TOWARDS AN INDIUM SINGLE-ION OPTICAL CLOCK

E. PEIK, J. ABEL, TH. BECKER, M. FRIES, J. VON ZANTHIER, H. WALTHER
Max-Planck-Institut für Quantenoptik
and Sektion Physik der Ludwig-Maximilians-Universität,
Hans-Kopfermann-Str. 1, 85748 Garching, Germany

We are investigating the $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_0$ transition of a single trapped laser-cooled ${}^{115}\mathrm{In}^+$ ion as a possible optical frequency standard. This line with a natural linewidth of only 1.1 Hz, corresponding to a Q factor of $1.2 \cdot 10^{15}$, is highly immune to systematic frequency shifts due to external perturbations. For sideband laser cooling and fluorescence detection of the indium ion the $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ transition at 230.6 nm is excited. Temperatures below 100 μK and a mean vibrational quantum number $\langle n \rangle < 1$ of the ion in the trap have been reached. For the clock transition a resolution of $3 \cdot 10^{-13}$ (350 Hz linewidth) has been obtained so far, limited by the laser linewidth. The absolute frequency of the ${}^1S_0 \rightarrow {}^3P_0$ transition has been measured by making a link to known reference frequencies of molecular iodine and methane using a frequency chain.

1 Introduction

A single laser-cooled ion in a radiofrequency trap represents a good approximation to the spectroscopic ideal of a motionless point-like absorber in a perturbation-free environment. It is consequently regarded as a perfect system for an optical atomic clock of very high accuracy [1]. With a frequency-stable laser locked to a narrow absorption resonance of a trapped ion a relative stability $\sigma_y(1\mathrm{s}) = 10^{-15}$ and an accuracy of a few parts in 10^{-18} seems possible. High resolution spectroscopy of forbidden optical transitions has yielded sub-kHz linewidths in Hg^+ (see [2] and the contribution of J. Bergquist in this volume), Ba^+ [3] and, more recently, in Sr^+ [4], Yb^+ [5] and In^+. In the first four alkali-like ions, quadrupole transitions between S and D states are investigated. We are studying the earth-alkali-like spectrum of In^+, where the lowest two levels $5s^2\,{}^1S_0$ and $5s5p\,{}^3P_0$ are connected by a hyperfine-induced electric dipole transition at 236.5 nm wavelength which has a natural linewidth of only 1.1 Hz [6]. Fig. 1 shows the relevant energy levels of In^+.

As a candidate for a primary optical frequency standard In^+ has three main advantages: (i) Since the electronic angular momenta in both states of the clock transition ${}^1S_0 \rightarrow {}^3P_0$ are vanishing the coupling of the transition to external electromagnetic fields is very small. In fact, the ${}^3P_0$ state is not a pure $J = 0$ state but contains small hyperfine admixtures of the $5s5p$ levels ${}^3P_1$ and ${}^1P_1$. These perturbations are responsible for the non-vanishing electric dipole moment between this state and the ground state [6]. For the same reason there exists a small difference between the Landé g-factors of ${}^1S_0$ and ${}^3P_0$. This leads to a linear Zeeman shift of the clock transition which we measured to 223 Hz/G for the $m_F = \pm 1/2 \rightarrow m_F = \pm 1/2$ Zeeman components. Shielding of magnetic fields to the 10 μG level will ensure the predicted 10^{-18} reproducibility of the frequency standard. We estimate the quadratic Stark shift of the clock transition to be below 1 $\mathrm{mHz}/(\mathrm{V}/\mathrm{cm})^2$. Since both levels have no quadrupole moment they do not couple to the electric field gradient of the trap. (ii) To excite the clock transition a reliable and frequency-stable laser source is required. For the In^+ transition there is the advantage that it coincides with the fourth harmonic of the 946 nm laser line of Nd:YAG. The availability of this diode-laser-pumped solid-state laser is of significance for the long-term operation of the optical frequency standard. For the measurements described here, still a conventional setup made from discrete optical elements is used [7,8], however, in the future it is conceivable to use a monolithic ring-laser [9,10] – like those employed in the laser interferometers for the detection of gravitational waves – offering high intrinsic stability and a compact design. (iii) Laser cooling of In^+ is performed by using the narrow intercombination line $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$. With a natural linewidth of 360 kHz, the photon scattering rate on this transition is sufficiently high to detect a single ion via the resonance fluorescence. At the same time the line is narrow enough to allow the resolution of the 1 MHz vibrational frequencies of the ion in the trap. In this parameter regime sideband cooling [1,11,12] permits to cool the ion to the motional ground state of the trap and to reach temperatures below 100 μK. This reduces the fractional second-order Doppler shift to values below 10^{-19}.

