

Quantum Logic for Precision Spectroscopy

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1 Introduction

Our current understanding of physics is based on the standard model of particle physics. Although this model has proven to be valid in many aspects, it is widely believed that it will be replaced by a more complete theory. Such a theory would need to be a quantum theory unifying all four fundamental forces. Some of the candidates for such a “theory of everything” allow for a variation of fundamental constants. The idea of changing physical constants was first conceived by Dirac who related in his “Large Number Hypothesis” the ratio between the strength of electro-magnetic and gravitational interaction with the age of the universe. As the universe continues to age, one of the fundamental constants describing electromagnetism and gravitation must change as well, according to the hypothesis. In the meantime many extensions and variations of this hypothesis have been postulated. Observing such a temporal variation of fundamental constants would revolutionize our current understanding of physics. Even restricting the rate for a possible variation to smaller and smaller values could aid to test predictions of theories beyond the standard model. It is convenient to consider only dimensionless constants, since one would otherwise not be able to distinguish between a change in its value or its dimension. Possible experimentally accessible candidates for a time variation (among others) are the fine-structure constant α , describing the strength of electro-magnetic interactions, and the electron-to-proton mass ratio m_e/m_p [1].

In 2001 the discussion about a change in the fine-structure constant was spurred by a publication of Webb and coworkers [2] who claimed to have observed a change in α on cosmological timescales. For their analysis, they recorded spectra of light emitted by quasars. The spectra show absorption lines of atoms and molecules found in interstellar clouds that have been imprinted onto the light as it passed through these clouds on its way to earth. A comparison with present-day laboratory spectra indicated that around 10^{10} years ago, α may have had a value

$(5.4 \pm 1.2) \cdot 10^{-6}$ smaller than today. Similar investigations by other collaborations could not confirm this result [3, 4], leaving the outcome of the dispute open. Currently, one of the limitations of the analysis is the precision with which laboratory spectra are known [5]. An approach to study the possible variation of fundamental constants on laboratory time scales consists of a long-term highly accurate frequency comparison of atomic or molecular transitions that depend differently on a change in the constant. An example is the recently performed comparison of the single-ion mercury and aluminum optical clocks [6]. Over the course of almost a year, frequency ratio measurements between the clock transitions of these two ions have been performed that yielded an upper bound for a relative change in α of $(-1.6 \pm 2.3) \cdot 10^{-17}/\text{year}$.

In the following, we will describe two projects that are currently being set up at the QUEST Institute for Experimental Quantum Metrology with the goal of helping to solve the question of a possible time variation of fundamental constants.

2 Quantum logic and precision spectroscopy

Common to both projects is the use of quantum logic techniques developed for quantum information processing with trapped ions. The idea is to use a string of trapped ions in a linear Paul trap that are strongly coupled via their mutual Coulomb repulsion. The external motion of the ions is most conveniently described in a normal-mode picture, involving motion of all ions. Using laser pulses of appropriate frequency and duration, it is possible to change not only the internal electronic state of the ions, but also their motional state in the trap. Cirac and Zoller proposed to use such laser pulses to entangle ions and perform quantum logic gates using the normal modes in the trap as a “quantum bus” [7].

Shortly after this seminal idea, a controlled-NOT (C-NOT) gate between a trapped atomic ion and its motional mode in the trap was de-

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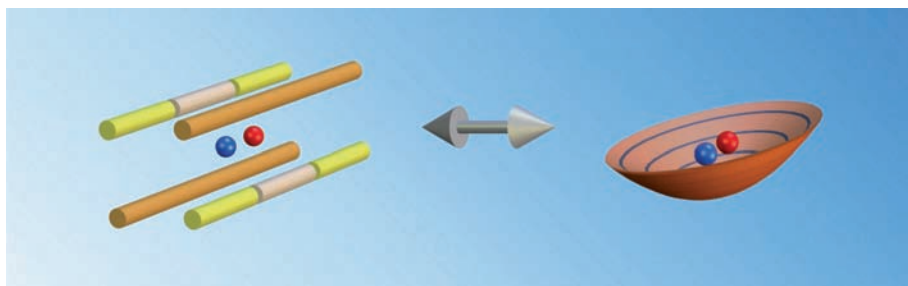


Figure 1:
Two ions trapped in a linear Paul trap. Left: Ions are trapped via radio-frequency (orange) and DC (yellow and grey) fields applied to electrodes. Right: The resulting potential is harmonic in three dimensions. The motion of the ions is strongly coupled via the Coulomb repulsion.

