Direct frequency comb spectroscopy of trapped ions

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Direct frequency comb spectroscopy of trapped ions is demonstrated for the first time. It is shown that the $4s^2S_{1/2} - 4p^2P_{3/2}$ transition in calcium ions can be excited directly with a frequency comb laser that is upconverted to 393 nm. Detection of the transition is performed using a shelving scheme to suppress background signal from non-resonant comb modes. The measured transition frequency of $f = 761.905.012.7(0.5)$ MHz presents an improvement in accuracy of more than two orders of magnitude.

Optical frequency comb lasers provide a phase coherent link between radio frequency sources and optical frequencies [1, 2]. As a result optical frequencies can be counted, which has made spectroscopy and optical clocks possible with an extremely high accuracy on the order of $10^{-16} - 10^{-17}$ [3, 4]. In most experiments a continuous wave (CW) laser is used as a probe of an atomic or molecular transition, which is then calibrated against a frequency comb (see e.g. [5] for an experiment on atoms, [6] for references to measurements on tightly confined ions, and [7, 8] for measurements on ions in the weak binding limit).

However, frequency combs can also be used for direct excitation, without the need of a CW laser. This is possible because the pulsed output of a comb laser (as seen in the time domain) is equivalent to many equidistant narrow band modes in the frequency domain. Direct frequency comb spectroscopy has been demonstrated in beam experiments [9, 10], atomic vapour cells [11] and cold neutral atoms in magneto-optical traps [12, 13], but not yet on cold ions in an ion trap.

Ion traps provide the opportunity to simultaneously trap different species. By laser cooling one type of ion, the other ions in the trap can be sympathetically cooled [14, 15]. A system to trap and cool ion species, can then be used without big modifications to trap and cool other types of atomic and molecular ions. Frequency combs offer a wide spectrum of frequencies through coherent broadening in non-linear optical fibers [16] and doubling in nonlinear crystals or higher harmonic generation in gas jets [17]. The combination of direct frequency comb spectroscopy and ion trapping thus provides the possibility to perform spectroscopy on various ionic transitions in a single system.

In this Letter we demonstrate for the first time direct frequency comb spectroscopy of ions in a trap. It is shown that this technique can be applied using an upconverted frequency comb at 394 nm, without amplification of the comb pulses. Furthermore, we combine it with a shelving scheme [18] to suppress background signal from non-resonant comb modes, resulting in a very good signal to noise-ratio despite the low power per comb mode. Calcium ions are used for this experiment because a more accurate calibration of the $4s^2S_{1/2} - 4p^2P_{3/2}$ state is of interest for the search for a change of $\alpha$ over timespans of many billion years [13]. Apart from this application, trapping and laser cooling of the calcium ion has been widely studied [20, 21, 22], in particular for atomic clocks [23, 24] and quantum computation [25].

The spectroscopy is performed in a linear Paul trap, which is described in detail inFig. 1. The ion trap is mounted in a vacuum chamber, evacuated to a pressure of $3 \times 10^{-9}$ mbar. Calcium ions are produced by a two-step ionization process, involving the frequency comb laser. Evaporated calcium ions from an oven are first excited at 422 nm to the $4p^1P_1$ state by a frequency-doubled CW Ti:sapphire laser (Coherent 899), and subsequently ionized by the doubled frequency comb ($\lambda < 391$ nm). This comb is also used for the spectroscopy on the ions (see below). The total system is schematically depicted in Fig. 1.

In order to reduce the Doppler effect on the single-photon spectroscopy transition, laser cooling is applied on the $4s^2S_{1/2} - 4p^2P_{1/2}$ transition using a 397 nm diode laser (Toptica DL100). This laser is set at a detuning $\Delta f \approx 10$
MH from resonance, locked to a wavemeter (Atos model LM-007) within \( \approx 6 \) MHz. The relevant energy levels are shown in Fig. 2. The spatial mode of this laser is cleaned up in a single mode fiber to reduce background from scattered light. The remaining 0.3 mW is focused to a beam diameter of \( w_0 = 0.5 \) mm in the trap. Since the excited ions have a 7% chance to decay to the long-lived \( 3d^2D_{3/2} \) state, a repumper diode laser at 866 nm is used (\( P = 1 \) mW, Toptica DL100). Enough cooling power is available to crystalize the ion cloud, which reduces the Doppler broadening. The center of the trap is imaged onto a pinhole to remove background radiation and scattered light from the electrodes. Fluorescence from the \( 4s^2S_{1/2} \rightarrow 4p^2P_{3/2} \) transition is then observed using a photomultiplier tube (PMT, Philips XP2020Q). From this fluorescence we determined a lifetime for the ions in the trap of \( \tau \approx 8 \) minutes.

