Publishable Summary for 17FUN07 CC4C
Coulomb Crystals for Clocks

Overview

Optical clocks and frequency standards are the most precise measurement devices available today. However, further improvements were needed to extend their use in applications in fundamental metrology. This project investigated laser-cooled trapped ions as a reference for a next generation of optical clocks of highest accuracy. While most precise optical clocks with trapped ions have been based on single ions, this project investigated ensembles of up to hundreds of ions in a Coulomb-coupled solid-like state, known as Coulomb crystals (CCs). This multi-ion approach provides higher signal-to-noise for clocks of improved stability and enabled the investigation of small frequency shifts caused by collisions or interactions. Different combinations of clock and coolant ions were studied and have opened the opportunity to perform precision measurements on previously inaccessible systems like highly charged argon ions and thorium ions which possess an optical nuclear transition. The main results were the development and implementation of loading and cooling methods for a range of different ions including the radioisotope ^{229}Th. Two-ion, two-species clock operation has been demonstrated and precision frequency determinations have been performed for sympathetically cooled $^{115}In^{+}$ and $^{40}Ar^{13+}$, where the result on the latter represents a breakthrough in precision measurements with highly charged ions.

Need

Within the SI system of units the realisation of the unit of time is in a pivotal position, as the unit second is included in the definition of six of the seven base units via the defining constants. Progress in research on optical clocks continues at a rapid pace towards lower uncertainties, now evaluated in the 10^{-19} range. New reference systems with specific advantages in terms of accuracy or stability required research on new experimental methods as well as on relevant atomic, molecular and nuclear data.

The control and understanding of the structure and dynamics of laser-cooled two species Coulomb crystals, which have not been studied in detail until now, is essential for improved optical clocks and frequency standards, and needed for the optimisation of sympathetic cooling and spectroscopy. Sympathetic cooling, in which one ion species is laser-cooled and the other ion species is cooled via the Coulomb interaction, allows the study of a wider range of relevant ions for optical clocks. However, it introduces additional Coulomb interaction between ions and with the time-dependent electric field of the ion trap, and required further investigation concerning frequency shifts induced by these interactions.

Existing methods of trap loading in optical clocks have been optimised for singly-charged species and based on evaporation or laser ablation, combined with electron impact or photoionisation. The investigation of the radioactive ^{229}Th isotope, with a half-life of 7920 years, required efficient loading methods for Th^{3+} and higher charge states in order to operate a nuclear optical clock with minimum source activity.

Ion clocks are affected by collisions with background atoms and molecules, resulting in a number of effects from frequency shifts, excitation or quenching of metastable levels to ion loss through charge exchange or chemical reactions. In order to reliably exclude or estimate systematic shifts in the low 10^{-18} level, it was necessary to systematically study the effects of collisions.

In this new interdisciplinary field between atomic and nuclear physics, the required advanced experimental infrastructure has often not been available in a single, highly specialised laboratory. Therefore, transportable laser spectroscopy equipment was required.

Objectives

The specific objectives of the project were:

1. To minimise the effects of kinetic energy and interaction between the ions of laser-cooled Coulomb crystals to reduce systematic frequency shifts in optical clock applications by investigation of the structure and dynamics of laser-cooled two-species Coulomb crystals of ions of different masses and charge states.
2. To implement sympathetic cooling of clock relevant ions (including highly charged ions) with suitable coolant ions to reduce systematic frequency shifts.
3. To provide reliable estimates on collisional frequency shifts due to the background gas in trapped ion optical clocks by investigating collisions of trapped ions with neutral atoms and molecules. To identify and eliminate causes of ion loss caused by charge exchange or chemical reactions that lead to the formation of molecular ions.
4. To develop efficient sources of ^{229}Th (thorium) ions in charge states Th^{3+} and Th^{4+}, based on recoil ions from ^{233}U (uranium). This should allow loading of an ion trap in ultrahigh vacuum from a source of less than 10 kBq ^{233}U activity.
5. To develop transportable equipment for laser cooling, high resolution spectroscopy and precision optical frequency measurements that enable common experiments to be carried out at nuclear physics laboratories and optical metrology laboratories.
6. To disseminate the results among the nuclear physics and optical metrology communities in order to advance fundamental research to metrology and enable further applications of optical clocks.