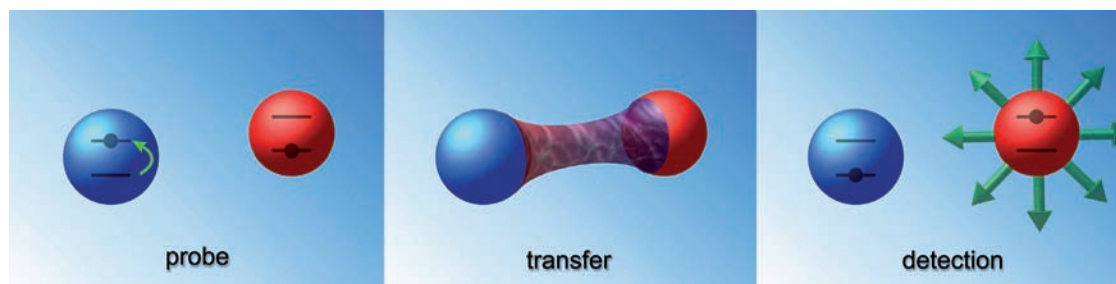
monstrated in a pioneering experiment [8]. After this demonstration of exquisite coherent control over the internal and external degrees of freedom in an atomic system, quantum information processing with trapped ions has flourished and impressive results, such as teleportation, error-correction and the implementation of the Deutsch-Jozsa and semi-classical Fourier transformation algorithms have been achieved. Current state of the art is the manipulation of up to eight qubits [9, 10] and two-ion gate fidelities exceeding 99.3% [11]. These accomplishments were only possible through efficient decoupling of the internal and external atomic degrees of freedom from the decoherence-inducing environment and manipulation of the internal states with highly coherent light fields. The same requirements hold true for precision spectroscopy,

proposal, a spectroscopy ion is trapped simultaneously with a logic ion in the same potential well of a linear Paul trap (see Figure 1).

Owing to the strong motional coupling between ions, laser cooling of the logic ion sympathetically cools the spectroscopy ion. Using quantum logic protocols, the internal state information of the spectroscopy ion after probing the transition can be faithfully transferred to the logic ion, where it is detected with near unit efficiency (Figure 2).

Furthermore, the internal state of a spectroscopy ion within the spectroscopy manifold of states can be deterministically prepared using this technique [14]. Therefore, quantum logic techniques allow us to fulfill all requirements to perform precision spectroscopy on ions that would otherwise be inaccessible due to the lack

Figure 2:
Schematic description of quantum logic spectroscopy. Left: The transition of the spectroscopy ion (blue) is probed with a laser. Middle: The state information is mapped to the logic ion (red). Right: Efficient state discrimination using electron shelving on the logic ion.



where a frequency measurement of an unspoiled atomic or molecular transition is performed. In fact, the two fields have a lot in common and already a number of quantum logic techniques beneficial for precision spectroscopy have been proposed [12, 13] and demonstrated [14, 15].

Precision spectroscopy is typically performed by laser cooling of an atomic sample to reduce Doppler shifts and increase interrogation times. Then, the transition of interest is probed with a frequency stabilized laser. The excitation probability is detected via either direct fluorescence or the electron shelving technique involving a third atomic level. In both cases a cycling transition is required to collect a sufficient number of photons for state discrimination. This severely restricts the number of accessible species for high precision spectroscopy. Dave Wineland suggested in 2001 the use of quantum logic techniques to overcome this limitation [12]. In his

of a cycling transition. This spectroscopy technique is particularly promising for ions with cycling transitions in the vacuum ultraviolet spectral range where no laser source is currently available, and for atomic and molecular ions with a complex internal level structure with many possible decay channels from the excited state. The described protocol works best for excited states with a long lifetime, such that spontaneous decay during the state transfer step is negligible. In a variation of the scheme, quantum logic spectroscopy can also be performed on transitions with a short excited state lifetime. Possible applications of this new technique are manifold. It opens up the possibility for unprecedented high resolution spectroscopy on a wide range of sympathetically cooled atomic and molecular species with special spectroscopic properties, such as an improved sensitivity to a variation of fundamental physics constants [16, 17] or atomic clocks [14].

