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Absolute frequency and isotope shift measurements of the cooling transition in singly ionized indium

Y.H. Wang[1], R. Dumke[2], J. Zhang[1], T. Liu[1], A. Stejskal[1], Y.N. Zhao[1], Z.H. Lu[1,a], L.J. Wang[1,b],
Th. Becker[3], and H. Walther[3]

[1] Max-Planck Research Group, Institute of Optics, Information and Photonics, University Erlangen-Nuremberg, Günther-Scharowsky-Str. 1, Building 24, 91058 Erlangen, Germany
[2] School of Physical and Mathematical Sciences, Nanyang Technological University, 1 Nanyang Walk Block 5, Level 3, Singapore 637616, Singapore
[3] Max-Planck Institute of Quantum Optics, Hans-Kopfermann-Str.1, 85748 Garching, Germany

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Abstract. We report greater than two orders of magnitude improvements in the absolute frequency and isotope shift measurements of the In^+ $5s^2$ $^1\mathrm{S}_0$ $(F = 9/2)$–$5s5p$ $^3\mathrm{P}_1$ $(F = 11/2)$ transition near 230.6 nm. The laser-induced fluorescence from a single In^+ in a radio-frequency trap is detected. The fourth-harmonic of a semiconductor laser is used as the light source. The absolute frequency is measured with the help of a frequency comb referenced to a Cs atomic clock. The resulting transition frequencies for isotopes $^{115}\mathrm{In}^+$ and $^{113}\mathrm{In}^+$ are measured to be 1 299 648 954.54(10) MHz and 1 299 649 585.36(16) MHz, respectively. The deduced cooling transition frequency difference is 630.82(19) MHz. By taking into account of the hyperfine interaction, the isotope shift is calculated to be 695.76(1.68) MHz.

PACS. 31.30.Gs Hyperfine interactions and isotope effects, Jahn-Teller effect – 32.30.Jc Visible and ultraviolet spectra – 06.20.F– Units and standards

1 Introduction

Ion trapping is a well studied technique for high resolution spectroscopy and for optical frequency standards [1–8]. For these applications, a single ion is preferred instead of a cloud of ions. A single ion in a radio-frequency trap can be cooled down to the lowest vibrational state within the Lamb-Dicke regime. Since a single ion is well isolated from the experimental surroundings, the collisional broadening, Doppler broadening, and transit-time broadening have almost no effect on the observed spectrum.

We wish to investigate the frequency stability and possible systematic shifts of a single indium ion for use as an optical frequency standard. In order to build an optical frequency standard based on a single indium ion, it is helpful to cool the ion down to the vibrational ground state. Therefore, it is necessary for us to know the exact frequency of the cooling transition before proceeding to optical frequency measurements. For the indium ion (In^+), the cooling transition is the $5s^2$ $^1\mathrm{S}_0$–$5s5p$ $^3\mathrm{P}_1$ intercombination transition, which has a 360 kHz linewidth. Throughout the paper, we refer to this intercombination transition as the cooling transition.

There are two stable indium isotopes, $^{115}\mathrm{In}$ and $^{113}\mathrm{In}$. Figure 1 shows the energy levels and relevant transitions of both singly ionized isotopes. Several groups have measured the cooling transition in $^{115}\mathrm{In}^+$, yet their results are not entirely consistent. Paschen and Campbell made an extensive survey of the $^{115}\mathrm{In}^+$ spectrum in the wavelength range 210–950 nm [9]. Later, Peik et al. measured the cooling transitions at 230.6 nm using an ion cloud stored in a radio-frequency trap. Their result was calibrated by tellurium Doppler-free saturation absorption spectroscopy [10]. They reported that the absolute frequency of the transition $5s^2$ $^1\mathrm{S}_0$ $(F = 9/2)$–$5s5p$ $^3\mathrm{P}_1$ $(F = 11/2)$ was 1 299 648 872(600) MHz, 9 GHz lower than reference [9]. Larkins and Hannaford confirmed the value of reference [10] and claimed an order of magnitude better in precision (1 299 648 692(60) MHz) [11]. Their measurements were made on a cloud of In^+ emitted from a neon-filled hollow-cathode lamp.