Progress beyond the state of the art

This project has extended the work of the earlier EMRP SIB04 'ion clock' and EMPIR 15SIB03 'OC18' projects in terms of i) new reference transitions, ii) a multi-ion approach at higher ion numbers, iii) the study of sympathetic cooling of the ions of interest for the clock, together with ions that are suitable for laser cooling, and iv) the study of collisional frequency shifts that have so far only been estimated based on simplified models.

This project has complemented the single-ion experiments that have been performed so far, and also offered the opportunity to increase the frequency stability of the optical clock through the simultaneous interrogation of many ions, resulting in a higher signal-to-noise ratio of the signal that steers the clock oscillator.

Objective 1: Structure and dynamics of laser-cooled two-species Coulomb crystals

This project has performed systematic studies of the structure and dynamics of larger Coulomb crystals, including the interaction between ions and the trap potential, in order to find ion configurations and trap parameters that minimise second order Doppler, Stark and quadrupole shifts, while providing extended interrogation times for higher ion numbers. The findings are applicable to a wide class of trapped ions of interest for optical clocks, including Yb^+ (ytterbium), with an extremely narrow electric octupole transition, highly charged ions that are sympathetically cooled by singly charged coolant, and thorium ions in different charge states that are investigated for the ^{229}Th nuclear clock.

Objective 2: Sympathetic cooling of clock relevant ions

In this project, sympathetic cooling in two-species crystals has been investigated. Well established techniques have been extended to sympathetically cool the clock ions. The number of simultaneously interrogated ions as well as the coherent interrogation time was enlarged beyond 1s with the aim of improving the achievable stability (10^{-16}) of ion clocks while maintaining their high accuracy.

Objective 3: Reliable estimation of collisional frequency shifts and identification and elimination of causes of ion loss

Interactions of trapped ions in collisions with neutral atoms or small molecules from background gas may lead to a systematic frequency shift and have been the most common cause for ion loss, through the formation of molecular ions. In a single-ion clock in ultrahigh vacuum, collisional frequency shifts had not been detected, but quantitative experimental data has been highly desirable for reliable and comprehensive uncertainty evaluations in the low 10^{-18} range. The formation of a molecular ion in a single-ion clock is usually a rare event, occurring once on the timescale of minutes to hours, but inevitably leads to an unintended interruption of the clock operation. Quantitative experiments on small frequency shifts and an identification of rare reaction events have been facilitated by the experiments with large Coulomb crystals that have been carried out in this project.

Objective 4: Efficient sources of ^{229}Th and other elements in intermediate (triply charged) and higher charge states

This project has investigated the loading of the trap from α-decay recoil ions in an ultrahigh-vacuum environment (UHV), eliminating the use of buffer gas that is incompatible with high-resolution spectroscopy. This work has provided a simple, robust and reliable ion source for use in different types of ^{229}Th nuclear clocks. Experiments in an electron beam ion trap (EBIT) provide access to higher charge states.

Objective 5: Transportable equipment for laser cooling, high resolution spectroscopy and precision optical frequency measurements

In order to provide a basis for joint experiments on optical frequency metrology at institutes that do not have the infrastructure and expertise in laser equipment, transportable equipment for laser cooling of ions (Sr^{+}), for high resolution spectroscopy of selected reference transitions and for the transfer of optical reference frequencies in the range of 1540 nm which are suitable for transmission through optical fibres has been developed. This project has addressed the subject of large Coulomb crystals with 10^{2} to 10^{4} ions and novel reference systems proposed in highly charged ions and the ^{229}Th nuclear clock.