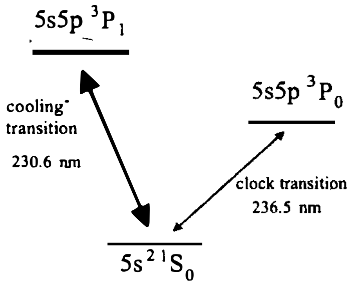

Figure 1: The lowest energy levels of the In^+ ion.

2 Sideband cooling of the indium ion

The ion is trapped in a quadrupole radiofrequency trap that is a geometrical variant of the original Paul trap and basically consisting of only a ring electrode (Paul-Straubel trap [13,14]). This type of trap is relatively simple to fabricate in miniature size (1 mm diamter of the ring), making it easy to confine the ion to a region in space that is smaller than the optical wavelength (Lamb-Dicke regime). It is also a geometrically quite open structure that allows good optical access to the trapped ion. The trap is driven with a RF field at 10 MHz; the oscillation frequencies in the time-averaged pseudopotential are 1.4 MHz in the axial and 0.9 MHz in the radial directions.

Sideband-cooling is performed through laser excitation of the $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ intercombination line at a wavelength of 230.6 nm. In this transition the hyperfine component $F \rightarrow F+1$ (where $F = I = 9/2$) is excited with circularly polarized light in vanishing magnetic field, so that optical pumping between the Zeeman sublevels results in a closed two-level system. The laser radiation is produced by a frequency-doubled stilbene-3 ring dye laser. The laser is frequency-stabilized to a reference cavity by using a high-bandwidth electronic servo system, resulting in a laser linewidth below 10 kHz.

In order to increase the initial cooling power for a highly vibrationally excited ion and to be able to record high resolution spectra of the cooling transition we use a bichromatic cooling method [15]. The laser beam is passed through an electro-optical modulator with variable modulation frequency. Through phase modulation two weak sidebands containing about 0.1% of the total power each were created around the original laser frequency. The carrier of this spectrum was far detuned (by about −40 MHz) and efficiently cooled the ion in higher vibrational levels, when excited e.g. by collisions with molecules of the residual gas. The high-frequency phase modulation sideband was used to cool the ion to the quantum ground state when tuned to the first low-frequency vibrational sideband and could also be scanned over the carrier frequency by changing the drive frequency of the electro-optical modulator. An experimental excitation spectrum obtained in this way is shown in Fig. 2. The spectrum is dominated by the carrier and the excitation of vibrational sidebands is very weak as can be seen from the magnified deviations between the data and a fitted single Lorentzian curve.