With the advance of the frequency comb technique, it is now possible to perform higher precision measurement of the $^{115}\mathrm{In}^+$ cooling transition. In addition, the cooling transition of the $^{113}\mathrm{In}^+$ ion can also be measured very accurately. So far, only one group has reported the cooling transition measurement for $^{113}\mathrm{In}^+$ [10]. The isotope shift can be deduced from the absolute frequencies of the

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[a] e-mail: `zlu@optik.uni-erlangen.de`
[b] e-mail: `lwan@optik.uni-erlangen.de`

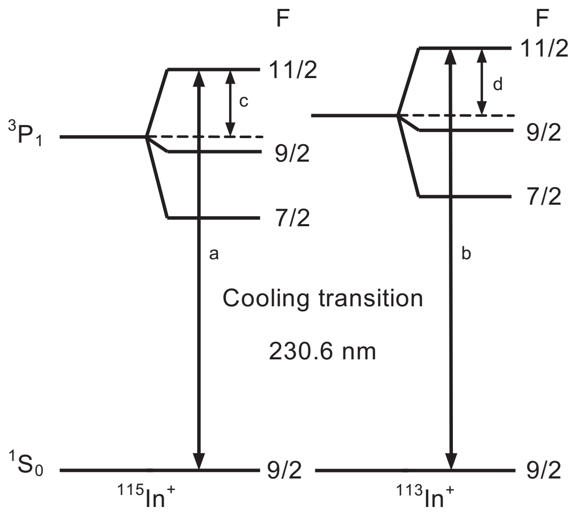

Fig. 1. Relevant In$^+$ energy levels and transitions. The vertical energy axis is not drawn to scale.

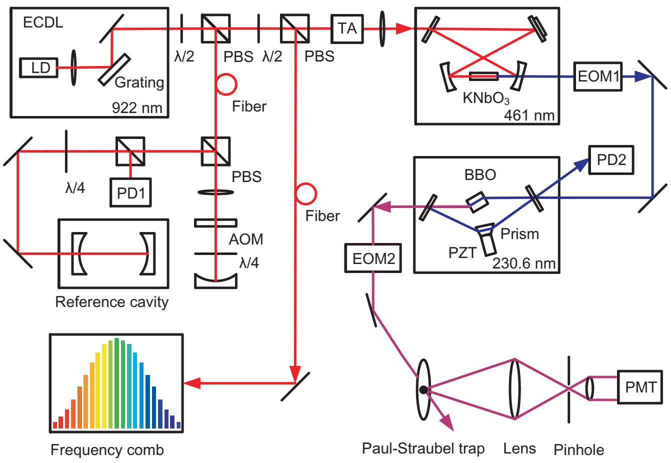

Fig. 2. (Color online) Schematic diagram of the experimental set-up. ECDL: external cavity diode laser; LD: laser diode; TA: tapered amplifier; EOM: electro-optical modulator; PD: photo diode; PBS: polarization beam splitter; AOM: acousto-optical modulator; PMT: photo-multiplier-tube.

cooling transition in ^{115}In$^+$ and ^{113}In$^+$. The cooling transition frequency difference of 0.64(7) GHz for In$^+$ was first reported in reference [10]. Its accuracy was limited by the tellurium spectroscopy calibration technique.

The measurement of optical isotope shifts have contributed to our understanding of nuclear charge distributions and of atomic structures [12,13]. The isotope shift is due to the differences in nuclear mass (mass shift) and nuclear charge distribution (field shift) [14]. Normally, the mass shift is dominant for lighter elements. For heavier elements, the field shift dominates and the measurement provides us information about the difference in the mean square nuclear charge radius between isotopes. By taking advantage of experimental precision achievable for the optical transition frequencies, it allows one to determine nuclear charge radii much more accurately (less than 0.1 fm) than from electron scattering measurements, despite the fact that the nuclear size is five orders of magnitude smaller than the atomic size.

