Progress toward a single indium ion optical frequency standard

T. Liu, Y. H. Wang, V. Elman, A. Stejskal, Y. N. Zhao, J. Zhang, Z. H. Lu, and L. J. Wang
Institute of Optics, Information and Photonics, Max-Planck Research Group and University Erlangen-Nuremberg
91058 Erlangen, Germany
Email: lwan@optik.uni-erlangen.de

R. Dumke
School of Physical and Mathematical Sciences, Nanyang Technological University, Nanyang Walk Block 5, Level 3, Singapore 637616, Singapore

Th. Becker and H. Walther
Max Planck Institute of Quantum Optics, Hans-Kopfermann-Str. 1, 85748 Garching, Germany

***Abstract*—We report an improved absolute frequency measurement of the $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ narrowline clock transition at 236.5 nm, for a single trapped and laser cooled $^{115}In^+$. Using a narrowline laser as the local oscillator, a linewidth of 43 Hz for the transition is resolved. For absolute frequency measurement we use an optical frequency comb referenced to a cesium clock. The transition frequency is found to be 1, 267, 402, 452, 900, 967 (63) Hz, averaged over 13 days of separate measurements. The accuracy is about 5.0 × 10^{-14}. And a preliminary absolute frequency measurement is shown with a Hydrogen maser as the reference. We discuss possibilities for further improvement.**

I. Introduction

An optical frequency standard promises higher stability and accuracy than the present-day microwave standards. It offers a unique opportunity to test fundamental physics, improve frequency metrology, and advance technological applications. Through intensive investigation of different systems for atomic optical clocks [1-16], a stable and accurate optical frequency standard is well within the reach.

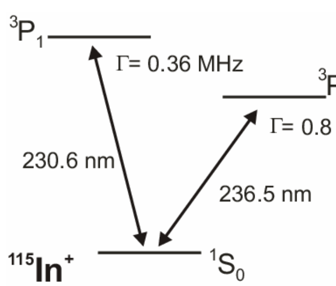

Figure 1. Partial energy levels of $^{115}In^+$. The clock transition $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ has a frequency of 1267 THz (236.5 nm) and a natural linewidth of only 0.8 Hz. The cooling transition $5s^2\ ^1S_0$ - $5s5p\ ^3P_1$ at 230.6 nm has a linewidth of 360 kHz.

A single indium ion ($^{115}In^+$) (Fig. 1), trapped and cooled to the lowest vibrational states well within the Lamb-Dicke regime, can serve as such a frequency reference [17]. The reference is a $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ narrowline clock transition with a natural linewidth of 0.8 Hz, leading to a quality factor of $\nu/\Delta\nu = 1.6 \times 10^{15}$ [15]. The clock transition is between states with vanishing total electronic angular momentum J=0, which couples only weakly to external electric and magnetic fields. In particular, the transition is insensitive to the electric quadrupole shift caused by the field gradient of the trap. These make the $^{115}In^+$ a favorable candidate for an optical frequency standard, with low sensitivity to environmental disturbances [17]. With a frequency stabilized laser locked to the clock transition, a stability of $\sigma_y(1s) = 1 \times 10^{-15}$ and an accuracy of lower than 10^{-17} are possible.

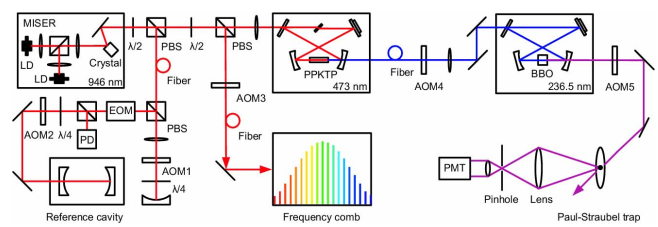

Figure 2. The experimental setup. MISER: monolithic isolated end-pumped ring Nd:YAG laser;LD, laser diode; PBS, polarized beam splitter; AOM, acousto-optical modulator; EOM, electro-optical modulator; PPKTP, periodically poled KTP crystal; PD, photo detector; PMT, photomultiplier tube.