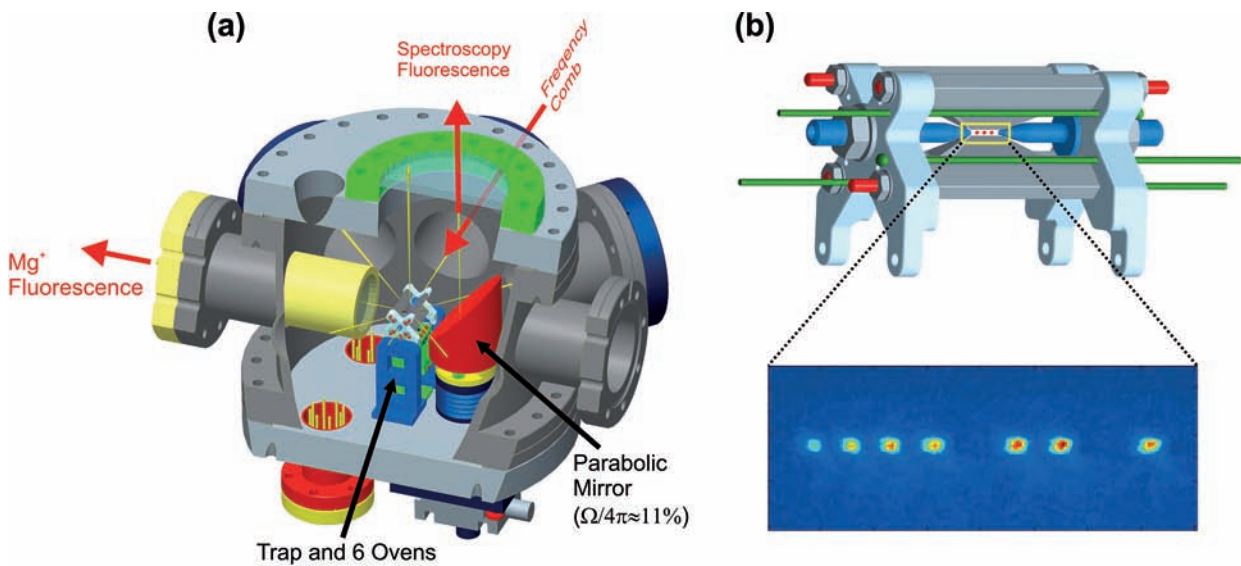
3 Direct frequency comb spectroscopy using quantum logic

Many atomic and molecular species with interesting spectroscopic features have a very complex level structure. As described above, quantum logic techniques can be employed to prepare and probe the ion within the spectroscopy manifold. Nevertheless, if too many levels are involved, initial state preparation can be challenging. Conventional spectroscopy typically circumvents this issue by probing very many atoms in a beam or gas cell, thus severely compromising the accuracy of the measurement due to level shifting and broadening effects. Another solution is the use of optical pumping to repump the atom from metastable states back into the

spectral output of the comb. Direct frequency comb spectroscopy has been implemented in a variety of flavors: (i) a single comb tooth interacts with a single transition [19]; (ii) the comb as a whole is used to drive two-photon transitions, possibly enhanced by near-resonant coupling to an intermediate level [20, 21]; and (iii) the comb is used as a broadband source that simultaneously interacts with many transitions in the atomic or molecular species [22].

We are currently setting up an experiment in which we plan to perform direct frequency comb spectroscopy on metal ions, such as Ca^+ , Ti^+ and Fe^+ , using quantum logic for state preparation and detection. Depending on the ion species, we will either use the comb to drive two-photon

Figure 3:
Experimental setup.
(a) Construction drawing of the vacuum chamber.
(b) Detail of the ion trap and a string of magnesium ions observed in the trap (false-color image).



spectroscopy manifold. For this, light at many different colors is needed to access all transitions. One such light source with a very large spectral bandwidth is an optical frequency comb [18]. Its invention by T.W. Hänsch and J.L. Hall has been awarded with the Nobel Prize in Physics in 2005. Today, two major frequency comb technologies have been established: Ti:Sa-based systems with repetition rates ranging from 80 MHz to up to 10 GHz and a spectral emission between 600 and 1200 nm, and fiber-laser based systems with repetition rates ranging from 100 to 200 MHz and a spectral emission between 900 nm and 2 μm . External frequency doubling and tripling in nonlinear crystals, and high-harmonic generation in gas jets allows to shift the output of the lasers to different spectral regions. The evenly spaced spectral comb emitted by these systems is phase and frequency stabilized to provide a fixed ruler in frequency space, acting as an array of thousands of phase-locked weak continuous-wave lasers. This makes frequency combs not only ideal instruments to count optical frequencies [18], but also to directly perform precision spectroscopy with the broad

transitions (Ca^+), or use it as a broadband source of light, interacting with many transitions simultaneously (Ti^+ , Fe^+). Optical transitions in these ions are used in the analysis of quasar absorption spectra and are currently known only to an accuracy ranging between a few 10 MHz to a few GHz [23]. To improve the analysis for a possible time variation of fundamental constants, accuracies on the order of 1 MHz are required [5].

