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1 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.

2 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.

3 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.

4 Results

4.1 Structure and dynamics of laser-cooled two-species Coulomb crystals

While the achievable gain in signal-to-noise ratio in an optical clock from the interrogation of many ions instead of a single one is evident, the influence of multi-ion operation on the accuracy of the clock is a much more complicated subject, involving the interactions among the ions and with the trap potential, and problems of the internal dynamics of the Coulomb crystal. In this project the consortium has undertaken systematic studies of the structure and dynamics of Coulomb crystals 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. Work has been performed on different species of trapped ions of interest for optical clocks: In^+ is a clock ion that is relatively insensitive to field-induced systematic shifts but difficult to cool because of an unusually narrow cooling transition. Yb^+ (ytterbium) possesses an extremely narrow electric octupole transition that potentially offers very long coherence times but requires sympathetic cooling to avoid heating during interrogation. Highly charged ions can be sympathetically cooled by a light singly charged coolant ion. Thorium ions in different charge states are investigated because of the low-energy isomer of ^{229}Th that is attractive as a reference for an optical nuclear clock.

In order to characterize motional frequency shifts from the Doppler effect, cooling rates and phonon distributions have been calculated at LUH for In^+/Yb^+ two-species Coulomb crystals in different configurations for ion numbers ranging from 3+10 to 5+5 for the two species. The resulting kinetic energy and relativistic Doppler shift are nearly independent of the spatial ion configuration, but the cooling times differ by about two orders of magnitude. Spatially resolved measurements of excess micromotion indicate that an uncertainty below 10^{-18} in the relativistic Doppler shift is achievable over extended regions (>1 mm) along the axis of the linear trap. LUH has also determined heating rates in small mixed-species In^+/Yb^+ Coulomb crystals.

Spectroscopy of large crystals of 10^2-10^4 Ca^+ ions has been studied in calculations and simulations of frequency shifts and line shapes at ISI. Accompanying experiments have been performed with small ion numbers. For the experiment on large Ca^+ crystals, the tuneable RF transformers have been built for drive at 24.7MHz with tuning range ± 1 MHz where the magic frequency of the Ca^+ ion is located and compensates the relativistic Doppler and the AC Stark shift induced by the trap. The Ramsey sequence implementation for line-shift analysis and software defined multiple servo-loop controller for sequential locking of the clock laser to two and more Zeeman levels have been completed. Clock operation of Ca^+ ion clock and pilot reference measurement against the local timescale at ISI (H-maser) have been tested. The compensation of the B-field fluctuation on basis of the sequential lock at two Zeeman transitions was verified successfully. Experiments have been performed on measurements of line-shift of the Ca^+ $D_{5/2}$ carrier for a single ion at different positions in the trap, for the RF drive frequency far from the magic frequency and at RF magic frequency. These developments have provided an experimental basis for measurements of the lineshape in Coulomb crystals with different ion numbers.

Since the trapped-ion optical clock would be sensitive to frequency shifts induced by fluctuations in the electric trapping potential, the instability of DC voltages applied to the ion trap has been measured and the result has been used to infer the instability of quadrupole shifts on the clock transitions in Yb^+ and In^+ at LUH and PTB. Co-trapping and cooling of Sr^+ and Yb^+ has been demonstrated at PTB in a setup that is suitable for operation as an optical clock. Laser systems with narrow linewidth and well controlled frequency have been developed for cooling and interrogation in long-term stable clock operation. PTB has investigated the micromotion and heating rates for the Yb^+/Sr^+ combination. Spectroscopy experiments with the two-species combination Yb^+/Sr^+ have not yet been possible because of an anomalously high heating rate in the trap. Precision frequency measurements of the clock transition Sr^+ with single ions for the characterization of the blackbody radiation shift have been performed against the established single-ion clock with Yb^+ at PTB.

MPG has performed micromotion measurements with a single Be^+ ion, confined in a novel superconducting ion trap. Motional modes of the confined ion were characterized via excitation by an intensity-modulated cooling laser beam. Pending the implementation of retrapping highly charged ions from the electron beam ionization trap, the secular mode structure of sporadically appearing mixed-species BeH^+/Be^+ crystals was characterized.

During the prolongation phase of the project it has been possible for PTB, LUH, and MPG cooperation to perform first two-ion, two-species clock operation and precision frequency determinations of optical frequency ratios for the two novel combinations of (clock ion, coolant ion): ($^{115}In^+$, $^{172}Yb^+$) and ($^{40}Ar^{13+}$, $^{9}Be^+$). Stability comparisons and frequency ratio measurements have been performed against the single-ion optical clock with Yb^+ and the optical lattice clock with Sr atoms at PTB. While a full evaluation of the measurements is still pending, they indicate systematic uncertainties in the low 10^{-17} range, representing a significant improvement for these novel species of optical clocks. The objective has been successfully achieved.