The absolute frequency of the $^{115}In^+$ $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ clock transition has been measured twice before [18,19]. In Ref. [19], the transition frequency was determined to be 1,267,402,452,899.920(230) kHz, where the reference was a methane-stabilized He-Ne laser at 3.39 µm. The accuracy was limited by the uncertainty of the He-Ne laser itself. Since then, the short-term frequency stability of the clock laser was further improved [20, 21]. The linewidth of the clock transition was measured to be 170 Hz, corresponding to a fractional resolution of 1.3×10^{-13} [15].

Here, we report a new absolute frequency measurement of the clock transition using an optical frequency comb referenced to a microwave cesium clock (Agilent 5071A). The absolute frequency is determined to be

$$f_{In^+} = 1,267,402,452,900,967(63)\ Hz$$

Using an ultra-stable clock laser, we observed a transition linewidth of 43 Hz on the clock transition. This is approximately four times narrower compared to the earlier best result [15].

II. Experiments and Results

The experimental setup is shown in Fig. 2, and the apparatus consists of three main parts: the narrow linewidth IR light source with two frequency doubling systems for the generation of UV light (236.5 nm); the Paul-Straubel trap with a laser cooled single $^{115}In^+$ ion; a frequency comb system referenced to the cesium clock for absolute frequency measurement.

A. Ion trapping and cooling

For our experiment, a single $^{115}In^+$ ion is confined in a Paul-Straubel trap. This trap setup has been described in detail before [15, 22], and we only give some brief descriptions here. The Paul-Straubel trap is a 1 mm ring made of copper beryllium alloy. A 10 MHz RF driving field with a voltage of 1 kV is applied to the ring electrode. The three secular frequencies of the trap are 1.52, 0.87, and 0.93 MHz respectively. The ions are produced by a two-stage photoionization process using a 410 nm laser [23]. We start initially with a cloud of ions trapped in the electro-dynamic potential. Fig. 3(a) shows a spectrum of the ion cloud. The asymmetric Doppler profile in the spectrum is caused by the asymmetric cooling and heating effect of the cooling laser when it crosses the atomic transition. After heating out the ions until only two ions remain in the trap, we can observe the onset of formation of a two-ion crystal. This is marked by an abrupt decrease of the Doppler spectrum width when the cooling rate becomes high enough such that the crystal is formed (Fig. 3(b)). On resonance the photon count rate is about 2 kHz. Finally, Fig. 3(c) shows the spectrum of a single ion. The fluorescence count rate of 1 kHz is almost exactly half of that of a two-ion crystal. The fluorescence signal from the ions is collected using a high numerical aperture imaging system into a photomultiplier tube. The combined efficiency of the entire detection system is about 0.1 %.

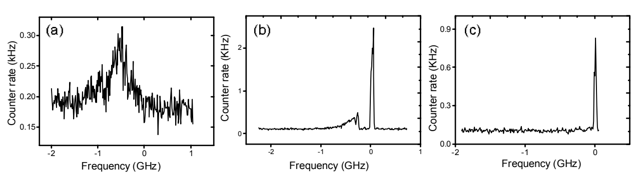

Figure 3. Scan the cooling laser over the resonance. (a) A cloud of ions is trapped. (b) Two ions are trapped. (c) A single ion is trapped.

The cooling laser for the $5s^2\ ^1S_0$ - $5s5p\ ^3P_1$ transition is provided by a frequency quadrupled semiconductor laser system. A diode laser at 922 nm is frequency stabilized via the Pound-Drever-Hall (PDH) locking technique [24, 25] to a reference cavity. The frequency stabilized laser beam is further amplified from 30 mW to 500 mW, using a tapered amplifier. The second harmonic at 461 nm is generated via a frequency doubling cavity with an output power of 120 mW. The output pumps a second frequency doubling cavity to generate 1 mW UV light at 230.6 nm. The UV light passes through an electro-optical modulator (EOM) which creates sidebands tunable between 0 and 100 MHz.