In our experiment, we plan to trap a single spectroscopy ion together with the logic ion Mg^+ in a linear Paul trap inside a vacuum chamber. Light from the optical frequency comb is applied along the symmetry axis of the trap (see Figure 3).

Our first candidate is Ca^+ , for which accurate data on the $^2\text{S}_{1/2} \leftrightarrow ^2\text{P}_{3/2}$ transition is missing [24]. Spectroscopy is performed in several steps (see Figure 5): (i) the ion crystal is cooled to the ground state of axial motion via Raman sideband cooling on Mg^+ ; (ii) the optical frequency comb is adjusted in its frequency and applied for a few hundred milliseconds to the spectroscopy ion; and (iii) scattering of photons heats

the ion crystal to a higher vibrational quantum number, which can be detected with near unit efficiency on the Mg^+ ion. Performing these steps in a sequence and adjusting the frequency comb spacing or offset allows the determination of the global electronic level scheme of the atomic system [21] by comparison with a theoretical model [see Fig. 5(c)].

A similar method will be used to perform spectroscopy on Ti^+ and Fe^+ . In principle, it should be possible to use the same versatile apparatus to investigate a wide variety of spectroscopy ions, provided the spectral range of the frequency comb covers all involved atomic transitions. We will collaborate with other QUEST

4 A single-ion quantum logic optical clock

Another application of quantum logic spectroscopy is the development of next-generation optical clocks [26]. Up to now, atomic species considered for optical clocks had to fulfill several criteria: they needed to provide a proper transition for laser cooling and detection, and a narrow-linewidth clock transition with high resilience to external field perturbations. With quantum logic spectroscopy, the only requirement is a suitable clock transition, since all other steps will be performed via the logic ion. This has been successfully demonstrated with aluminum and beryllium as the clock and logic ion species [14].

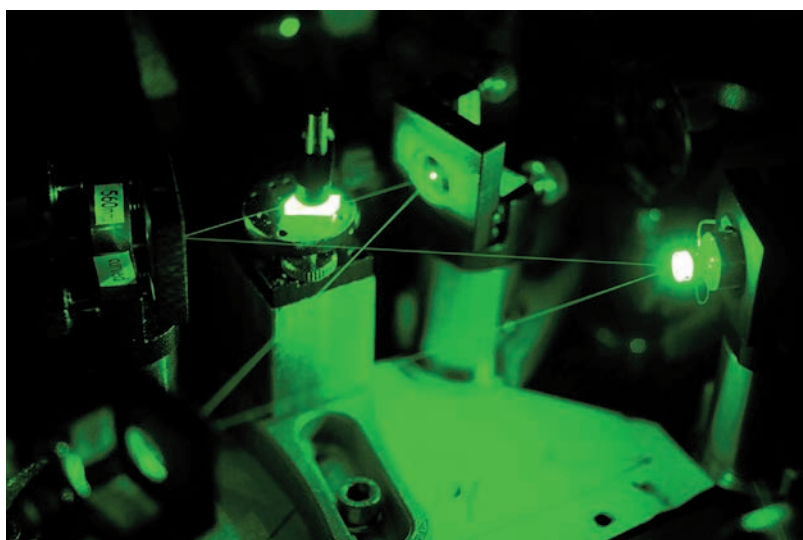


Figure 4:
Detail of the experimental setup. Photo of the last frequency-doubling stage to generate the ultraviolet light used for cooling and detecting magnesium ions. Four mirrors form a bow-tie resonator for the pump light at 560 nm which is converted to 280 nm in a non-linear crystal. Source: Universität Innsbruck.

partners (U. Morgner and LZH) to extend optical frequency combs into the near ultra-violet spectral regime, relevant to many atomic ion species.