4.2 Sympathetic cooling of clock relevant ions in two-species Coulomb crystals

While laser cooling of single ions had been well-established, as well as cooling of single-species linear Coulomb crystals in the context of quantum information experiments, cooling of dual-species Coulomb crystals had been explored much less, especially for chains larger than two ions. Here, the different charge-to-mass ratio of the two ion species offers an additional degree of freedom that influences how the ions participate in motional modes. Through resolved sideband cooling before clock interrogation the thermal energy of the ions can be reduced. But due to coupling to any fluctuating electric fields, e.g. from noise in the trap electrodes, the ions heat up during periods without laser cooling, which limits the maximum interrogation time even after ground state cooling. This problem can be mitigated by sympathetic cooling with a second ion species during clock operation. For an Al^+ clock employing quantum logic spectroscopy, continuous sympathetic Doppler cooling of the logic ion $^{25}Mg^+$ during clock interrogation had been applied at NIST in the USA to compensate for the gain of thermal motion due to heating. The light shift induced by the cooling laser added only a small contribution to the total uncertainty. With an improved trap design that provides smaller and controllable heating rates, lower uncertainties can be achieved, if the two-ion crystal is initially cooled to its motional ground state. In this case, for periods of up to a few seconds, the second-order Doppler shift due to thermal motion and its uncertainty are smaller than during continuous Doppler cooling. For very long coherent interrogations, like in Ramsey interrogation with extended dark times, even heating rates of only a few quanta of motion per second can pose a limitation. In this case, resolved sideband cooling on at least one co-trapped ancillary ion can compensate for the heating and allow for small shifts due to thermal motion. The selection of the ancillary ion requires in-depth investigations of expected residual motion and available cooling techniques. While electromagnetically-induced-transparency (EIT) cooling can provide favorable cooling times even for larger crystals, work in this project has focused on resolved sideband cooling.

Sympathetic cooling is the most efficient when the charge-to-mass ratios of the two ion species are similar. The second important property of the coolant ion is the availability of a suitable cooling transition: a strong, cycling dipole transition for Doppler cooling and a narrow transition if sideband cooling is to be used. The alkaline-earth-metal ions are frequently used. The heavier ones, Ca^+, Sr^+ and Ba^+, as well as the lanthanide Yb^+ feature both types of transitions at convenient wavelengths. As such, the metastable *D* levels in these ions are too long-lived for efficient sideband cooling. By coupling the *D* level to a short-lived excited state using a 'quenching' laser, the effective linewidth of the cooling transition can be tuned. Although sideband cooling a Coulomb crystal to the motional ground state minimizes the residual motion, it gains energy through anomalous heating. The speed at which a mode gains energy is expressed by its *heating rate*, the number of quanta excited per second. This limits the minimum motion during sideband cooling but also the maximum coherent interrogation time of the ion even after initial ground-state cooling. Typical heating rates for linear traps are in the range of a few to few hundred quanta/s. The heating rates for the different modes can differ significantly, especially if the micromotion of the ion in the trap is not well compensated. If sideband cooling via an ancillary ion is used during long clock interrogations, the light required to cool the ancillary ion will imprint an ac Stark shift, called light shift, which can easily exceed 100 Hz. Various variations of Ramsey's method of separated oscillatory fields have been investigated to enable high-accuracy clock operation even for cases with significant light shift from the probe laser. Furthermore, the light shift can be canceled by properly adjusting the intensity of a beam causing a light shift of opposite sign. Also, the fact that the light shift contains scalar and tensor part needs to be considered and addressed using techniques that suppress other tensor frequency shifts.

The cooling efficiency of a mixed-species ion Coulomb crystal strongly depends on the cooling time for a specific crystal motional mode. Different crystal configurations can have a very different impact on a certain mode and thus dramatically change the overall cooling efficiency. The cooling rates of individual modes depends on the projection of their mode vector onto the wave vector of the incident cooling laser. Also, one needs to differentiate between mono-species Coulomb crystals and mixed-species crystals, such as In^+–Yb^+ crystals, as investigated in this project at LUH and PTB and discussed in the following.

A two-species crystal contains less cooling ions than a single-species crystal of same size, which reduces the overall cooling rate just by the number of participating cooling ions. As a consequence of the ions' mass imbalance, the modes have asymmetric amplitudes. These modes turn out to be either "Yb^+-like", i.e. the Yb^+ ions have a stronger oscillation amplitude, or, analogously, "In^+-like". This effect is even more pronounced in the radial direction. Modes with a smaller amplitude for the Yb^+ cooling ions feature longer cooling times, as the reduction of kinetic energy depends on the projection of the cooling laser onto the mode. In the extreme case of zero oscillation amplitude of Yb^+ ions in a certain mode, no cooling would occur. Furthermore, the radial Yb^+ and In^+-like modes appear to oscillate with a similar frequency as the single ion, which is 1 MHz for Yb^+ and 1.47 MHz for In^+, which is also observed in more complex linear crystals, e.g. containing 3 Yb^+ and 10 In^+¸ ions. This is a more desired composition due to a high signal-to-noise ratio for clock interrogation in In^+ ions, while the cooling efficiency based on the Yb^+ ion positions will be investigated in the following. For this particular composition of a mixed-species crystal, motional modes have been calculated for all of the 146 possible crystal configurations, i.e. the individual positions of In^+ and Yb^+ ions in a 13-ion Coulomb crystal. Figure 1 shows the minimum and maximum mode cooling time, as well as the mean value over all modes.

The maximum cooling time varies from 21 ms radially (1 ms axially) in the optimal crystal configuration to more than 10 s for the worst-case configuration in radial and more than 100 s in axial direction, which is a difference by up to 5 orders of magnitude. To gain a better insight on how an optimal or a worst-case configuration looks like, Figure 2 depicts the first six optimal and the last six worst-case configurations. Although there seems to be not much of a difference between, e.g. configurations no. 4 and no. 142, the maximum cooling time differs extremely. As a general conclusion drawn from looking at all configurations, symmetry within the crystal should be avoided. Also, the cooling Yb^+ ions should be rather equally distributed over the whole crystal length instead of being close neighbors.