Results

Objective 1: Structure and dynamics of laser-cooled two-species Coulomb crystals

The calculation of electric quadrupole shifts produced by the field gradients of surrounding ions shows the range of trap parameters and ion configurations that are suitable for precision experiments and the realisation of a multi-ion frequency standard. Different two-species Coulomb crystals with different mass ratios and geometric configurations has been realised experimentally, demonstrating deterministic ion loading and sorting routines. In total, four different combinations of clock and coolant ions have been investigated and characterized. First clock operation has been demonstrated in two specific pairs of ions. The objective has been successfully achieved.

Objective 2: Sympathetic cooling of clock relevant ions

Measurements of heating rates below 10 quanta/s in two-ion crystals have shown that 1s interrogation times are achievable. Heating rates of higher-order modes in multi-ion-crystals have been measured and are similar to the low rates on the order of 0.1 quanta/s observed for single ions. A trap loading method based on a combination of laser evaporation from the metal and resonant photoionisation allows for an isotope-selective rapid (re-) loading with minimal perturbation of the trap structure and other trapped ions. A superconducting ion trap has been built and successfully used for trapping Coulomb crystals (a 'first in the world'). The high Q-value of the circuit provides very low RF heating through RF noise. Coolant ions Sr^{+}, Be^{+} and Yb^{+} have been characterised for sympathetic cooling of the spectroscopy ions Th^{3+}, highly charged Ar ions, In^{+} and Yb^{+} in different configurations of two-species Coulomb crystals. The objective has been successfully achieved.

Objective 3: Reliable estimation of collisional frequency shifts and identification and elimination of causes of ion loss

Cryo-cleaning of the argon and helium buffer gas from condensable organic residuals has allowed to extend storage times of the highly reactive Th^{+} and Th^{2+} ions by a factor 2.5 to 1600 s and 900 s for Th^{+} and Th^{2+} respectively. In Th^{+} and Th^{2+}, molecule formation has been counteracted by ultraviolet photo dissociation lasers. These results are highly relevant for future experiments with Th ions, that are most strongly affected by molecule formation. Collisional population and quenching rates in the range of 10^{-2} to 10^{-4} per s have been analysed for a metastable level in Yb^{+}, through the observation of Coulomb crystals over extended periods of time. The result has shown the high sensitivity of the experiment to study such rare events and enables the use as a diagnostics of background pressure in the ultrahigh-vacuum range. Experimental tests for collisional frequency shifts were performed on Yb^{+} ions. Clock operation and frequencies were monitored against a stable reference while the base pressure (mainly He) was increased by a factor of approximately 100. As no significant frequency shift has been observed, the pressure shift at base pressure could be constrained in the 10^{-18} range. The objective has been successfully achieved.

Objective 4: Efficient sources of ^{229}Th and other elements in intermediate (triply charged) and higher charge states

The demonstration of efficient laser ablation loading of an electron beam ion trap has identified a practical method to build sources of rare isotopes like ^{229}Th. A recoil ion source has been designed and built. It uses recoil ions from radioactive α-decay and transfers them to an ion trap for precision spectroscopy experiments. A buffer-gas free trapping region is obtained by using a gate valve with integrated ion optics that separates the ion trap from the recoil ion source. The objective has been successfully achieved.

Objective 5: Transportable equipment for laser cooling, high resolution spectroscopy and precision optical frequency measurements

A transportable 1542 nm reference laser, passively stabilized to an ULE reference cavity, has been set up and is used to link phase-stable optical frequency transfer connections via telecom fibres. A Doppler-compensated fibre link at 1542 nm has been established between ISI-Brno and BEV-Vienna and comparison measurements have been performed, connecting the reference laser to the BEV primary Cs standard and hydrogen masers. A scheme has been established to connect the 150 nm Th-229 nuclear transition wavelength to the transfer laser. The objective has been successfully achieved.