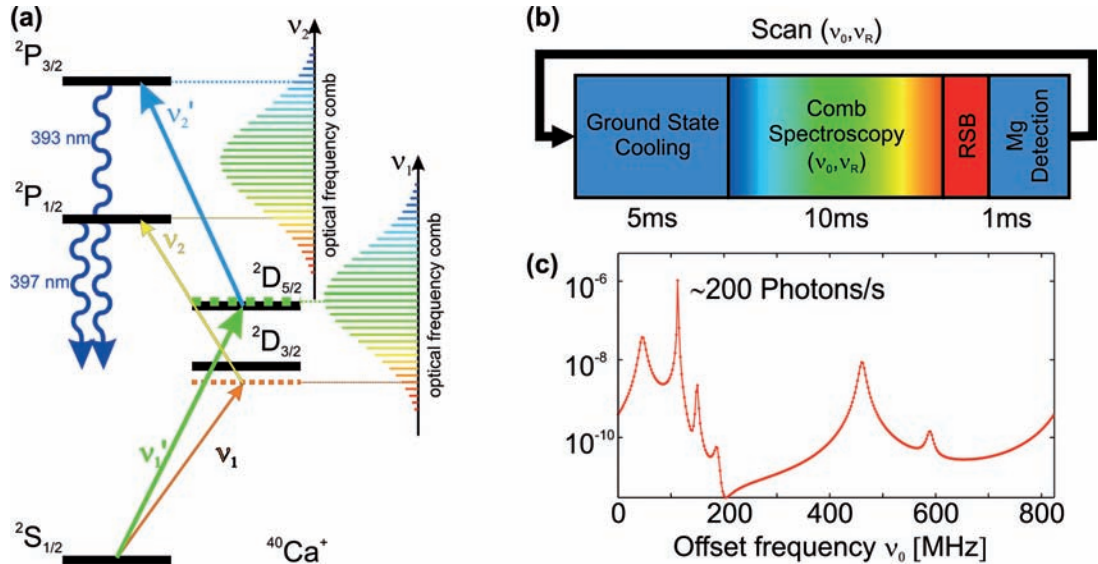
The quantum logic techniques described above can be employed not only for precision spectroscopy, but also to cool the rotational states of a molecule [25]. Having ground-state cooled molecules at our disposal allows us to perform a large variety of fascinating experiments. One possibility is to conduct precision measurements of various electronic and rovibrational transitions using the direct frequency comb spectroscopy techniques developed above.

This would be particularly interesting for the astrophysically important CaH^+ molecule, for which no experimental data is available. We will also explore the possibility of implementing our scheme with other small molecules including H_2^+ and HD^+ , which are relevant for m_e/m_p measurements.

In the meantime, the aluminum ion optical clock has advanced to being one of the most accurate clocks in the world [6]. The limitations in this experiment are frequency shifts due to micro- and secular motion of the ion in the trap. Micromotion is most likely introduced by charging of the electrodes by ultraviolet laser photons. Heating of the ion crystal during interrogation is responsible for a systematic uncertainty in the amplitude of the secular motion and therefore the second-order Doppler shift. Both effects can be significantly reduced in a new setup, featuring a trap with lower heating rates of the secular motion and different electrode material, less susceptible to the photo effect. We plan to set up such an experiment with aluminum as the clock ion species and a new trap design to overcome the limitations described above. Having a clock with such a high accuracy available at PTB will pave the way not only to establish the single-ion aluminum clock as a secondary frequency standard, but also to perform precision measurements with unprecedented resolution. One example for such an experiment would be to compare the aluminum ion frequency standard to other

Figure 5: Direct Frequency Comb Spectroscopy of Ca^+ .

(a) Partial level scheme of Ca^+ and excitation by optical frequency comb. (b) Quantum logic spectroscopy sequence. (c) Density matrix simulation of the number of scattered photons on the $P_{1/2} - S_{1/2}$ transition as a function of the frequency comb's offset frequency, scanned over one repetition rate (824 MHz).



frequency standards at PTB over the course of many years and derive an upper limit for a temporal change in the fine-structure constant on a laboratory time scale.

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References

- [1] See "More Accurate Clocks – What are They Needed for?" in this issue, p. 16.
- [2] *J. Webb et al.*: Further evidence for cosmological evolution of the fine structure constant. *Phys. Rev. Lett.* 87, 091301 (2001).
- [3] *R. Srianand, H. Chand, P. Petitjean, B. Aracil*: Limits on the time variation of the electromagnetic fine-structure constant in the low energy limit from absorption lines in the spectra of distant Quasars. *Phys. Rev. Lett.* 92, 121302 (2004).
- [4] *R. Quast, D. Reimers, S. A. Levshakov*: Probing the variability of the fine-structure constant with the VLT/UVES. *Astron. Astrophys.* 415, L7 (2004).
- [5] *J. C. Berengut et al.*: Laboratory spectroscopy and the search for space-time variation of the fine structure constant using QSO spectra. arXiv: physics/0408017 (2004).