The group at LUH has also identified simple and robust ordering procedures to bring a two-species ensemble of ions of different charge-to-mass ratio nearly deterministically into two types of ion distributions along linear chains: with the heavier ions occupying the central region of the chain, and with both species at alternating positions (see Figure 3).

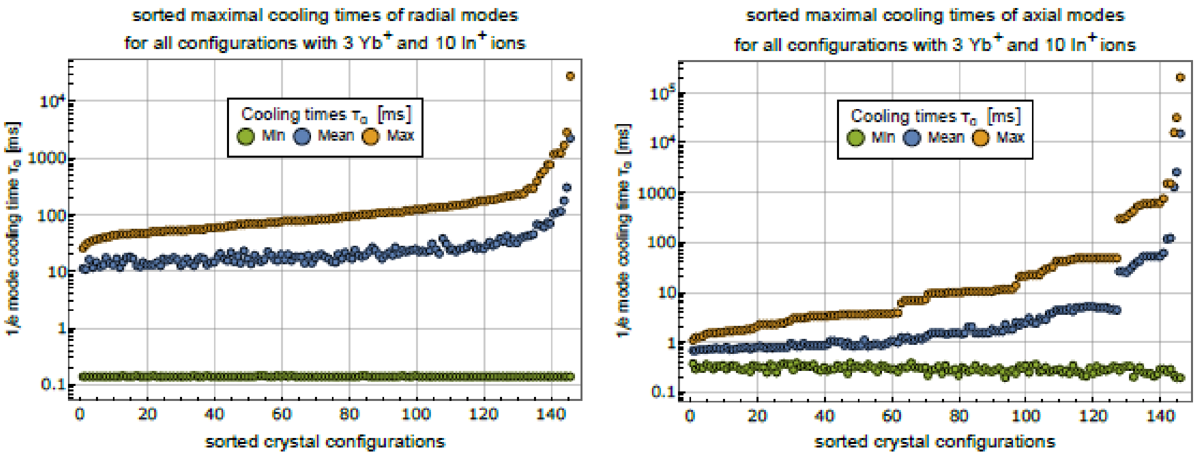

Figure 1. Maximum, minimum, and mean cooling times for radial (left) and axial (right) crystal modes occurring for individual crystal configurations. Trap secular frequencies are 1 MHz radially and 150 kHz axially.

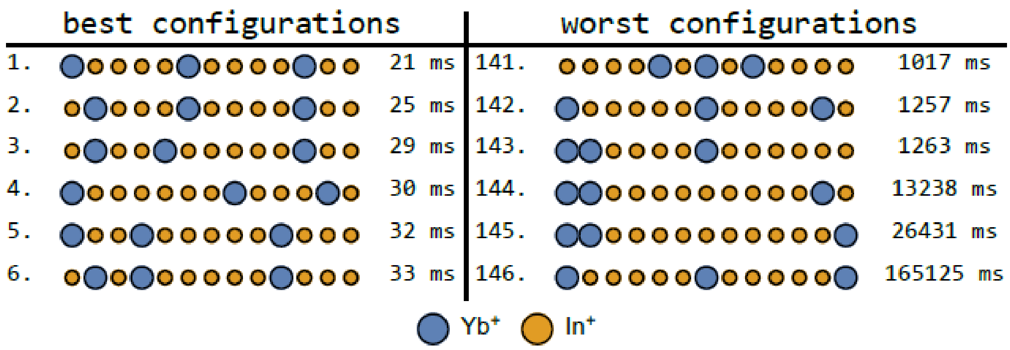

Figure 2. Optimal and worst-case configurations for a 13 ion Coulomb crystal sorted with respect to the maximum cooling time, as shown in Figure 1. Trap secular frequencies of 1 MHz (radial) and 150 kHz (axial) were used for the calculation. As a reference: the cooling time for a 13 Yb^+ ion crystal would be 0.13 ms for all modes.

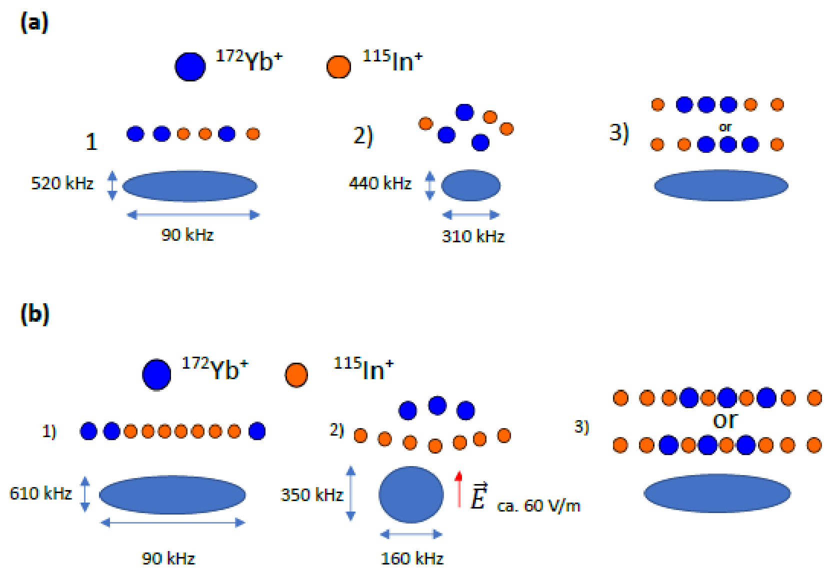

Figure 3. Ordering techniques for two-species crystals. (a) The heavier species can be moved to the crystal center by changing the confinement to yield a 2D or 3D intermediate configuration, before restoring the original trapping potential. (b) The additional application of a radial electric field separates the species radially and produces interleaved configurations.