Direct frequency comb spectroscopy relies on the equivalence of phase coherent pulses (as emitted from a comb laser) and an equidistant mode structure in the frequency domain. These frequency comb modes can be described by the repetition frequency \( f_{\text{rep}} \) of the pulses and a carrier-envelope offset frequency \( f_{\text{ceo}} \). The frequency of the \( n \)th mode is equal to \( f_n = \pm f_{\text{ceo}} + n \times f_{\text{rep}} \). Both \( f_{\text{rep}} \) and \( f_{\text{ceo}} \) are radio frequencies, which are locked to a frequency standard (in our case a Stanford PRS10 Rubidium atomic clock, referenced to the Global Positioning System).

The frequency comb laser for the experiment is based on Ti:sapphire. Chirped mirrors are used in the laser for dispersion management, and are chosen such that the output is maximized at the desired wavelength of \( \lambda = 788 \) nm. This light is frequency doubled in a 3 mm BBO-crystal to obtain the right wavelength range to excite the \( 4s^2S_{1/2} \rightarrow 4p^2P_{3/2} \). By matching the angular dispersion induced by a grating to the wavelength derivative of the phase-matching angle \( [26] \), a wide bandwidth can be phase-matched. This method is used (Fig. 1 inset) to obtain a frequency comb in the blue with a FWHM of 13 nm and a power of \( P \approx 2 \) mW.

The calcium ions are excited by focusing the upconverted comb light in the trap to an elliptical beam with a major axis \((1/e^2 \) width\) of 0.8 mm, and a minor axis of 0.4 mm. The generated spectrum is sufficiently broad for both probing the \( 4s^2S_{1/2} \rightarrow 4p^2P_{3/2} \) transition and ionizing excited neutral calcium. However, only one comb mode is resonant at the time in the ions because a single-photon transition is probed. All other \( \approx 10^5 \) modes do not contribute to the signal, but do give background signal due to scattered photons. This signal-to-noise issue is overcome by employing a ‘shelving’ scheme (see e.g. [18]). The scheme we use is depicted schematically in Fig. 2. Ions that are excited to the \( 4p^2P_{3/2} \) state, have a 7% chance to decay to the \( 3d^2D_{5/2} \) state, which has a lifetime of \( 1.2 \) s [27]. Trapped in this state they can no longer contribute to the signal. Since the fluorescence from the cooling laser is monitored, less signal will be observed in this situation (Fig. 2b)). However, if a second repumper laser is used on the \( 3d^2D_{5/2} \rightarrow 4p^2P_{3/2} \) transition at \( \lambda = 854 \) nm, ions will be pumped back into the ground state and participate in the cooling cycle (Fig. 2a).

In order to scan over the transition, the frequency comb repetition rate is varied. A small change in this parameter effectively leads to a frequency scan of the comb near the resonance. All shown results are measured by scanning in both increasing and decreasing frequency direction, in order to eliminate systematic effects due to calcium ion loss.

**FIG. 2:** (Color online) Schematic view of the energy levels and the shelving principle. Arrows indicate laser beams, the wavy lines spontaneous fluorescence. The second repumper at 854 nm is periodically blocked using a chopper. If this repumper is present (situation (a)) all ions stay in the cooling cycle, a blocked repumper leads to transfer of the ions into the dark state, and the fluorescence from the cooling laser decreases (situation (b)).