Impact

The project has made 40 conference presentations, 29 peer-reviewed scientific publications (including 7 master theses) and 30 other dissemination activities. 15 training events have been organised. The impact of this work has been predominantly on the scientific community and on the long-term development of metrological capabilities at the frontiers of measurement science. Longer-term economic impact from knowledge transfer to industry is foreseeable.

A strong interest from the scientific community in topics of quantum metrology, which are being investigated in this project have led to 5 invitations for project group leaders to deliver lectures at well-known international summer schools with a target audience of PhD students and Postdocs. Furthermore, the largest newspaper in Finland; Helsingin Sanomat; featured a 2-page article on the Finnish timescale and the VTT ion clock in July 2019. As an initiative of the precision physics and quantum information community during the COVID-19 shutdown, a virtual “Seminar on Precision Physics and Fundamental Symmetries” has been arranged at https://indico.cern.ch/category/12183/ and has been attended by around 300 participants. Work from this project has been presented in two talks of the seminar series. The international online workshop “Novel Movements for Optical Clocks and Sensors” was organised jointly by 17FUN07 CC4C and 17FUN03 USOQS on 20-22 September 2021. It featured presentations by five invited speakers, while the results of the projects were presented by seven speakers from the two consortia. The workshop was very successful with more than 170 attendees from 24 different countries.

Impact on industrial and other user communities

Optical clocks are attracting interest in different sectors, such as space, aerospace, telecommunications and energy networks. Key subsystems of optical clocks (e.g. laser systems, reference resonators, frequency combs, ion traps and optical traps) are commercially available from several vendors. European SMEs have a significant market share in this sector. The European Quantum Technologies Flagship and in particular the application of trapped laser-cooled ions in quantum computing has identified further applications and has intensified the industrial developments in these fields. PTB has been scientific coordinator for the German nationally funded quantum technology project ‘opticlock’ www.opticlock.de/en/info/, which has developed a fully automated Yb^+ single-ion clock as a demonstrator for a commercial product. The present project has strengthened and expanded the relations between NMIs, academic institutes and industry through knowledge exchange and cooperation. The industrial development of optical and electronic systems has been supported by the participating NMIs via guidance on target specifications for novel applications and ad hoc support in the characterisation of commercial prototypes.

Impact on the metrology and scientific communities

This project has led to improvements in the operational reliability and precision of optical clocks with trapped ions, which are among the most precise optical frequency standards available today. The interrogation of many ions has improved the stability and reduced the averaging time required to obtain a targeted statistical uncertainty. It has enabled the investigation of new reference systems, including a nuclear transition, that are promising candidates for optical clocks. This has provided input to the selection of suitable reference systems for a redefinition of the SI second, an essential contribution to fundamental metrology and to the long-term development of the SI system of units.

This project has been fostering new interdisciplinary links and has led to an exchange of technology and know-how between high-precision optical frequency metrology and nuclear physics. High precision methods for optical frequency standards that have been developed by NMIs have been made available for a wider class of systems of scientific interest, such as highly charged ions This contributes to an improved understanding of the structure of atoms, molecules and nuclei, and to tests of fundamental physics through precision spectroscopic studies and frequency measurements on selected systems of high sensitivity (e.g. for violations of Einstein's equivalence principle).

Impact on relevant standards

This project has developed and strengthened the high-level metrological infrastructure in the measurement of time and frequency, and in the longer term this will improve the capabilities in time scale generation and time dissemination. The consortium has liaised with the time section of BIPM, and has reported to the Consultative Committee for Time and Frequency (CCTF) for its meetings in 2020 and 2021. It has been working with the Consultative Committee for Length - Consultative Committee for Time and Frequency (CCL-CCTF) Working Group on Frequency Standards (WGPSFS) and the EURAMET Technical Committee for Time and Frequency (TC-TF). The consortium has contributed to the work of the CCTF Task Force on updating the Roadmap for the redefinition of the second, especially on the topic of atomic frequency standards and possible redefinition approaches. The task group has discussed several options for the use of optical frequency standards in time scales and for a redefinition of the second. Members of the consortium have been contributing to the preparation of an online survey that has been conducted in 2021, addressed to concerned institutional bodies and stakeholder communities on several topics, including the redefinition of the second via an optical frequency standard.