Complementing the work on In^+/Yb^+, the consortium has also studied the combinations Yb^+/Sr^+ in cooperation of VTT and PTB and Be^+ together with highly charged ions in cooperation of MPG and LUH. Optimal strategies have been developed for sympathetic cooling of $^{171}Yb^+$ via sideband cooling of $^{88}Sr^+$ in small Coulomb crystals.

For continuous cooling of $^{171}Yb^+$ during clock operation on the extremely narrow electric octupole (E3) transition, that could potentially benefit from a coherence time exceeding many seconds, $^{88}Sr^+$ is an interesting coolant ion as the wavelengths of its $S_{1/2} \rightarrow D_{3/2}$ and $S_{1/2} \rightarrow D_{5/2}$ sideband cooling transitions are close to the magic wavelength at about 670 nm of the $^{171}Yb^+$ E3 clock transition, so that the Sr^+ cooling laser radiation will not produce any significant light shift for the Yb^+ clock. A special interest of this ion combination is that both ion species possess clock reference transitions that have partially complementary advantages and that are both recognized as secondary representations of the SI second. This opportunity will be investigated further in the EMPIR JRP 20FUN01 TSCAC project “Two-species and composite atomic clocks”.

Using a novel algorithmic cooling protocol for transferring phonons from poorly to efficiently cooled modes, LUH and MPG have demonstrated experimentally the simultaneously cooling of two motional modes of a Be^+-Ar^{13+} mixed Coulomb crystal close to the zero-point energies, despite the weak coupling between the ions. The lowest temperature reported for a highly charged ion, with a residual temperature of only $T \lesssim 200$ μK has been obtained in each of the two modes, corresponding to a residual mean motional phonon number below 0.4.

In summary, within this project sympathetic cooling has been implemented in several experiments and studied in two-ion, two-species operation for the combinations (clock ion, coolant ion): ($^{115}In^+$, $^{172}Yb^+$), ($^{171}Yb^+$, $^{88}Sr^+$) and ($^{40}Ar^{13+}$, $^{9}Be^+$). The objective has been successfully achieved.

4.3 Reliable estimation of collisional frequency shifts and identification and elimination of causes of ion loss

In optical clocks, trapped ions are confined in ultra-high vacuum environment with typical pressure in the 10^{-8} Pa range. Nevertheless, collisions between trapped ions and particles of the background gases (either atoms or molecules) affect the clock operation. In particular, the potential governing the neutral-ion collisions is governed at a large inter-particle distance R by an attractive term that originated from the interaction between the charge of the ion and the electric dipole induced on the neutral particle. The potential scales with R^{-4} and it is much longer-ranged than the neutral-neutral interaction which scales with R^{-4}. As a result, trapped ions collide in general more with the background gas than trapped neutral particles.

The project work on this subject has been directed towards providing a detailed study of the effects of ion-neutral collisions in the operation of an optical clock. In particular, collisions can be divided in two categories. First, elastic ion-neutral collisions, i.e. collisions that affect the ion only by changing its motional state, causing e.g. an increase of the ion kinetic energy, or a dephasing during the interrogation. Second, inelastic ion-neutral collisions, i.e. collisions in which the ion undergo a change in its internal or chemical state. Inelastic collisions can be divided into three main types: collisional quenching (in which the ion changes its internal state, and which can be either endothermic or exothermic), charge-exchange collisions (collisions of the kind $A+B^+ \rightarrow A^++B$), and molecular formation (processes of the kind $A+B^+ \rightarrow AB^+$). Inelastic collisions are in general more critical, as they cause an interruption of the clock interrogation.

Ion-neutral collisions were intensively studied in different environments and with different elements (Yb^+, In^+, Ca^+, Sr^+, Th^+). Because of the typical ultra-high vacuum environments of ion-based clocks, collisional events are scarce, making it challenging to detect and characterize the effects of ion-neutral collisions. Two main strategies were used. The first was to perform extremely long measurements by collecting the data for extended periods of time (up to several days). The second was to increase the pressure in the vacuum chamber, e.g. by turning off the ion pump, or by increasing the temperature of a getter pump. However, the pressure in the vacuum chambers could not be increased by more than two orders of magnitude, as the whole ion clock operation was excessively damaged by the background pressure. In order to tackle the problem with a different strategy, a new experimental apparatus was realized, in which Ba^+ ions can be immersed in an ultracold gas of Li atoms. This approach is promising for studying atom-ion collisions at considerably higher rates, and at a controlled temperature.

The effects of elastic collisions were studied by looking at possible frequency shifts of an ion-based clock as a function of the background gas pressure, and by looking at position changes of ions in a Coulomb crystal. A collisional fractional shift of $2(7) \times 10^{-18}$ was deduced at PTB for the E2 transition of the Yb^+ clock operating at 10^{-8} Pa (Figure 4). The effects of inelastic collisions were studied by looking at molecular formation and at collisional quenching. In particular, molecules formed by Sr^+, Ca^+, Th^+, Yb^+ were studied at VTT, ISI, PTB, and LUH, respectively. In the case of Ca^+, Th^+ and Yb^+ photodissociation was observed, in the case of Th^+ and Yb^+ after illuminating the molecular ion with UV light (see Figure 5). Additionally, collisional quenching was studied with Sr^+ (VTT), Ca^+ (ISI), and Yb^+ (PTB and LUH). In Ca^+ and Yb^+ quenching events were observed in which the ion changed its state from the long-lived excited state (D-state or F-state) to the ground state. Interestingly, in Sr^+ there was evidence of endothermic events in which the ion was passing from the $D_{3/2}$ level to the more energetic $D_{5/2}$ level after a collision, making it evident that the collisional physics can lead to very different observations depending on the chemical elements involved. With respect to the new setup for ultracold atom-ion collisions realized at INRiM, first results include the realization of a Li magneto-optical trap and the confinement of Ba^+ ions in a Paul trap. Remarkably, this new setup represents the first ion trapping experiment in Italy. The objective has been successfully achieved.

