Direct frequency-comb spectroscopy of a dipole-forbidden clock transition in trapped $^{40}Ca^{+}$ ions

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We demonstrate direct frequency-comb (FC) spectroscopy of the dipole-forbidden $4s^2S_{1/2}$–$3d^2D_{5/2}$ transition in trapped $^{40}Ca^{+}$ ions using an unamplified FC laser. The excitation is detected with nearly 100% efficiency using a shelving scheme in combination with single-ion imaging. The method demonstrated here has the potential to reach hertz-level accuracy, if a hertz-level linewidth FC is used in combination with confinement in the Lamb–Dicke regime. © 2010 Optical Society of America

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Optical frequency combs (FCs) provide a direct link between optical and microwave frequencies [1,2]. This link enables carrying the accuracy of a microwave frequency standard to the optical domain, or transferring the extreme accuracy achieved in optical spectroscopy (currently reaching below the 10^{-17} level [3]) to any other part of the optical spectrum and to the microwave domain. In addition, the development of broadband titanium-doped sapphire FC lasers with a wavelength span of hundreds of nanometers [4,5] makes high-accuracy frequency calibration possible over a very wide wavelength range. These FCs can be employed to calibrate an additional laser that is used for excitation, but it is also possible to directly use the light of the comb modes to perform the metrology [6–9]. This technique of direct FC spectroscopy (DFCS) is advantageous in that the full comb spectrum is available for excitation, without the need for an additional probe laser.

Trapped laser-cooled ions provide an ideal system for DFCS, as they allow for long interaction times. Furthermore, different ionic species can be trapped simultaneously and be sympathetically cooled and detected by the laser-cooled ions [10,11]. The application of DFCS to trapped ions [9] thus facilitates spectroscopy on various ionic transitions in a single measurement setup. To our knowledge, the possibility of DFCS of trapped ions has been demonstrated only for strong resonance lines. The range of applicability of this method would be greatly extended if also weak and narrow transitions could be detected. For example, such a "clock" transition could be used as an *in situ* frequency reference for the calibration of other transitions in ions or as a probe for external fields in the ion trap. The concept of DFCS on narrow transitions has been investigated for cold neutral calcium atoms on a transition with a linewidth of 374 Hz, using the amplified modes of an FC [12]. In this Letter, we show that DFCS can be performed on significantly weaker transitions, using an ion trap and even without amplification of the FC laser. This is demonstrated on a laser-cooled crystal of calcium ions, in which the dipole-forbidden $4s^2S_{1/2}$–$3d^2D_{5/2}$ transition at 729 nm (natural linewidth 0.14 Hz) is induced with DFCS. On this transition, one can potentially reach hertz-level accuracy, as was shown previously by Chwalla *et al.* using cw laser excitation [13].

In our experiment, calcium ions are produced by two-photon excitation involving a frequency-doubled portion of the FC [9]. The created ions are trapped in a segmented linear Paul trap (Fig. 1), which consists of four cylindrical molybdenum electrodes with a radial spacing of $2 \times r_0 = 7$ mm. Two of the rods are segmented into five pieces of 12 mm length each, to which different dc voltages can be applied. These potentials serve to confine the ions to the trap axis and compensate for possible stray electric fields. The end caps of the trap are formed by the electrodes adjacent to the central region; hence, the spacing between the end caps is $z = 12$ mm. A voltage of $V_{ec} = 10$ V is applied to these electrodes to provide axial confinement.

The other two electrodes are unsegmented cylindrical rods of 60 mm length. To these electrodes, an rf voltage is applied, with frequency $\Omega = 2\pi \times 2.6$ MHz and amplitude $V_{RF} = 150$ V. This corresponds to a Mathieu stability parameter $q_r = 0.2$ and a radial secular trapping

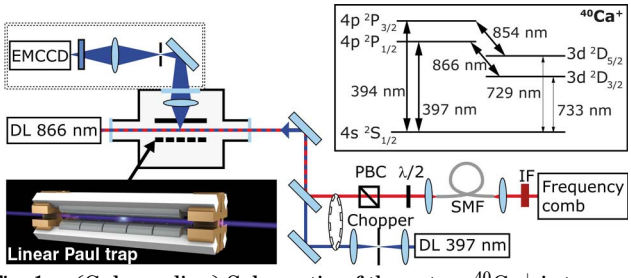

Fig. 1. (Color online) Schematic of the setup: $^{40}Ca^{+}$ is trapped in a linear Paul trap and Doppler cooled using two diode lasers (DLs). Fluorescence at 397 nm due to the cooling laser is spatially filtered and detected by an EMCCD camera. A simplified energy level scheme for $^{40}Ca^{+}$ is shown in the inset. IF, interference filter; $\lambda/2$, half-wave plate; PBC, polarizing beam splitter cube; SMF, single-mode fiber.

frequency of $2\pi \times 202$ kHz. The pressure inside the vacuum chamber is 10^{-11} mbar, for which the lifetime of a string of confined ions is larger than 8 h.