In this paper, we report on absolute frequency measurements of the $5s^2\ {}^1\mathrm{S}_0\ (F = 9/2)$–$5s5p^3\mathrm{P}_1\ (F = 11/2)$ cooling transitions for both ^{115}In$^+$ and ^{113}In$^+$. We also determine the isotope shift from our measured results and by using known hyperfine structure constants. These measurements are done on a single laser-cooled ion, stored in a radio-frequency trap. A compact, tunable and narrow-linewidth diode laser, frequency quadrupled to 230.6 nm is used as the light source. The absolute frequencies are determined by a frequency comb [15,16], which is referenced to a Cs atomic clock. Our results show a greater than two orders of magnitude improvement compared to previous results.

2 Experimental method

The experimental set-up is shown in Figure 2. It consists of three main parts: the cooling laser source at 230.6 nm, the Paul-Straubel ion trap [17], and a frequency comb system for absolute frequency measurement.

The cooling laser is based on a semiconductor laser system (Toptica Photonics SHG110), similar to that used in reference [18]. An extended cavity diode laser (ECDL) in Littow configuration emits near-infrared (NIR) light at 922 nm with a 30 mW power. The light is further amplified to 500 mW by a tapered amplifier. The NIR light is frequency doubled to 461 nm by using a potassium niobate (KNbO$_3$) crystal. After the first frequency doubling, approximately 100–150 mW of blue light is produced. The blue light is frequency doubled again by a BBO crystal, producing approximately 1 mW of ultraviolet (UV) light at 230.6 nm.

The free-running ECDL has a typical linewidth of 1 MHz at 922 nm. Taking into account the frequency quadrupling system, we have a 4 MHz linewidth in the UV, which is too broad compared to the 360 kHz linewidth of the cooling transition. Therefore, it is important to reduce the NIR laser linewidth. This is achieved by locking the laser to a high-finesse cavity using the Pound-Drever-Hall locking technique [19]. The sideband for locking is created by modulating the current of the ECDL at 20 MHz. The performance of the ECDL after locking is investigated by looking at the error signal. The estimated linewidth is less than 10 kHz in the NIR.

The Paul-Straubel trap is placed in a stainless steel vacuum chamber. The trap consists of three ring electrodes made of beryllium copper alloy. The center ring has an inner hole diameter of 1 mm, which is used to trap a small amount of ions and confine them to within the Lamb-Dicke regime. A 10 MHz RF frequency is applied to the center ring with a 35 V DC offset. This results in a time averaged pseudo-potential with oscillation frequencies of 1.5 MHz in the axial direction and 0.9 MHz in the radial direction. These secular frequencies are measured by applying a driving frequency to one of the outer

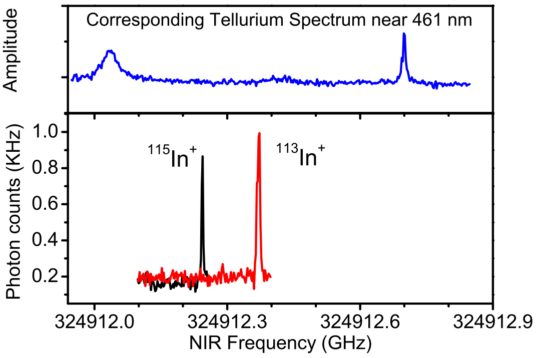

Fig. 3. Upper: Tellurium saturated absorption Doppler-free spectrum, recorded at 461 nm by scanning the cooling laser. Lower: single ion laser-induced-fluorescence signal. The upper spectrum is at the doubled frequency.

ring electrodes and observing the drop in the fluorescence signals as the driving frequency is swept through the resonances.

The natural mixture of indium (95.7% ^{115}In, 4.3% ^{113}In) is evaporated from an oven by applying a 1.8 A current for a few seconds. The indium ions are created through photonionization process with a single 410 nm diode laser. This wavelength is close to the $5s^25p\ ^2\mathrm{P}_{1/2}$–$5s^26s\ ^2\mathrm{S}_{1/2}$ transition as well as the $5s^26s\ ^2\mathrm{S}_{1/2}$ to the autoionizing $5s5p^2\ ^2\mathrm{D}$ state [20,21]. Initially, a cloud of ions is trapped. By applying a blue-detuned laser, the ions are heated and subsequently some ions leave the trap region. This procedure is repeated approximately five times to obtain a single ion. The fluorescence signal from the ions is collected by a three-lens system ($f/1.4$), focused to a 200 μm pinhole, and then detected by a solar-blind photomultiplier tube. The overall detection efficiency is 0.1%.