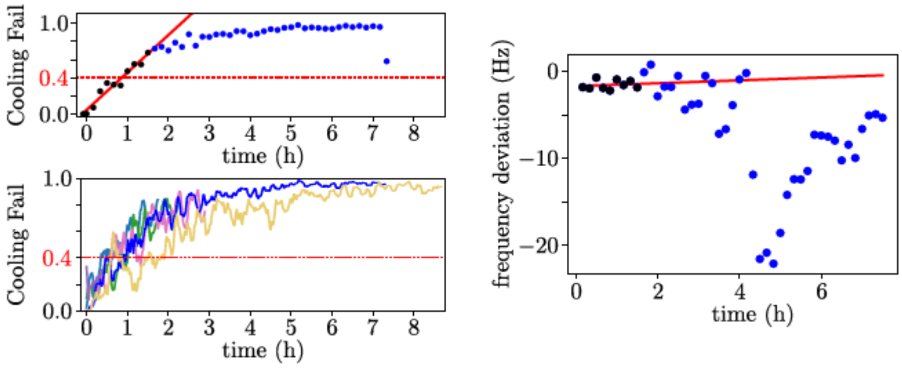

Figure 4. Collisional frequency shift measurements by shut down of the ion pump. Top left: Increase in the number of collisions resulting in loss of fluorescence from the ion, denoted as cooling-fail events, after the ion pump has been shut down. The initial increase is linear. At about 0.4 cooling-fail events per atom interrogation, the performance of the clock degrades significantly. Bottom left: Comparison of the cooling-fail events for multiple experiment runs (multiple shutdowns of the pump). The pressure increase is similar in each experiment, but the time until pump restarts (which marks the end of one experiment) varies from about one to about eight hours. Right side: Measurement of the atomic frequency deviation with respect to a stable reference (i. e. a second atomic clock) during one pump shutdown experiment (the one shown on the upper left side). Since clock performance degrades after a certain pressure increase, only the first part of the frequency measurement marked by the black dots is used analysis and is fitted by a linear function (red line). From a total of six measurements, a collisional frequency shift of 120(480) mHz on the E2 transition is deduced, with the accuracy limited by the statistical uncertainty.

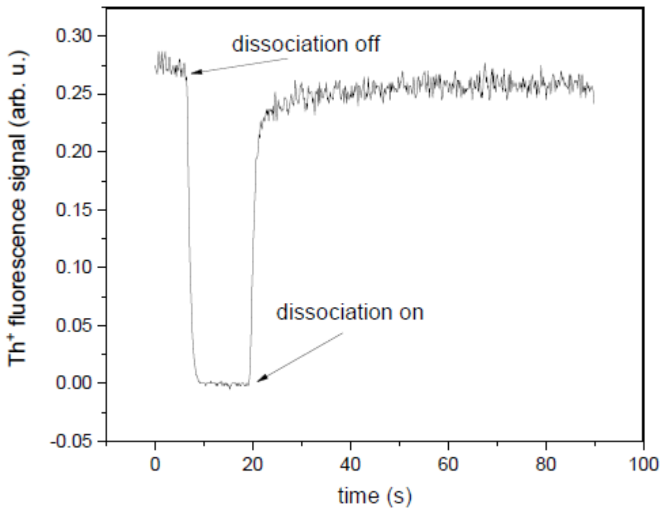

Figure 5. Photodissociation of ThX¸ compounds in CH_4 buffer gas. Th^+ ions are initially stored in the trap while applying the 266 nm laser radiation and using 5×10^{-3} Pa of methane as buffer gas. The dissociation laser is switched off 7 s after the start of the measurement, followed by a rapid decrease in fluorescence signal due to the predominant formation of $ThCH_2^+$ molecules. Switching the 266 nm laser on initiates dissociation of the molecules, visible as a reappearing fluorescence signal. The final fluorescence signal level is smaller than the initial, possibly due to further reactions to non-dissociable molecules with impurities of the buffer gas, while the 266 nm laser is switched off.

4.4 *Efficient ion sources for trap experiments with ^{229}Th, Th^{3+} and other elements in intermediate (triply charged) and higher charge states*

Because of its electronic level structure, Th^{3+} is the most suitable charge state of thorium ions for high-resolution laser spectroscopy experiments and for a highly precise nuclear clock based on trapped laser-cooled $^{229}Th^{3+}$ ions. In laser ablation loading of an ion trap from a solid ^{229}Th material, the efficiency decreases steeply with increasing charge. While well suited for experiments with Th^{+}, this method is therefore not advisable for Th^{3+} if an ion source with minimal amount of radioactive material is desired. It was decided to use $^{229}Th^{3+}$ ions originating from a ^{233}U recoil ion source with a ^{233}U activity of less than 10 kBq, the exemption level for work with this radio isotope in a common laboratory without specific radiation requirements. An additional motivation for using recoil ions arises from the fact that 2 % of the emitted thorium nuclei are in the isomeric state, making experiments with the isomer, for the study of its properties, possible without the need to actively excite it from the ground state.

