pair.](image)

3. Ultra-stable laser system

The \( ^{27}\text{Al}^+ \) optical clock requires an ultra-narrow linewidth laser as a clock laser. To develop the first generation clock laser, two 1070 nm diode lasers from Toptica are phase stabilized to two independent 10 cm long all ULE cavities through PDH locking technique. The stabilized diode laser is quadrupled to 267.4 nm as the clock laser.

The thermal noise limit of one reference cavity which works at TEM\(_{00}\) mode is estimated to be 7.7×10\(^{-16}\) @ 1s, dominated by the thermal noise of the mirror substrates material. The linewidth of TEM\(_{00}\) modes of these two cavities are measured to be 13.6 kHz and 4.5 kHz, respectively. As the cavity length variation directly broadens the locked laser linewidth, the cavities are sealed in two vacuum chambers with pressures less than 1×10\(^{-6}\) Pa. The vacuum chambers are temperature controlled at the measured ULE cavity’s zero crossing temperature \( T_0 \) of 36 °C and the temperature fluctuation is about 1 mK in few hours’ time. In addition, the vacuum chambers are placed on active vibration isolation (AVI) stages and the whole systems are enclosed with a heavy box made of lead and iron plate to reduce the effect of acoustic noise and air disturbance. The ULE cavities are placed on four viton bars on their notched surfaces where the mounting positions are optimized to minimize the sensitivity to vibration through finite-element analysis (FEA) with Comsol Multiphysics.

The EOMs used in the PDH locking scheme are temperature controlled to reduce the residual amplitude modulation (RAM), and modulation depths are about 1.08 to reach a maximum slope for the PDH error signal. The error signals are sent to locking circuits (FALC, Toptica) that feed back to the current and piezo control of the diode lasers. We obtain a 2 MHz current feedback bandwidth and a 2 kHz piezo feedback bandwidth. As shown in figure 5, a beat note with a linewidth of 0.88 Hz is
obtained between the two locked lasers, indicating an absolute linewidth of 0.6 Hz for one single laser. The Allan deviation of the beat note is $1.2 \times 10^{-15}$ at 1 s which is close to the thermal noise limit contributions of the two cavities.

Figure 5. The Allan deviation and spectral noise density of the beat note between lasers.

In order to develop better ultrastable clock lasers that can reach an instability of $10^{-16}$ or even $10^{-17}$, noise contributions due to temperature fluctuations, optical power fluctuations and residual amplitude modulation are analysed to identify the limiting factors.

For the noise contribution due to temperature fluctuations, the thermal time constant $\tau$ is measured to be about 1 day by changing the temperature of the vacuum chamber with a step of 1 K and use an optical frequency comb to monitor the frequency of the locked laser. The temperature fluctuation of the cavity can be written by the following formula:

$$T_{in}(\omega) = \frac{T_{out}(\omega)}{\sqrt{1+(\omega \tau)^2}}. \quad (1)$$

$T_{out}(\omega)$ is the vacuum chamber temperature in the frequency domain, and $T_{in}(\omega)$ represents the ULE cavity temperature. The slope of the CTE $\beta$ near the zero crossing temperature is measured to be $-1.2 \times 10^{-9}/K^2$ by measuring the relationship between the cavity temperature and the locked laser frequency. The influence of temperature variation of the cavity on the Allan deviation $\sigma_{in}(\tau)$ can be estimated by Eq. 2.

$$\sigma_y(\tau) = \beta(T - T_0)\sigma_{in}(\tau). \quad (2)$$

For conservative estimation, $T - T_0$ is chosen to be 1 K, which is larger than the real temperature difference between the zero crossing temperature $T_0$ and the controlled temperature. As a result, $\sigma_y(\tau)$ is lower than the thermal noise limit of the 10 cm cavity at 1 s to 1000 s. In order to reduce the noise contributed from temperature fluctuations, larger thermal time constant of the vacuum chamber and better temperature control at $T_0$ will help.

The power circulating in the cavity is very strong while the laser is resonant with the cavity. The total power in the cavity can be estimated to be

$$P_{cavity} = P_{in} F / \pi, \quad (3)$$

where $F$ is the cavity finesse, $P_{in}$ is the laser power coupled into the cavity. In our case the measured finesse is about 300,000 for one of the cavities and $P_{in}$ is about 10 $\mu$W, therefor the power circulating in the cavity $P_{cavity}$ is near 1 W. The laser beam will induce a local heating on the mirror coatings and substrates that causes the cavity length change, and will result in the fluctuation of the locked laser frequency. In order to evaluate the influence of power fluctuations, we first measure the frequency sensitivity to power $k$ by modulating the coupling power and measure the laser frequency change. When the the coupling power changes by 5 $\mu$W, the laser frequency changes 35.5 Hz, which corresponds to a sensitivity of 7.1 Hz/$\mu$W. After that we measure the laser power in front of the cavity when the laser is locked to the cavity and get the influence of the power fluctuation $\sigma_{P_{in}}(\tau)$ on the Allan deviation

$$\sigma_y(\tau) = k P_{in} \sigma_{P_{in}}(\tau) / \nu. \quad (4)$$
Here, $\nu = 280.7$ THz is the laser frequency. The Allan deviation contributed by the laser power fluctuation is lower than $10^{-15}$ during 1 s to 100 s. By active stabilization, it can be improved to the $10^{-17}$ level.

RAM is one of the dominant noises in ultrastable laser systems based on the PDH technique. In the ideal case, the error signal should be zero when the laser is on resonance with the cavity. But RAM introduces an offset voltage in the error signal which biases the locking point from the cavity transmission peak and fluctuates with time. The RAM can be caused by many effects, such as temperature fluctuation of the EOM, misalignment of polarization between the input beam and the principal axis of the EOM crystal, and etalon effect. When the laser is locked to the reference cavity, the RAM induced fractional frequency instability can be represented as [9]:

$$\sigma_y(\tau) = \sigma_{\text{RAM}}(\tau)/\nu,$$ (5)

where $\Gamma$ is the cavity linewidth. We implement an EOM temperature stabilization to reduce the RAM. We also carefully align the polarization of the injection beam to the axis of the crystal and suppress the etalon effect. It is shown that the Allan deviation caused by RAM is smaller than $10^{-15}$ from 1 s to 1000 s. It can be further reduced by active feedback control to reach the $10^{-17}$ level.

### 4. Conclusion and Outlook

We demonstrate Raman sideband cooling the $^{25}\text{Mg}^+$ ion to the motional ground state and sub-Hz linewidth ultra-stable lasers for Al’ optical frequency standard. The achieved stability of the lasers is approaching the thermal noise limits of the cavities. We also investigate the noise contributions due to temperature fluctuations, optical power fluctuations and RAM to identify the limiting factors.

For our second generation clock laser, the diode lasers will be locked to 30 cm long ULE cavities with fused silica mirrors to reach an instability of $1 \times 10^{-16}$ at 1 s. One of the cavities is a commercial product with a sandwich structure, with an additional ULE ring optically contacted to the back surface of the fused silica mirrors [10]. The second cavity is a homemade one with a re-entrant structure which has a wide tuning range of the zero crossing temperature of CTE to room temperature [11].

For the third generation clock laser, the laser will be locked to a 6 cm long cryogenic cavity with sapphire mirrors to reach an expected instability of $2 \times 10^{-17}$ at 1 s. The designed square shape cavity will be maintained at 4 K where the CTE of cavity ($4 \times 10^{-11}/\text{K}$) and the slope of CTE ($3 \times 10^{-11}/\text{K}^2$) are both very small. A pulse tube cryostat for housing the cavity is currently under development.

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