Longer-term economic, social and environmental impacts

Long-term impact of this research will result from the pivotal role of atomic clocks in the revised SI and from the expected widespread use of quantum technologies in communication, sensing and metrology. The results will allow the international metrological community to make better informed decisions towards a future redefinition of the SI second. Improved atomic clocks have relevance for technological applications, in sectors such as space, aerospace, telecommunications and energy networks. Trapped ion optical frequency standards offer excellent accuracy and have the best potential for miniaturisation of the "physics package" which is of major importance in their development as payloads on board satellites and aerospace vehicles.

List of publications

1. M. G. Kozlov et al., Highly charged ions: Optical clocks and applications in fundamental physics, Rev. Mod. Phys. **90**, 045005 (2018), https://doi.org/10.1103/RevModPhys.90.045005

2. Detti et al., A compact radiofrequency drive based on interdependent resonant circuits for precise control of ion traps, Rev. Sci. Instr. **90**, 023201 (2019), https://aip.scitation.org/doi/10.1063/1.5063305

3. E. Perego et al., A scalable hardware and software control apparatus for experiments with hybrid quantum systems, Rev. Sci. Instr. **89**, 113116 (2018), https://doi.org/10.1063/1.5049120

4. T. Leopold et al., A cryogenic radio-frequency ion trap for quantum logic spectroscopy of highly charged ions, Rev. Sci. Instr. **90**, 073201 (2019); https://doi.org/10.1063/1.5100594

5. R. Lange et al., Coherent Suppression of Tensor Frequency Shifts through Magnetic Field Rotation, Phys. Rev. Lett. **125**, 143201 (2020), https://doi.org/10.1103/PhysRevLett.125.143201

6. T. Sikorsky et al., Measurement of the Th229 Isomer Energy with a Magnetic Microcalorimeter, Phys. Rev. Lett. **125**, 142503 (2020), https://doi.org/10.1103/PhysRevLett.125.142503

7. P. V. Bilous et al., Electronic Bridge Excitation in Highly Charged Th229 Ions, Phys. Rev. Lett. **124**, 192502 (2020), https://doi.org/10.1103/PhysRevLett.124.192502

8. M. Pimon et al., DFT calculation of 229thorium-doped magnesium fluoride for nuclear laser spectroscopy Journal of Physics: Condensed Matter **32,** 255503 (2020) https://iopscience.iop.org/article/10.1088/1361-648X/ab7c90/pdf

9. P. Micke et al., Coherent laser spectroscopy of highly charged ions using quantum logic, Nature **578**, 60 (2020), https://doi.org/10.1038/s41586-020-1959-8

10. L. Timm et al., Energy localization in an atomic chain with a topological soliton, Phys. Rev. Research 2, 033198 (2020), https://doi.org/10.1103/PhysRevResearch.2.033198

11. E. Perego et al.,Electro-Optical Ion Trap for Experiments with Atom-Ion Quantum Hybrid Systems Appl. Sci. **10**, 2222 (2020), https://doi.org/10.3390/app10072222

12. Lesundak et al., Optical frequency analysis on dark state of a single trapped ion, Optics Express **28**, 13091-13103 (2020), https://doi.org/10.1364/OE.389411

13. R. Lange et al., Improved Limits for Violations of Local Position Invariance from Atomic Clock Comparisons, Phys. Rev. Lett. **126**, 011102 (2021), https://journals.aps.org/prl/pdf/10.1103/PhysRevLett.126.011102