^{233}U recoil sources have been produced and characterized by TU WIEN. When ^{233}U alpha-decays, an alpha particle of ~5 MeV is emitted, transferring a recoil of 84 keV onto the “freshly born” ^{229}Th daughter nucleus. If deposited in a thin layer, ^{233}U can hence be used as a source for 229^{Th}. TU WIEN has performed numerical simulations using the TRIM software package to evaluate the mean path of Th recoil ions in uranium to estimate the required U layer thickness and expected energy distribution of the Th recoil ions. It was found find that the recoil yield saturates at a layer thickness exceeding 10 nm. The value slightly depends on the exact chemical composition of the U layer, simulations were performed for metallic U and UO_2. Unfortunately, TRIM software does not allow to make a prediction on the distribution of charge states, but it gives the energy distribution of the escaping Th recoil ions. The result suggests, that about 0.2 % of all decay events produce Th recoil ions with an energy <10 eV, that could be trapped directly ion an ion trap. The overwhelming majority of recoil ions however have significantly higher energies, motivating the search for a slowing/focussing ion optics element.

For a 10 nm ^{233}U layer, one would expect a ^{229}Th ion yield of about 1 x 10^5 Th recoil ions per s and cm^2. TU WIEN established a new molecular plating produce to produce large, homogeneous, thin layers of ^{233}U. A new cell was set up with support from external cooperation partners at RIKEN (Japan), GSI (Germany) and the university Mainz. Crucial aspect in the design is the “parallel plate” arrangement, that produces highly homogeneous deposition. To determine the actual ^{229}Th yield produced by the fabricated ^{233}U sources, implantation experiments have been performed: a ^{233}U layer was held with < 1 mm distance to a virgin PIPS alpha detector for 20 days under vacuum. After this time, the ^{233}U source and the alpha decay of the implanted recoil nuclei (was detected for another 20 days. A quantitative analysis of such implantation allowed us to determine the absolute ^{229}Th recoil ion yield. A maximum yield of 110 ^{229}Th ions/(s · cm^2) was found. This corresponds to a recoil ion implantation efficiency of 3.8 % (recoil ions per alpha decay), almost two orders of magnitudes below the theoretical predictions. These findings are consistent with samples produced in LMU Munich and McGill University.

In order to investigate the opportunity of slowing and trapping recoil ions without the use of a buffer gas cell, ISI has performed a design study on the feasibility of a vacuum transfer ion optics, selecting the charge state $^{229}Th^{4+}$ as a test case. The properties of the ion source pose a challenge for the design of the transfer optics: the large size of the source area, the wide angular spread of the emerging Th-ions and their enormous energy range. In particle-optical terms, the source has low brightness, that is, the volume of the phase space occupied by the beam (which is invariant in time-independent focusing and retarding fields), is very high. The transfer optics will inevitably have limited throughput due to geometric and chromatic aberrations. Arrangements utilizing time dependent fields could equalize the energy range, but they impose some spatial collimation of the incoming beam, leading to losses.

As a choice for a transfer optics, a rotationally-symmetrical electrostatic lens has been considered, operating at 21 kV based on the recoil energy of 84 keV and the 4+ charge state of the ions (see Figure 6). In optimising the design, a trade-off between the accelerating and decelerating modes is made by putting the planar ion source on a negative potential. The ions first decelerate in the first section. The main focusing action is in the region between the focusing electrodes and the output aperture, which limits ions with the most divergent trajectories. The geometrical acceptance of the system at the nominal energy was estimated to 3 %. All in all, the transmission of the system (the proportion of ions that pass through) would be on the order of magnitude of 10^{-3}.