It is very easy to trap single $^{115}\mathrm{In}^+$ ions because of their abundance. In order to trap rarer $^{113}\mathrm{In}^+$, we use the fact that the cooling transition frequency in $^{113}\mathrm{In}^+$ is higher than that in $^{115}\mathrm{In}^+$. When the laser frequency is in between the two cooling transition frequencies, the $^{115}\mathrm{In}^+$ ion will be heated, and the $^{113}\mathrm{In}^+$ ion will be cooled. Typically, an $^{113}\mathrm{In}^+$ ion can be trapped in 15–20 attempts.

Figure 3 (lower panel) shows laser-induced-fluorescence (LIF) signals for both isotopes by scanning the free-running cooling laser frequency. The upper panel is the corresponding tellurium spectrum near 461 nm.

For our experiment, the bichromatic cooling technique [22] is further developed to cool the ion down to the vibrational ground state. The UV light passes through an electro-optical modulator EOM2 which creates a sideband that can be tuned in the range of 0 to 100 MHz. The carrier (the incident UV light) is red-detuned a few orders of micromotion sidebands away from the In$^+$ resonance. It produces nearly no fluorescence signal from the ion. As a consequence, the ion can be efficiently Doppler-cooled from the higher vibrational states. The UV sideband is red-detuned to the first dominant secular sideband (1.5 MHz) to cool the ion to its vibrational ground state.

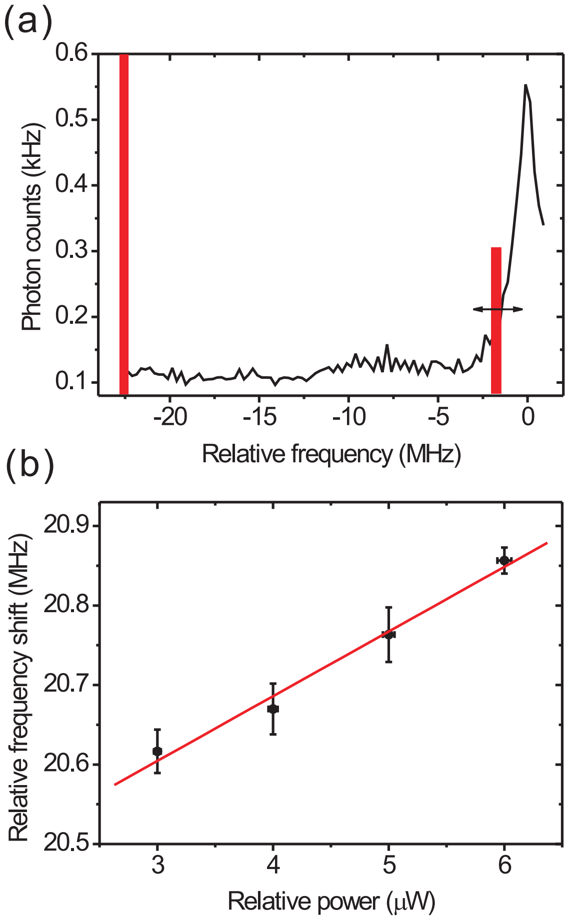

Fig. 4. (a) Bichromatic cooling of the ion transition spectrum. The line at −25 MHz indicates the strong carrier of the cooling light and the line at −2 MHz indicates the sideband which can be tuned with respect to the carrier. The spectrum is recorded by scanning the weak sideband over the resonance transition, indicated by the arrow. (b) Power-dependent ac Stark shift of the cooling transition. Measurements are fitted with a linear line, and the intercept gives the real transition frequency.

The spectrum which is shown in Figure 4a is recorded by scanning the UV sideband across the cooling transition. The micromotion of the ion is minimized by optimizing the DC offset voltage applied to the trap.