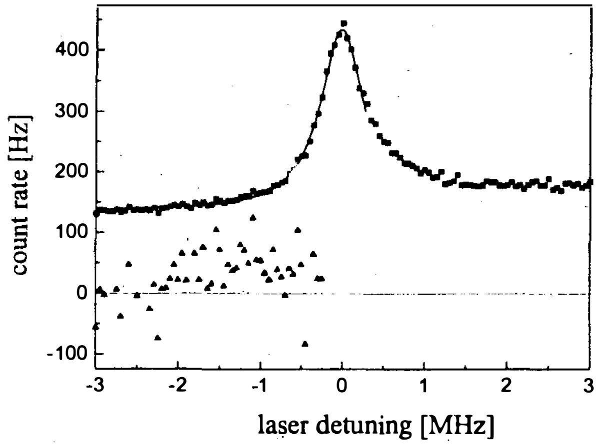

Figure 2: Excitation spectrum of the cooling transition of a single indium ion obtained with bichromatic sideband cooling (averaged over 32 scans). Below: deviations between the data points and a fitted Lorentzian curve (solid line) in tenfold magnification. From the height of the low-frequency vibrational sidebands at −0.9 MHz and −1.4 MHz detuning the mean vibrational quantum number can be determined: $\langle n \rangle = 0.7(3)$.

In the region of laser detunings corresponding to the low-frequency vibrational sidebands an increase of the signal by about 2(1)% of the height of the Lorentzian is observable. In the Lamb-Dicke limit the height of the first red sideband relative to the carrier is given by $\langle n \rangle k^2 x_0^2$ where k is the wavenumber of the cooling laser light and $x_0 = \sqrt{\hbar/2m\omega}$ the extension of the ground state wavefunction (ω: vibrational frequency). We can therefore determine the mean vibrational quantum number $\langle n \rangle = 0.7(3)$, corresponding to a temperature of about 60 μK. This result indicates that the ion is in the vibrational ground state with a probability exceeding 50% for a basically unlimited time duration (several minutes in the actual experiment).

3 High-resolution spectroscopy of the clock transition

Spectroscopy of the narrow ${}^1S_0 \rightarrow {}^3P_0$ line is performed in optical-optical double-resonance using Dehmelt's idea of electron shelving [1]: Excitation of the metastable ${}^3P_0$ level leads to a cessation of the single-ion fluorescence signal on the cooling transition until the level decays or the valence electron is brought back to the ground state by a stimulated process. This method leads to a strong quantum amplification (the absorption of one photon prevents the subsequent scattering of some 10^5 to 10^6 photons) and allows detection of transitions to the metastable state with practically 100% efficiency.

The laser system used for excitation of the $^1S_0 \rightarrow {}^3P_0$ resonance is described in [8]. It consists of a diode-pumped Nd:YAG laser emitting at 946 nm. This laser contains all the necessary tuning elements and is frequency-stabilized to a passive resonator of high finesse. A second diode-pumped Nd:YAG laser is used for power-amplification and efficient generation of the second harmonic at 473 nm wavelength. It is a ring laser, containing only a Nd:YAG crystal and a $KNbO_3$ crystal for frequency doubling. Infrared light from the stable master laser is coupled into this laser to transfer the frequency stability via injection locking. The blue light is coupled into a passive enhancement cavity to generate the UV radiation at 236.5 nm with a BBO crystal.

In order to obtain high-resolution spectra of the clock transition any light-shift and line-broadening by the cooling laser have to be avoided. Both laser beams are applied alternately and blocked using mechanical shutters. After a clock-laser pulse of 20 ms duration the cooling laser is turned on and the fluorescence photons are counted in a 40 ms time interval. If the count rate corresponds to the single-ion fluorescence level the excitation attempt of the clock transition is regarded as unsuccessful and repeated either at the same or at a different frequency of the clock laser. If the ion is not fluorescing the cooling laser is kept switched-on to wait for the decay of the metastable state (lifetime 140 ms) and an excitation is registered. Typically the frequency of the clock-laser radiation was changed in steps of 8 Hz and four excitation attempts were performed at each frequency.