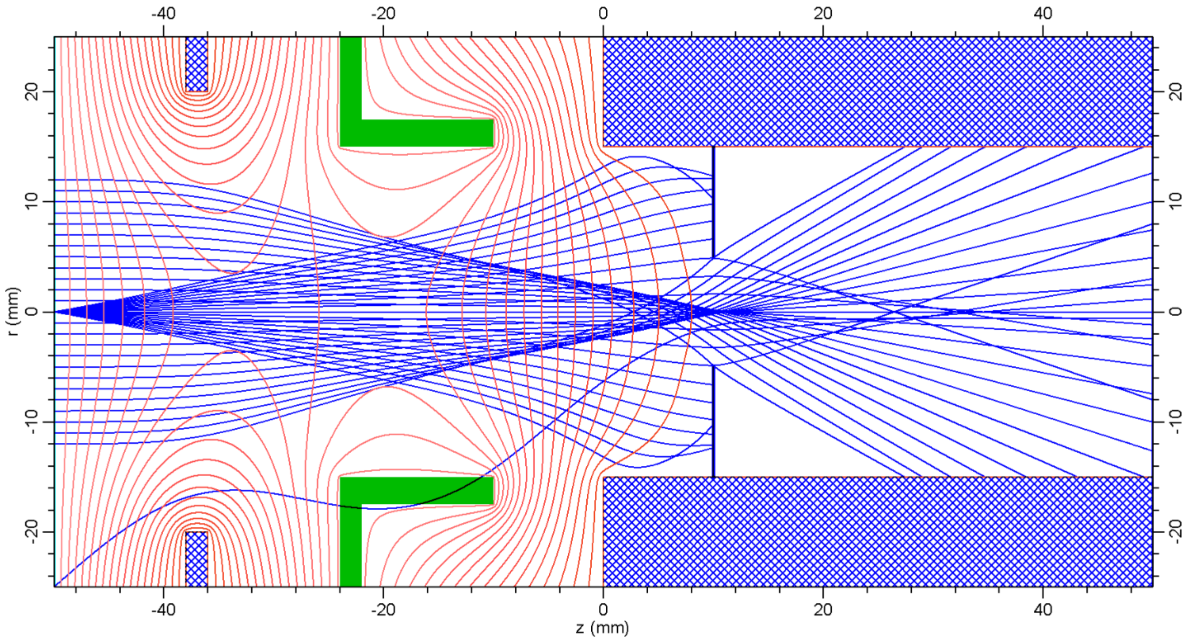

Figure 6: Design of an electrostatic axially symmetrical transfer lens: the ion source is on the left-hand border and is at the potential of -21 kV, the focusing electrode (green) is at -18 kV, and the blue parts are at the ground potential. The trajectories are plotted for Th^{4+} ions at the initial energy of 84 keV. The trajectories parallel to the axis do not meet in an axial focal point due to geometrical aberrations and the excessive source size. Similarly, the trajectories starting from the axial point exhibit a large aberration caused by the widespread in the starting angle. The voltages can be scaled so that the optimum lens action is achieved for different initial energy ranges.

The approach of using direct loading without buffer gas is burdened with a high uncertainty in the kinetic energy (up to 84 keV), direction, and charge state of the recoil ions. Several concepts of focusing optics (static and triggered-dynamic) for Th^{4+} have been investigated by ISI but found low overall efficiency (below 1%) even in the most optimistic scenarios, leading to an expected ion yield below 1 s^{-1}. Seen the successes of buffer-gas based approaches, e.g. at cooperation partner LMU Munich, this approach was not pursued for the experimental system developed at PTB. Instead, helium is used as a buffer-gas to reduce the kinetic energy of the ions. The stopping range of $^{229}Th^{3+}$ in 1000 Pa of He is a few centimeters. The ionization potential of He is 24.6 eV, above the ionization potentials of Th^{3+} and Th^{2+}, so that these charge states should be preserved, while higher charge states that are originally present from the recoil ion source undergo charge exchange.

To realize the aim of an ultrahigh vacuum in the ion trap section of the vacuum system, the crucial element in the PTB setup is a gate valve to physically separate the buffer gas source region form the ion trapping region. After loading of the ion trap for a few seconds, the gate valve will be closed, restoring UHV conditions within a few seconds and enabling long trapping times for Th^{3+}, provisionally extending over several minutes.

A sketch of the $^{229}Th^{3+}$ loading system and ion beamline is shown in Figure 7. The first sections have been developed in cooperation with LMU Munich and JGU. The text refers to the labelling in the drawing. As in the first stage, the ^{233}U source is located (1) in a helium buffer-gas environment (about 3000 Pa). Here, the ^{229}Th ions, and all other isotopes and charge states out of the decays from the source are decelerated via the interactions with the buffer gas. An RF ion funnel (2) next to the source collects and guides all ions towards a de Laval nozzle (3). The nozzle opens the only connection from the buffer-gas cell to the downstream vacuum region, acting as a first differential pumping stage. Here the ions are injected into a first RF quadrupole ion guide (4). A gate valve (5) with integrated ion optics bridges the quadrupole (RFQ) with the second vacuum chamber for differential pumping. The chamber contains a quadrupole mass filter (5) to select the $^{229}Th^{3+}$ ions out of the source background of different isotopes, charge states and molecules. An electrostatic deflector (7) behind the mass filter bends the ions at right angle towards the ion trap (8). This deflector provides access on two optical axes to the ion source and ion trap. The deflector is contained in the last section for differential pumping in front of the trap. To minimize the losses on $^{229}Th^{3+}$ ions, all parts of the system use UHV compatible materials, excluding all organic elastomer or polymer compounds because of the high reactivity of Th ions with carbon hydrides. Special care has also been given to the purity of the buffer-gas. For that, the helium gas supply is connected to a gas-purification system including a commercial heated getter gas purifier and a liquid nitrogen cryo trap. The system has been built and characterized at PTB. A flux of $^{229}Th^{3+}$ of about 10 ions per second is available for trapping and spectroscopy measurements. The objective has been successfully achieved.

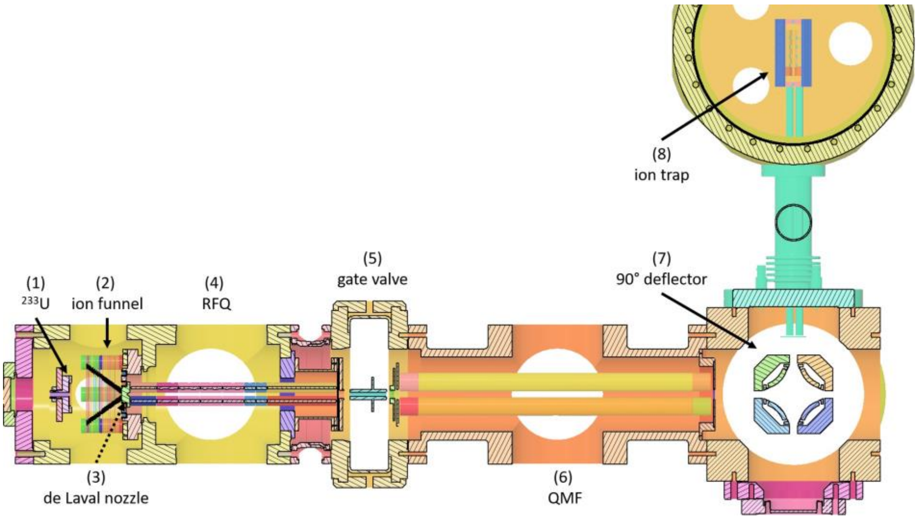

Figure 7: A schematic overview of the ion beamline for loading of $^{229}Th^{3+}$ from a recoil ion source. A detailed description is given in the text.