The trapped $^{40}Ca^{+}$ ions are Doppler cooled on the $4s^2S_{1/2}$–$4p^2P_{1/2}$ transition, using a grating-stabilized diode laser at 397 nm (Toptica DL100, $P = 0.1$ mW after spatial filtering) in combination with a repumper diode laser at 866 nm (Toptica DL100, $P = 0.8$ mW) [14]. Fluorescence photons emitted by the ions are imaged onto an electron multiplying CCD camera (EMCCD, Andor iXon^{Em}+), with a telescope comprising one lens inside the vacuum with an effective focal length EFL = 25 mm, and a second lens outside the vacuum with a focal length of $f = 100$ mm. To reduce stray light, the beam is spatially filtered at the intermediate focus in between the two lenses.

The $4s^2S_{1/2}$–$3d^2D_{5/2}$ transition is excited with light near $\lambda = 729.5$ nm from an unamplified FC. Both the carrier-envelope offset frequency (f_{ceo}) and the repetition frequency (f_{rep}) of the FC are locked to a Stanford PRS10 rubidium atomic clock, which is referenced to the Global Positioning System (GPS). The FC has a power of $P = 0.12(2)$ μW per mode at the transition frequency, focused to a spot size of $w_0 = 0.17$ mm. A 60 nm bandwidth interference filter with 99% transmission at 729.5 nm and an optical density of 7 in the wavelength range of 760–870 nm is used in this beam to suppress excitation of the dipole allowed $3d^2D_{5/2}$–$4p^2P_{3/2}$ and $3d^2D_{3/2}$–$4p^2P_{3/2}$ transitions. To avoid an ac-Stark shift by the 397 nm laser, the cooling and comb laser are alternately chopped mechanically at a frequency of 59 Hz.

The fluorescence of the ions due to the cooling laser is monitored with the EMCCD camera at a speed of 1 Hz. Excitation of the clock transition is detected by observing the disappearance of fluorescence from the cooling laser throughout the lifetime of the $D_{5/2}$ level. This shelving method yields near 100% detection probability, enabling us to detect the transition despite a very low excitation rate of ~7 per 1000 s per ion, over a background count rate of ~0.9 per 1000 s per ion.

Typical images as recorded in this process are shown in Figs. 2(a) and 2(b). Integration of the signal over several image rows results in the traces shown below the images, where the distinction between dark and bright ions is made based on a threshold at 50% of the peak fluorescence intensity. The count rate is limited to 20 counts/100 s for a string of 10 ions, to avoid ion heating and count ambiguity. An example of the detection of dark ions on resonance in 100 s is shown in Fig. 2(c). To record the transition, the FC is scanned over the transition in frequency steps of $\Delta f = 1.2$ MHz every 100 s, resulting in a total time of 30 min per scan. The corresponding measured signal is shown in Fig. 3 (the error bars indicate a $\sqrt{N}$ count uncertainty and are based on the underlying Poissonian statistics of the ion excitation process [15]).

There are several contributions to the current frequency uncertainty budget, all of which have the prospect to be reduced to the hertz level. The largest of these is caused by a residual time overlap between the cooling laser and the FC excitation period, resulting in an ac-Stark shift of $\Delta f = 0.15(0.10)$ MHz. The repumper also causes a small shift of $\Delta f = -0.01(0.01)$ MHz. Other systematic