To figure out the residual magnetic field effect on the single ion, a Zeeman shift measurement is done. The cooling light is circularly polarized and pumps the ion into its extreme Zeeman sub-levels ($m_F=\pm 9/2 \leftrightarrow m_F=\pm 11/2$) so that we can treat the cooling transition as an effective two-level transition. By changing the offset magnetic field via changing the current in a set of Helmholtz coils, the magnetic sublevels of the ion are split and the effective cooling transition is shifted. Depending on the polarization of the light with respect to the quantization axis provided by the magnetic field, the transition can be shifted toward lower or higher frequencies. The crossing point of the two shifts for the two different circular polarizations gives the zero magnetic field in this direction. Stray magnetic fields are thus compensated in all three directions to 10 mG level.

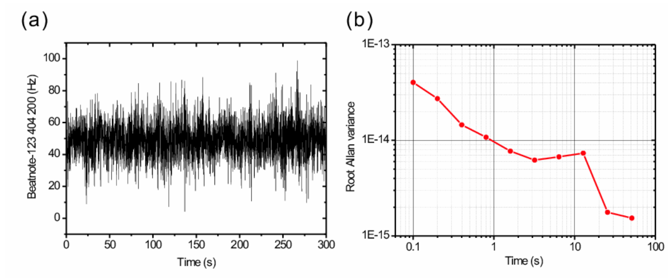

Figure 4. (a) The beatnote at 123.4042 MHz is measured with two frequency counters in a dead-time free configuration using a gate time of 0.1 s. The relative linear cavity drift of 1.2 Hz/s is subtracted from the data. (b) The Allan variance shows a relative frequency stability to be 4×10^{-15} at 3 s and the relative linewidth to be less than 1.34 Hz.

B. Narrow linewidth clock laser

The monolithic isolated end-pumped ring Nd:YAG laser (MISER) for the high resolution spectroscopy has been described in detail before [20,21,26-28]. The performance of this clock laser is checked again by locking it onto two independent high-finesse ultra-stable cavities. The cavities, one made of ULE and the other of Zerodur, are mounted in two separate temperature stabilized, active-vibration isolated vacuum chambers. A typical 300s beat note frequency measurement between these two independently locked laser beams is shown in Fig. 4(a). The corresponding Allan deviation is shown in Fig.4 (b). This measurement shows a relative laser linewidth of less than 1.34 Hz, with an integration time up to 3s. This value is similar to the best result achieved before.

C. Absolute frequency measurement

In order to observe the spectrum of the narrow clock transition, the ultra-stable IR laser is frequency quadrupled. As shown in Fig. 2, the first frequency doubling system is employed to produce 473 nm blue light with a periodically-poled KTP crystal (PPKTP). A 13 m long single-mode fiber is used to transfer the blue light to the ion trap laboratory and AOM4 is implemented for the fiber noise compensation. A second frequency doubling system using BBO crystal converts the 473 nm blue light into UV light near 236.5 nm on resonance with the clock transition.

The absolute frequency measurement of the $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ transition in $^{115}In^+$ consists of two parts: measuring the IR light (946 nm) frequency and the center frequency $f_{spectrum}$ of the quantum-jump spectrum by scanning AOM5 (in UV).

Spectroscopy of the clock transition is performed with optical-optical double resonance and quantum jump detection [17, 29]. The cooling and clock laser systems are used to excite the ion from $5s^2\ ^1S_0$ to $5s5p\ ^3P_1$ and $^3P_0$ states, respectively. With this electron shelving method, nearly 100 % detection efficiency of the transition to the metastable state $^3P_0$ can be achieved.

To prevent the AC Stark shifts induced by cooling laser, the cooling laser and clock laser beams are applied alternately by means of two mechanical shutters. The sequence of the shelving detection scheme and typical quantum jump signals are given in Fig. 5(a), and (b), respectively. Fig. 5(c) is a single scan spectrum for a clock laser power=100 nW (in UV). The clock laser beam waist is ≈ 20 μm. This saturated spectrum shows a spectral broadening linewidth of 1 kHz with maximum transition probability of 50 %.