14. E. Peik et al., Nuclear clocks for testing fundamental physics, Quantum Sci. Technol. **6** 034002 (2021), https://doi.org/10.1088/2058-9565/abe9c2

15. K. Beeks et al., The Thorium-229 low-energy isomer and the nuclear clock, Nature Review Physics **3**, 238–248 (2021), https://doi.org/10.1038/s42254-021-00286-6

16. J. Seres et al., High harmonic generation in AlN due to out-of-surface electron orbitals OSA Continuum 4, 47-54 (2021), https://doi.org/10.1364/OSAC.404942

17. D. Kalincev et al., Motional heating of spatially extended ion crystals, Quantum Sci. Technol. **6,** 034003 (2021), https://doi.org/10.1088/2058-9565/abee99

18. S. A. King et al., Algorithmic Ground-state Cooling of Weakly-Coupled Oscillators using Quantum Logic, Phys. Rev. X 11, 041049 (2021), https://doi.org/10.1103/PhysRevX.11.041049

19. J. Stark et al., An ultralow-noise superconducting radio-frequency ion trap for frequency metrology with highly charged ions, Rev. Sci. Instrum. **92**, 083203 (2021), https://doi.org/10.1063/5.0046569

20. N.-H. Rehbehn et al., Sensitivity to new physics of isotope-shift studies using the coronal lines of highly charged calcium ions, Phys. Rev. A **103**, L040801 (2021), https://doi.org/10.1103/PhysRevA.103.L040801

21. J. Kiethe et al., Finite-temperature spectrum at the symmetry-breaking linear to zigzag transition, Phys. Rev. B **103**, 104106 (2021), https://doi.org/10.1103/PhysRevB.103.104106

22. M. Oral et al., Simulation of Motion of Many Ions in a Linear Paul Trap, Int. J. Mod. Phys. A **34**, 1942003 (2019), DOI:10.1142/S0217751X1942003X, https://arxiv.org/abs/2112.02203

23. L.J. Spiess, Setup of a vibration-suppressed cryogenic system for a RF ion trap with minimum micromotion, Master Thesis, Ruprecht-Karls-Universität, Heidelberg (2018) http://hdl.handle.net/21.11116/0000-0002-5FDF-A

24. M. K. Rosner, Production and preparation of highly charged ions for re-trapping in ultra-cold environments, Master Thesis, Ruprecht-Karls-Universitat, Heidelberg (2019), http://hdl.handle.net/21.11116/0000-0005-5B37-8

25. S. Bogen, Frequenz-, Leistungs- und Positionsstabilisierung von UV-Lasersystemen fur Frequenzmetrologie mit hochgeladenen Ionen, Master Thesis, Ruprecht-Karls-Universitat, Heidelberg (2019), http://hdl.handle.net/21.11116/0000-0005-1733-8

26. Ch. Warnecke, Imaging of Coulomb crystals in a cryogenic Paul trap experiment, Master Thesis, Ruprecht-Karls-Universität, Heidelberg (2019), http://hdl.handle.net/21.11116/0000-0004-F47D-D

27. D. Vadlejch, Characterization of micro-motion and its influence on systematic frequency shifts of quadrupole transition of Calcium ion trapped in Paul trap, Master Thesis, Brno University of Technology (2020), https://dspace.vutbr.cz/handle/11012/192380?locale-attribute=en

28. J. Grim, Spectroscopy of a photoionization transition of neutral Calcium atom emitted by laser ablation, Master Thesis, Brno University of Technology (2020), https://dspace.vutbr.cz/handle/11012/192381?locale-attribute=en

29. T. Pronebner, An ultra-stable Laser as an optical frequency standard, Master Thesis, Vienna University of Technology, https://repositum.tuwien.at/bitstream/20.500.12708/18346/1/Pronebner%20Thomas%20Tobias%20-%202021%20-%20An%20ultrastable%20laser%20as%20optical%20frequency...pdf

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Project start date and duration:		1 May 2018, 42 months
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