The absolute frequency measurement of the cooling transition is done by transferring 1 mW NIR light through a 60 m long single-mode fiber to another laboratory, where a Ti:sapphire femtosecond laser frequency comb (Menlo Systems FC-8004) is located. The frequency comb's repetition rate f_{rep} and offset frequency f_0 are both locked to a Cs atomic clock (Agilent 5071A).

The absolute frequency of the cooling transition is measured by beating the NIR light with the frequency comb, and then adding the center frequency of the bichromatic scanning spectrum of the EOM2. Due to the strong carrier light field, the measured cooling transition is ac Stark shifted. When the laser is red-detuned, the measured frequency is larger than the real value. The frequency shift is proportional to the intensity of the light

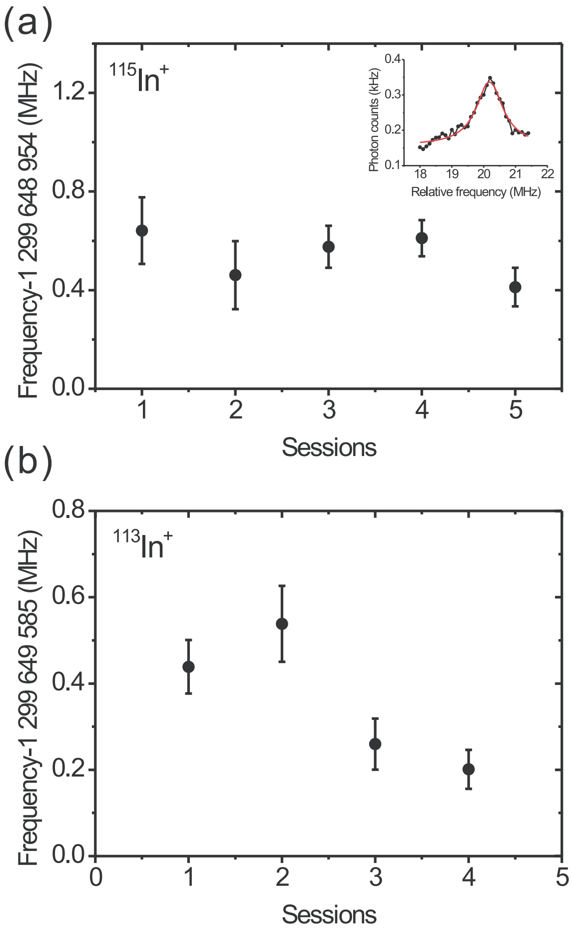

Fig. 5. Absolute frequency measurements for the cooling transition of (a) a single $^{115}\mathrm{In}^{+}$ ion, and (b) a single $^{113}\mathrm{In}^{+}$ ion. The error bars show the total statistical uncertainties from the frequency comb and ac Stark shift measurements. The inset in (a) shows one of the measured transition spectra.

field. This ac Stark shift should be taken into account for this high precision measurement. The measured light shift result is shown in Figure 4b, for various cooling laser powers. Each data point is the average of five measurements. The intercept f' after linear fitting is used as the intrinsic transition frequency. Hence, the absolute frequency is given by

$$f = 4(N f_{rep} \pm f_0 \pm f_{beat}) + f'. \tag{1}$$

Here N is the mode number of the frequency comb that beats with the laser, f_{beat} is the beat-note frequency. The method to determine N and the signs of the offset and the beat-note frequencies are described in reference [23].

3 Results and discussion

The absolute frequency measurements of both isotopes are shown in Figure 5. The error bars indicate the total statistical uncertainties from the frequency comb and light shift measurements. For $^{115}\mathrm{In}^{+}$ and $^{113}\mathrm{In}^{+}$, the frequencies of the cooling transitions are 1 299 648 954.54(10) MHz and 1 299 649 585.36(16) MHz, respectively. The first value is not consistent with reference [11], with a discrepancy of 260 MHz. However, our values are in good agreement with the values in reference [10]. Furthermore, we found that there was a similar discrepancy of 277 MHz between the recent, more precise narrow line transition frequency in reference [24] and the value reported in reference [11].