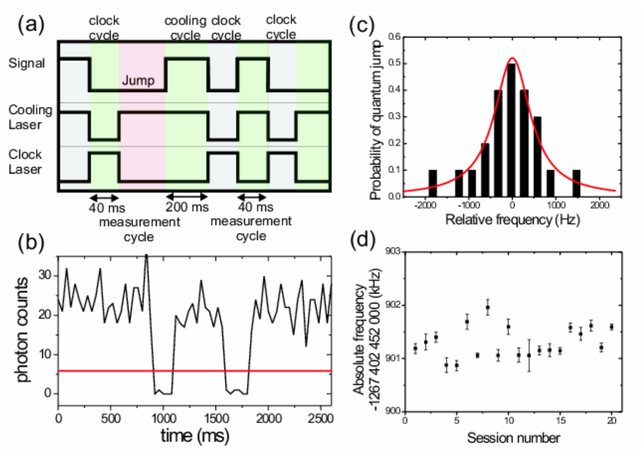

Figure 5. (a) The sequence of the electron shelving detection scheme. (b) A typical quantum jump signal. (c) A single scan spectrum taken for clock laser = 100 nW, leading to saturation broadening of the clock transition with a FWHM of 1 kHz ±100 Hz. It is fitted with Lorentzian function and the laser beam waist is ≈ 20 μm. (d) 20 sessions frequency measurements are made on 13 separate days in four months. The cesium clock is calibrated between the 9th and 10th sessions. The error bars indicate the total uncertainty from the comb measurements and the peak position determinations of the spectrum.

The absolute frequency of the clock transition is given by

$$f_{In^+} = 4(Nf_{rep} \pm f_0 \pm f_{beat} + f_{AOM3}) + 2f_{AOM4} + f_{spectrum} \quad (1)$$

Here N is the mode number of the frequency comb that beats with the clock laser, f_{beat} is the beatnote frequency. The method to determine N and the signs of offset and beatnote frequencies is described in [30]. All counters as well as the synthesizers that drive the AOMs are referenced to the cesium atomic clock. Fig. 5(d) shows our 20 measured results made on 13 separate days in four months. The cesium clock is compared against UTC time at the PTB between the 9th and 10th session measurements. It has an accuracy of 2.35×10^{-13}, which corresponds to a 298Hz correction to the measured value. The uncertainty of the calibration is about 2×10^{-14}, owing to the uncertainty of UTC (PTB) with respect to SI second.

A dominant systematic shift for In^+ is the linear Zeeman shift. The magnetic field dependences of 658±16 Hz/G and -620±41 Hz/G for the |F=9/2, m_F=±9/2> to |F=9/2, m_F=±7/2> components that we excite are further measured, and in good agreement with the value in [15]. The systematic uncertainty is thus less than 7 Hz from the magnetic field compensation. Other systematic frequency shifts such as the second-order Doppler shift or quadratic Stark shift due to stray fields are a few orders of magnitude smaller than the Zeeman shift at the ion temperature of 150 μK [31], and are negligible in the current level.

In Fig. 5(d), a weighted averaging method is used to determine the line center. The absolute frequency of the $^{115}In^+$ $5s^2\ ^1S_0$ - $5s5p\ ^3P_0$ clock transition after correction is determined to be

$$f_{In^+} = 1,267,402,452,900,967 \pm 57\,(stat.) \pm 26\,(sys.)\,Hz$$

Each data point shown in Fig.5(d) is the mean value of around one hour measurement. The error bars show the total uncertainty from the comb measurements and the peak position determinations of the spectrum. The value of 57 Hz is the statistical standard error, and 26 Hz is the systematic error, which is derived from the uncertainty of the cesium clock calibration and magnetic field compensation. The total uncertainty is 63 Hz, assuming uncorrelated statistical and systematic errors. This gives the accuracy of the current measurement at 5.0×10^{-14}. This new absolute frequency is approximately 1 kHz higher than the result in [19].