- [6] *T. Rosenband et al.*: Frequency Ratio of Al^+ and Hg^+ single-ion optical clocks; metrology at the 17th decimal place. *Science* 319, 1808 (2008).
- [7] *J. I. Cirac and P. Zoller*: Quantum computations with cold trapped ions. *Phys. Rev. Lett.* 74, 4091 (1995).
- [8] *C. Monroe, D. M. Meekhof, B. E. King, W. M. Itano, D. J. Wineland*: Demonstration of a fundamental quantum logic gate. *Phys. Rev. Lett.* 75, 4714 (1995).
- [9] *H. Häffner et al.*: Scalable multiparticle entanglement of trapped ions. *Nature* 438, 643 (2005).
- [10] *D. Leibfried et al.*: Creation of a six-atom 'Schrödinger cat' state. *Nature* 438, 639 (2005).
- [11] *J. Benhelm, G. Kirchmair, U. Rapol, T. Körber, C. F. Roos, R. Blatt*: Measurement of the hyperfine structure of the $S_{1/2} - D_{5/2}$ transition in $^{43}\text{Ca}^+$. *Phys. Rev. A* 75, 032506 (2007).
- [12] *D. Wineland, J. Bergquist, J. Bollinger, R. Drullinger, W. Itano*: Quantum computers and atomic clocks. In: Proc. 6th Symposium on Frequency Standards and Metrology (St. Andrews, Scotland, 2001), ed. P. Gill (World Scientific, Singapore, 2001), p. 361.
- [13] *C. F. Roos*: Precision frequency measurements with entangled states. arXiv:quant-ph/0508148 (2005).
- [14] *P. O. Schmidt, T. Rosenband, C. Langer, W. M. Itano, J. C. Bergquist, D. J. Wineland*: Spectroscopy using quantum logic. *Science* 309, 749 (2005).
- [15] *C. F. Roos, M. Chwalla, K. Kim, M. Riebe, R. Blatt*: 'Designer atoms' for quantum metrology. *Nature* 443, 316 (2006).
- [16] *J.-P. Uzan*: The fundamental constants and their variation: observational and theoretical status. *Rev. Mod. Phys.* 75, 403 (2003).

- [17] *S. G. Karshenboim*: Fundamental physical constants: looking from different angles. arXiv: physics/ 0506173 (2005).
- [18] *T. Udem, R. Holzwarth, T. W. Hänsch*: Optical frequency metrology. *Nature* 416, 233 (2002).
- [19] *V. Gerginov, C. E. Tanner, S. A. Diddams, A. Bartels, L. Hollberg*: High-resolution spectroscopy with a femtosecond laser frequency comb. *Opt. Lett.* 30, 1734 (2005).
- [20] *A. Marian, M. C. Stowe, J. R. Lawall, D. Felinto, J. Ye*: United time-frequency spectroscopy for dynamics and global structure, *Science* 306, 2063 (2004).
- [21] *A. Marian, M. C. Stowe, D. Felinto, J. Ye*: Direct frequency comb measurements of absolute optical frequencies and population transfer dynamics. *Phys. Rev. Lett.* 95, 023001 (2005).
- [22] *M. Thorpe, J. Ye*: Cavity-enhanced direct frequency comb spectroscopy. *Appl. Phys. B* 91, 397 (2008).
- [23] *D. C. Morton*: Atomic data for resonance absorption lines. III. Wavelengths longward of the Lyman limit for the elements Hydrogen to Gallium. *Astrophys. J. Suppl. Ser.* 149, 205 (2003).
- [24] *A. L. Wolf, S. A. van den Berg, C. Gohle, E. J. Salumbides, W. Ubachs, K. S. E. Eikema*: Frequency metrology on the $4s\ ^2S_{1/2}-4p\ ^2P_{1/2}$ transition in $^{40}\text{Ca}^+$ for a comparison with Quasar data, *Phys. Rev. A* 78, 032511 (2008).
- [25] *P. O. Schmidt et al.*: Spectroscopy of atomic and molecular ions using quantum logic. In: *Non-Neutral Plasma Physics VI: Workshop on Non-Neutral Plasmas 2006*, eds. M. Drewsen, U. Uggerhoj, H. Knudsen, vol. 862 (AIP, 2006), p. 305.
- [26] See "The Development of Accurate Optical Clocks" in this issue, p. 25.