The corresponding cooling transition frequency difference from our measurements is derived to be 630.82(19) MHz. Since the nuclear moments of the isotopes are slightly different, hyperfine interactions contribute a frequency shift, which can be expressed as [25]

$$\Delta f = \frac{1}{2}AK + \frac{B}{8}\frac{3K(K+1) - 4I(I+1)J(J+1)}{I(2I-1)J(2J-1)}, \tag{2}$$

where $K = F(F+1) - I(I+1) - J(J+1)$. A and B are the hyperfine constants that can be determined if all three hyperfine splittings for In^{+} are measured. However, due to the low transition rates of $^{1}\mathrm{S}_0(F = 9/2)$ to $^{3}\mathrm{P}_1(F = 9/2, 7/2)$, the LIF signal from a single ion is too weak to detect. Thus, we use the experimental results of $A_{^{115}\mathrm{In}^{+}} = 6960(90)$ MHz and $B_{^{115}\mathrm{In}^{+}} = -480(420)$ MHz for state $^{3}\mathrm{P}_1$, which were measured for an ion cloud [10]. Using another experimental result for the ratio of magnetic dipole moments ($A_{^{115}\mathrm{In}^{+}}/A_{^{113}\mathrm{In}^{+}} = 1.00213$) [26] and electric quadrupole moments ($B_{^{115}\mathrm{In}^{+}}/B_{^{113}\mathrm{In}^{+}} = 1.0138$) [27] for both isotopes, we can determine the hyperfine interaction shift $c - d = 64.94(1.67)$ MHz (see Fig. 1). The isotope shift that is measured from the centroid of the hyperfine splitting is then

$$\begin{aligned}(b-d)-(a-c) &= 630.82(19) + 64.94(1.67)\\ &= 695.76(1.68)\ \text{MHz}.\end{aligned} \tag{3}$$

For reference [10], the uncertainty of this value is limited by the cooling transition frequency measurement, which has a 70 MHz uncertainty. For our measurement, the uncertainty is decreased to 1.68 MHz and is only limited by the contribution of the hyperfine interaction.

Finally, we discuss the error budget of the measurement. The ion is well within the Lamb-Dicke regime, such that both recoil and first-order Doppler shifts vanish. Since the temperature of the laser-cooled single ion is lower than 1 mK [22], the second-order Doppler shift is negligible (below 1 mHz). The Zeeman shift ($m_F = \pm 9/2 \leftrightarrow m'_F = \pm 11/2$) of the cooling transition for both isotopes is 22 GHz/Tesla. The Earth magnetic field is compensated with Helmholtz coils in all three directions to $<5 \times 10^{-7}$ Tesla level, corresponding to an 11 kHz uncertainty. Typically, the beat-note measurements done by the frequency comb contribute a 15 kHz uncertainty at 230.6 nm. The cavity drift of 1 Hz/s (in the NIR) is accounted for through data analysis and has a negligible uncertainty. The main limitation in our experiment comes from the 1 MHz linewidth spectrum when doing bichromatic scanning, and linear fittings used afterward for canceling the ac Stark shift. This uncertainty contribution is approximately 50 kHz.

4 Conclusions

We have trapped and laser-cooled single $^{115}\mathrm{In}^+$ and $^{113}\mathrm{In}^+$ ions in a Paul-Straubel trap. The absolute frequencies and the isotope shift of the intercombination transition $5s^2\ {}^1\mathrm{S}_0$ $(F = 9/2)$–$5s5p\ {}^3\mathrm{P}_1$ $(F = 11/2)$ are measured with a frequency comb. In comparison to other groups' results, more than two orders of magnitude improvements are obtained by using a single ion.

The precision of our isotope shift measurement is limited by the contribution of the hyperfine interaction measurement accuracy. With a better measurement of the hyperfine splitting, it is possible to improve the isotope shift measurement precision to the level of hundreds of kHz. This, together with the advances in the theoretical calculation of the atomic structure of indium, can pave way to high-precision determination of the nuclear charge radius of indium.

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