Recently, we remeasure the absolute frequency of In^+ clock transition with a hydrogen maser as the reference. This preliminary measurement shows a smaller error bar in each measurement and decreased scattering of measured values. The main contribution should come from the improved stability of reference used in the measurement. Fig.6 shows ten measured absolute frequency in one continuous week together with the results based on Cs clock for comparison. The red lines in the figure stand for the weighted mean of the respective measurements based on different reference. The uncertainty is not figured out here and will be analyzed in the near future.

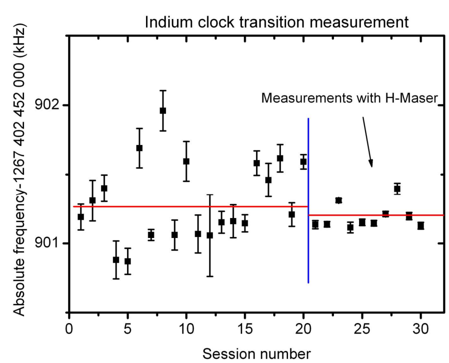

Figure 6. Preliminary measurment of absolute frequency of $^{115}In^+$ clock transition with a hydrogen maser as the reference.

Fig. 7 shows a high resolution spectrum of the clock transition. The clock laser power is reduced to 15 nW to avoid saturation broadening. The dominant linear cavity drift of 1

Hz/s is actively compensated by controlling the driving frequency of AOM2. In order to minimize any influence from the mid-term (20-30s) instability of the clock laser, we decrease the scanning time to less than 20 s. Further, high signal to noise ratio spectra with saturated power are obtained before and after the fine scanning with this weak power for the fine compensation of the cavity drift. Forty scans over the resonance are superimposed and fitting the spectrum with a Lorentzian curve results in a linewidth of 43 Hz (FWHM) of the center peak. However, there is still a broad background, which most likely results from the mid-term instability of the clock laser for the excitation.

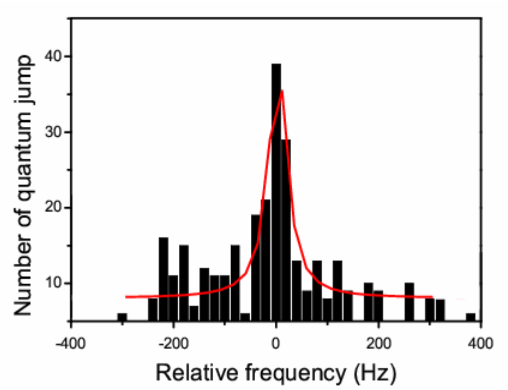

Figure 7. High resolution spectrum taken for clock laser = 15 nW to avoid the saturation broadening. Forty scans over the resonance are superimposed, and fitting the spectrum with a Lorentzian function results in a center linewidth of 43 Hz (FWHM). Each scan lasts less than 20 s. The broad background is most likely due to the mid-term instability of the clock laser.

Conclusions

The absolute frequency of the $^{115}In^{+}$ $5s^2$ $^1S_0$ - $5s5p$ $^3P_0$ transition has been measured and the accuracy is about four times improved and down to 10^{-14} level. Further investigation for the 1 kHz gap and more accurate and stable measurements will be done with a hydrogen maser as the reference in the near future. In addition, another high performance reference cavity for clock laser is being prepared for the high resolution spectroscopy. After careful examination of various possibilities which can cause a spectral broadening, we believe it is most likely due to the nonlinear drifting in the mid to long term of the clock laser. At present, the laser frequency variation is compensated in a linear fashion which is not immune to the higher order behavior. Finally, a new ion trap is being built to reduce the RF heating, which presently limits our trap stability. Currently, a linewidth of the clock transition spectrum of 43 Hz is observed, but with a relatively broad background. The work to further improve the results is still on-going, and is vital to achieve the natural linewidth of the clock transition.

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