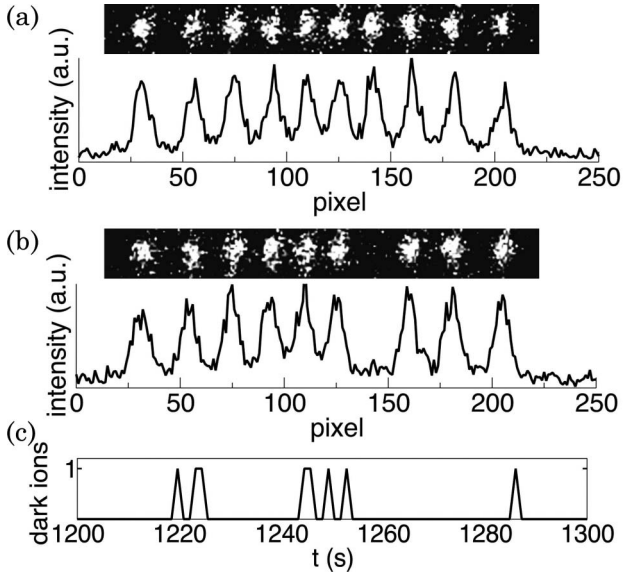

Fig. 2. Detection method: (a) (all bright ions) and (b) (one dark ion) show the camera image of a string of ions. The distance between the ions is ~15 μm. Integration over ~15 lines of the camera images results in the traces as shown underneath the images. The detection of dark ions close to resonance in 100 s is shown in (c).

effects we investigated (ac-Stark effect due to the FC, the Zeeman effect in combination with possible optical pumping by the cooling laser, the ac- and dc-Stark effect due to the trap electric field and comb accuracy) give a total contribution of <8 kHz. Correcting for all systematic shifts, we find a transition frequency 411042129.6(0.3) MHz, consistent with the value reported in [13].

In the current experiment, an FC with a 1.6 MHz FWHM mode linewidth was used. The measured linewidth of 5.5 MHz FWHM is mainly limited by Doppler broadening, and corresponds to $T = 14$ mK. To improve on the presented results, Doppler broadening may be

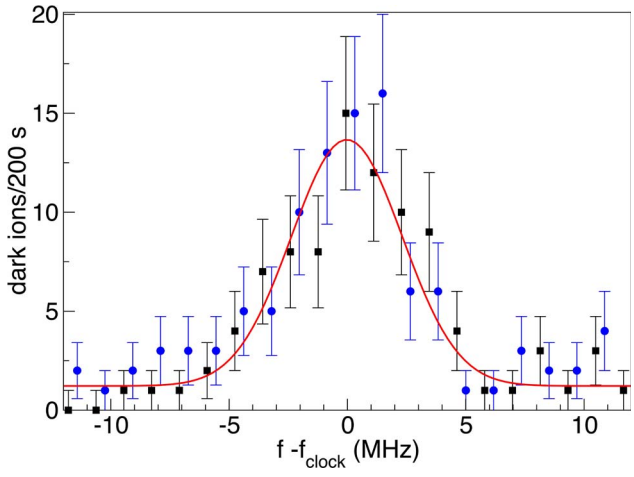

Fig. 3. (Color online) Graph of the resonance line, relative to the literature value of $f_{clock} = 411042129.776$ MHz [13,16], consisting of two measurement sets (one indicated by circles, the other by squares). For each set, the data is collected by scanning the FC over the resonance from both sides. The solid curve shows a Gaussian fit through all the data points.

removed by increasing the confinement of the ions by the trap such that the Lamb–Dicke regime is attained [17]. In addition, hertz-level FC linewidths may be achieved by locking to a high-finesse cavity [5]. On the assumption of an instrument linewidth of 23 Hz, as in [13], combined with such an FC, a 10^5 fold improved excitation rate can be obtained. Lowering the FC intensity by a factor of 10^2 then reduces the ac-Stark shift to <1.5 Hz, with still 10^3 times increased excitation rate. The higher excitation rate requires a repumper on the $3d^2D_{5/2}$–$3p^2P_{3/2}$ transition, for which light is available in the FC. With the aforementioned linewidths, in combination with a Poisson-limited noise, and a count rate of 5 counts/s, theoretically, a stability of $\sigma(\tau) = 10^{-13}/\sqrt{\tau}$ could be reached. Under these conditions, DFCS as demonstrated here could provide a relatively simple method to operate the combination of an ion trap and an FC as an optical frequency standard, with both stability and accuracy surpassing that of GPS-disciplined Rb standards by orders of magnitude.

The theory on further experimental requirements are described in [17] and shown to be experimentally achievable by [13].

In conclusion, we have demonstrated the feasibility of DFCS of a dipole-forbidden (clock) transition in an ion trap. The measured transition frequency is consistent with previous measurements. The combination of long ion storage times with single-ion fluorescence imaging and detection by electron shelving yields a very high sensitivity. We also point out that, with the use of an FC with narrow linewidth and ion confinement in the Lamb–Dicke regime, DFCS as demonstrated here has the potential to reach a hertz-level accuracy. In addition, the technique can be used to perform spectroscopy on other transitions in Ca^+ or on sympathetically cooled ions, with direct calibration to the calcium clock transition.

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