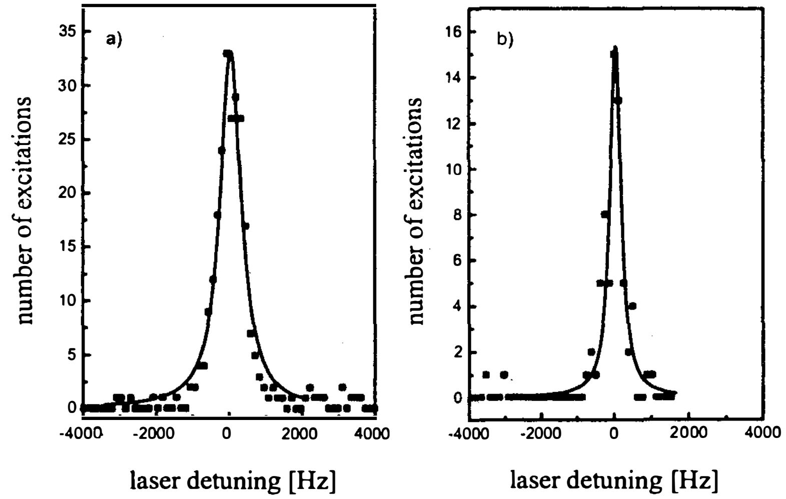

Figure 3: Excitation spectra of the $^1S_0 \rightarrow {}^3P_0$ resonance of a single indium ion obtained in optical-optical double resonance using electron shelving. a) Laser pulses of 20 ms duration and 1 μW power have been used. The line is saturation broadened to 700 Hz linewidth (FWHM). b) The laser power was reduced to 35 nW. The linewidth of 350 Hz is determined by the linewidth of the laser.

Two high-resolution spectra of the clock transition are shown in Fig. 3. At a laser power of 1 μW (spectrum 3a) the resonance is still saturation-broadened to 700 Hz linewidth (full width at half maximum on the ultraviolet frequency scale); at 35 nW laser power the linewidth is reduced to 350 Hz (spectrum 3b). This value corresponds to a spectroscopic resolution $\delta\nu/\nu$ of $3 \cdot 10^{-13}$. According to the above mentioned measurement of the ion's temperature and our present experimental control of electromagnetic fields and vacuum conditions no significant Doppler-, Zeeman-, Stark- or collisional broadening is expected beyond the level of the natural linewidth of 1.1 Hz. The smallest linewidth that we observe experimentally is presently determined by the laser linewidth. The value of 350 Hz for this linewidth is in agreement with an independent determination of the frequency stability of the laser using a second stable optical reference cavity. It is determined mainly by residual vibrations of the high-finesse cavity that is used for the frequency stabilization. Improvements of the isolation of this cavity will probably allow us to resolve the natural linewidth of the ion, leading to a resolution of $8 \cdot 10^{-16}$.

The realization of an optical clock requires first of all a narrow and stable resonance – as it is the case for a trapped indium ion – and secondly a precise frequency-determination in comparison with the cesium clock. We have measured the absolute frequency of the $^{115}\mathrm{In}^+$ $5s^2\,{}^1S_0$ - $5s5p\,{}^3P_0$ clock transition with an accuracy of 3.3 parts in 10^{11} [16]. For this measurement a frequency synthesis chain was used which links the indium transition to a methane stabilized He-Ne laser at 3.39 μm [17] and a Nd:YAG laser at 1064 nm whose second harmonic was locked to a hyperfine component of molecular iodine [18]. The 946 nm Nd:YAG laser whose fourth harmonic drives the In^+ transition was set to the frequency-midpoint between the 1064 nm Nd:YAG laser and a diode-laser at 852 nm using an optical frequency interval divider [19]. The frequency of the diode-laser was compared to another diode-laser at 848 nm that was phase locked to the fourth harmonic of the He-Ne laser at 3.39 μm. A frequency gap of 1.43 THz between the two diode-lasers was bridged with the help of an optical frequency comb generator [20]. The frequency of the $^{115}\mathrm{In}^+$ clock transition was determined to 1 267 402 452 914 (42) kHz, where the accuracy is limited by the uncertainty of the iodine reference.