**FIG. 3:** (Color online) Top: Measured fluorescence signal for the periods where the second repumper at 854 nm is on (red trace) and the periods where the repumper is off (black trace). The comb repetition frequency \( f_{\text{rep}} \) is scanned against time (upper axis) and frequency (lower axis). Bottom: Normalized fluorescence signal (corrected for decay, thin black trace) and the corresponding fit (thick red trace).
An important issue in frequency comb spectroscopy is the determination of the mode number $n_0$, usually based on previous measurements. The best known value for the $4s^2S_{1/2} \rightarrow 4p^2P_{3/2}$ transition has long been $\nu = 25 \, 414.40(15) \, \text{cm}^{-1}$, presented in [28], until a new value was reported of $\nu = 25 \, 414.4137 \, \text{cm}^{-1}$ [29]. Because no uncertainty was known for this measurement, it has been re-evaluated recently leading to a best value of $\nu = 25 \, 414.415(2) \, \text{cm}^{-1}$ [30]. The resulting $1\sigma$ accuracy of 60 MHz is insufficient to assign the mode number with confidence. We can, however, determine the mode by changing $f_{\text{rep}}$, which is graphically shown in Fig. 4. First an approximate mode number can be deduced from the earlier presented values, and the offset frequency $2 \times f_{\text{reo}}$ is taken into account (the factor two is due to the doubling of the comb). Using the selected mode number, one of the peak centers overlaps with the transition frequency, while the others deviate an integer times the repetition rate from this value. After the repetition frequency is changed by several MHz, the centers overlap at the peak with the correctly assigned mode number, but separate on neighbouring peaks (Fig. 4).

We have investigated several systematic effects on the transition frequency. The strongest is due to the always present AC Stark shift. The power dependence of this shift was measured to be $\Delta f_{\text{trans}} = 9.7(1.3) \, \text{kHz}/\mu \text{W} \times \nu_{\text{cooling}}$, where $\nu_{\text{cooling}}$ is the cooling laser power in MHz. The corresponding zero-crossing is $f_{\text{trans}} = 761 \, 905 \, 013.06(0.21) \, \text{MHz}$. In addition, at $\nu_{\text{cooling}} = 200(10) \, \text{MHz}$, the dependence of the measured transition frequency on the frequency comb laser power was measured to be $\Delta f_{\text{trans}} = 0.19(0.24) \, \text{MHz}/\mu \text{W} \times f_{\text{comb}}$.
where $P_{\text{comb}}$ is the comb laser power in mW (Fig. 5 (b)). The transition frequency is corrected for the corresponding shift at $P_{\text{comb}} = 1.9$ mW of $\Delta f_{\text{trans}} = 0.36(0.46)$ MHz. The two repumper lasers do not cause a significant light shift, since the 866 nm laser does not couple to either one of the levels involved in the measured transition, and the 854 nm repumper is not present during the shelving intervals. The recoil shift is calculated to be 32 kHz, which is small compared to the measurement accuracy. Stark shifts due to the trapping fields are estimated to be below 4 kHz. Since the comb laser beam is linearly polarized, there is no first order Zeeman shift, while higher order Zeeman shifts are negligible compared to the measurement accuracy (see [2]). This leaves the statistical uncertainty due to the comb laser to consider. For a measurement time per data point of 0.1 s, the Allan deviation is estimated to be $10^{-9}$. Since a typical scan consists of about 3000 points measured over 10 minutes, this deviation averages down further and does not contribute significantly to the uncertainty budget. Once corrected for the measured shifts, the unperturbed transition frequency of the $4s^2S_{1/2} - 4p^2P_{3/2}$ transition in $^{40}\text{Ca}^+$ is found to be $f = 761\,905\,012.7(0.5)$ MHz ($1\sigma$ uncertainty). This result is consistent with the previous most accurate result of $f = 761\,904\,994(60)$ MHz [30].

In conclusion, for the first time direct frequency comb spectroscopy on ions in a linear Paul trap has been demonstrated, using an upconverted comb laser and shelving detection. The obtained level of accuracy on the $4s^2S_{1/2} - 4p^2P_{3/2}$ transition in calcium ions is more than two orders of magnitude better than previous calibrations. The applicability of this method extends well beyond the probed ion and transition used for this experiment, if direct frequency comb spectroscopy were used on sympathetically cooled ions in a Paul trap.

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