



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 are needed to extend applications in communication, in navigation and in fundamental metrology. This project will investigate 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 are based on single ions, this project will investigate large ensembles of ions in a Coulomb-coupled solid-like state, known as Coulomb crystals (CCs). The multi-ion approach used in this project will provide higher signal-to-noise for clocks of improved stability, enable the investigation of small frequency shifts caused by collisions with neutral particles or interactions between the ions, and the opportunity to study new systems which may show improved uncertainty.

Need

Within the revised SI system of units, due for implementation in May 2019, the realisation of the unit of time will maintain a pivotal role, as the unit second will be 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 low 10^{-18} range. New reference systems with specific advantages in terms of accuracy or stability require 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 requires further investigation concerning frequency shifts induced by these interactions.

Existing methods of trap loading in optical clocks are 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, requires 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 is necessary to systematically study the effects of collisions.

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

Objectives

The specific objectives of the project are:

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 extends 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 will complement the single-ion experiments that have been performed so far, and also offer 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 will perform 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 will be 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 will be investigated for the ^{229}Th nuclear clock.

Objective 2: Sympathetic cooling of clock relevant ions

In this project, sympathetic cooling in two-species crystals will be investigated. Well established techniques will be extended to sympathetically cool the clock ions. The number of simultaneously interrogated ions as well as the coherent interrogation time will be 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 are the most common cause for ion loss, through the formation of molecular ions. In a single-ion clock in ultrahigh vacuum, collisional frequency shifts have not yet been detected, but quantitative experimental data is 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 will be facilitated by the experiments with large Coulomb crystals that will be 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 will investigate 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 will

provide 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) will 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 will be developed. This project will address 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 will be suitable for precision experiments and the realisation of a multi-ion frequency standard. A variety of different two-species Coulomb crystals with different mass ratios and geometric configurations has been realised experimentally, demonstrating deterministic ion loading and sorting routines.

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 will be 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 is successfully being used for trapping Coulomb crystals. The high Q-value of the circuit promises 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.

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^+ , molecule formation is further counteracted by a photo dissociation laser at 266 nm. This process shows a factor 4 improved efficiency in argon compared to helium, trapping times of 5 days (argon) and 1.5 days (helium) have been demonstrated. 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. This result shows the high sensitivity of the experiment to study such rare events and enables us to use them as a diagnostics of background pressure in the ultrahigh-vacuum range. First experimental tests for collisional frequency shifts were performed on Yb^+ ions, by looking at the transient response of the ion after switching off the ion pump in a sealed-off system. Clock operation and frequencies were monitored against a stable reference while the base pressure (mainly He) was increased by a factor of approximately 100.

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 identifies a practical method that can be applied to sources of rare isotopes like ^{229}Th . Recoil ion sources have been designed to slow down energetic recoil ions from radioactive α -decay and transfer them to an ion trap for precision spectroscopy experiments. A buffer-gas free trapping region will be obtained by use of a gate valve with integrated ion optics that will separate the ion trap from the recoil ion source after trap loading.

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 can be linked to 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 commenced, connecting the reference laser to the local primary Cs standard and masers. A scheme has been established to connect the 150 nm Th-229 nuclear transition wavelength to the transfer laser and has been adapted to the now more precisely known nuclear transition wavelength.

Impact

In the first 30 months of the project, 27 conference presentations have been given, 11 training events have been organised, 13 peer-reviewed scientific papers and 6 master theses have been published. The impact of this work will be 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.

Impact on industrial and other user communities

Although not yet in use, optical clocks are attracting much 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 already commercially available from several vendors. European SMEs have a significant market share in this sector. The new European Quantum Technologies Flagship will identify further applications in the wider fields of quantum information, communication, and sensors and will intensify the industrial developments in these fields. PTB is scientific coordinator for the 'opticlock' project, which has recently succeeded to develop a fully automated Yb⁺ single-ion clock as a demonstrator for a commercial product within the German quantum technology programme QUTEQA. CC4C is strengthening and widening the relations between NMIs, academic institutes and industry through knowledge exchange and cooperation. The industrial development of optical and electronic systems is 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 is leading 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 will improve the stability and reduce the averaging time required to obtain a targeted statistical uncertainty. It will enable the investigation of new reference systems, including a nuclear transition, that are promising candidates for optical clocks. This will provide 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 is fostering new interdisciplinary links, and lead 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 will be made available to a wider class of systems of scientific interest, such as highly charged ions This will contribute 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 will develop and strengthen the high-level metrological infrastructure in the measurement of time and frequency, and in the longer term it will improve the capabilities in time scale generation and time dissemination. The consortium is liaising with the time section of BIPM, and is reporting to the Consultative Committee for Time and Frequency (CCTF) for its 22nd meeting in October 2020 and March 2021. It is working with the 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 will contribute to the preparation of an online survey (to open in January 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

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This list is also available here: <https://www.euramet.org/repository/research-publications-repository-link/>

Project start date and duration:		1 May 2018, 42 months
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Internal Funded Partners:	External Funded Partners:	Unfunded Partners:
1 PTB, Germany	5 ISI, Czech Republic	
2 BEV-PTP, Austria	6 LUH, Germany	
3 INRIM, Italy	7 MPG, Germany	
4 VTT, Finland	8 TU WIEN, Austria	
RMG: -		