Parts of this frequency chain were already used in a previous measurement to determine the $1S \rightarrow 2S$ transition frequency in atomic hydrogen with an accuracy of 3.4 parts in 10^{13} [21]. An interesting perspective of this work is the possibility to link the two narrow optical-resonances of atomic hydrogen and the indium ion and to use the high precision of this frequency comparison in a search for temporal variations of the fine structure constant (see e.g. Ref. [22] and the contribution of Th. Damour in this volume).

Acknowledgments

We gratefully acknowledge the collaboration with A. Yu. Nevsky, M. N. Skvortsov, S. N. Bagayev, R. Holzwarth, J. Reichert, Th. Udem, and Th. Hänsch in the absolute optical frequency measurement.

References

1. H. Dehmelt, IEEE Trans. Instrum. Meas. **31**, 83 (1982).
2. J. C. Bergquist, W. M. Itano, F. Elsner, M. G. Raizen, and D. J. Wineland, in *Light Induced Kinetic Effects on Atoms, Ions and Molecules*, Eds.: L. Moi, S. Gozzini, C. Gabbanini, E. Arimondo, and F. Strumia (ETS Editrice, Pisa, 1991).
3. N. Yu, X. Zhao, H. Dehmelt, and W. Nagourney, Phys. Rev. A **50**, 2738 (1994).
4. J. E. Bernard, L. Marmet, and A. Madej, Opt. Commun. **150**, 170 (1998).
5. C. Tamm and V. Bühner, Abstract QFD5, European Quantum Electronics Conference, EQEC '98, Glasgow, 1998.
6. E. Peik, G. Hollemann, and H. Walther, Phys. Rev. A **49**, 402 (1994).
7. G. Hollemann, E. Peik, and H. Walther, Opt. Lett. **19**, 192 (1994).
8. G. Hollemann, E. Peik, A. Rusch, and H. Walther, Opt. Lett. **20**, 1871 (1995).
9. Th. J. Kane and R. L. Byer, Opt. Lett. **10**, 65 (1985).
10. I. Freitag, R. Henking, A. Tünnermann, and H. Welling, Opt. Lett. **20**, 2499 (1995).
11. D. J. Wineland and W. M. Itano, Phys. Rev. A **20**, 1521 (1979).
12. F. Diedrich, J. C. Bergquist, W. M. Itano, and D. J.Wineland, Phys. Rev. Lett. **62**, 403 (1989).

13. N. Yu, W. Nagourney, and H. Dehmelt, J. Appl. Phys **69**, 3779 (1991).
14. C. Schrama, E. Peik, W. W. Smith, and H. Walther, Opt. Commun. **101**, 32 (1993).
15. E. Peik, J. Abel, Th. Becker, J. v. Zanthier, and H. Walther, submitted to Phys. Rev. A.
16. J. v. Zanthier, J. Abel, Th. Becker, M. Fries, E. Peik, H. Walther, R. Holzwarth, J. Reichert, Th. Udem, T. W. Hänsch, A. Yu. Nevsky, M. N. Skvortsov, and S. N. Bagayev, submitted to Opt. Commun.
17. S. N. Bagayev, A. K. Dmitriyev, and P. V. Pokasov, Laser Phys. **7**, 989 (1997).
18. P. A. Jungner, S. Swartz, M. Eickhoff, Jun Ye, J. L. Hall, and S. Waltman, IEEE Trans. Instrum. Meas. **44**, 151 (1995).
19. H. Telle, D. Meschede, and T. W. Hänsch, Opt. Lett. **15**, 532 (1990).
20. M. Kourogi, T. Enami, and M. Ohtsu, IEEE Photon. Techn. Lett. **6**, 214 (1994).
21. T. Udem, A. Huber, B. Gross, J. Reichert, M. Prevedelli, M. Weitz, and T. W. Hänsch, Phys. Rev. Lett. **79**, 2646 (1997).
22. V. A. Dzuba, V. V. Flambaum, and J. K. Webb, Phys. Rev. Lett. **82**, 888 (1999).