4.5 Transportable equipment for laser cooling, high resolution spectroscopy and precision optical frequency measurements

This objective was targeted specifically at enabling joint experiments based on high resolution laser spectroscopy and high precision optical frequency measurements at nuclear physics laboratories that so far did not possess a direct connection with optical metrology laboratories. The objective was motivated by research directed towards a ^{229}Th optical nuclear clock. The aim was to establish a transportable laser infrastructure for laser cooling of thorium ions and to investigate sympathetic cooling of thorium ions for high resolution laser spectroscopy. Furthermore, suitable excitation sources towards direct nuclear laser excitation are investigated in the vacuum ultraviolet (VUV). As a prerequisite for frequency measurements, a subharmonic of the VUV frequency had to be linked to the SI standard. For this purpose, a fiber link has been established and characterized.

The direct laser excitation of the ^{229}Th nuclear clock transition requires a laser source in the VUV regime at a wavelength shorter than 200 nm. At the start of the present project, the transition wavelength was expected to be around 160 $\pm$ 10 nm based on results from electron and gamma spectroscopy. Due to results obtained during the course of the project by TU WIEN and cooperation partners, this target wavelength was corrected and refined to around 150 $\pm$ 3 nm. The design of the laser system was adapted accordingly in the duration of the present project. One of the main reasons for using a nuclear transition as an optical frequency is the narrowness of the spectroscopic line, expected in the sub-millihertz range (for a single Th-229 ion). As a consequence, the linewidth of the interrogation should also be narrow. Within the project the main approach to produce laser light at 160 nm (later 150 nm) was high-harmonic generation (HHG) of infrared femtosecond pulses.

This approach succeeded at TU WIEN in realizing a VUV frequency comb, covering the target range (160 $\pm$ 25 nm) using the 5th harmonic of a 800 nm TiSa femtosecond laser. We implemented the currently only VUV frequency comb system using solid-state HHG and demonstrated a frequency uncertainty of 2 MHz, corresponding to a relative uncertainty of 10^{-12}. Due to the complexity of the approach, the system is not yet transportable. However, as the primary IR frequency comb is stabilized to a local 10^{-13} RF reference (a Rb clock combined with a GPS-disciplined quartz), we can locally realize a 10^{-12} accuracy with this system.

A phase stable optical link between the BEV-PTP and TU WIEN is currently being established as part of a trilateral remote connection to a precision frequency reference at BEV-PTP that connects to ISI and that has been implemented in the project. The ISI, and BEV-PTP have set up an experimental system for remote synchronization of stable laser oscillators at a wavelength of 1542 nm, which is necessary for the stabilization of a spectroscopic laser for direct excitation of the Th-229 nuclear clock transition. This system is based on two independent highly-stable laser sources with a spectral line-width of less than a few Hz. At the same time, the stability of their optical frequencies is ensured using optical frequency combs (OFC), which are synchronized by active hydrogen masers (H-maser). Because the institutions ISI at Brno, CZ and the BEV-PTP, TU WIEN, AT are more than 150 km apart, a long-haul optical fiber link has been set up, which ensures the transmission of an optical frequency at the wavelength of 1542 nm.

To ensure the transmission of an optical frequency of 1542 nm between ISI and BEV-PTP, an optical fibre link with a length of 232 km had to be established. For this purpose, the cooperating partner CESNET, CZ (collaborator) was introduced to the cooperating work. CESNET operates transmission optical fibres in the Czech Republic as well as cross-border fibres (e.g. connections between Austria and the Czech Republic). Dark channels on the production communication fibre Austria - Czech Republic were used for the connection between ISI and BEV-PTP. CESNET fitted this fibre path with the necessary optical band-pass filters and equipped the 1542 nm channel with bidirectional amplifiers to compensate for the attenuation of the long-haul fibre link. Subsequently, the dedicated channels of the link were set to bidirectional operation. The inserted optical amplifiers were adjusted very precisely, so that spontaneous oscillations did not occur. The fibre link prepared in this way was subsequently equipped with BEV-PTP and ISI optical components. Furthermore, new electronics developed in ISI were involved in the control and induced fibre noise compensation of the link.

The ISI and BEV-PTP fibre link is the first operational segment of the planned delocalized composite clock network infrastructure – a coherent fibre link in which a stable optical oscillator (Hz linewidth) is disseminated between ISI and BEV-PTP and TU WIEN – Atominstitut (additional 24 km). In pilot experiments, BEV-PTP and ISI have investigated a remote beat note of two independent highly coherent laser sources working at 1542 nm. A 1542 nm low-noise laser locked via an optically referenced optical frequency comb to a 1540 nm cavity-stabilized laser (ISI custom design) at ISI is being transmitted from ISI to BEV-PTP over this Doppler-compensated fibre link. In BEV-PTP, the beat note between the link output and a local 1542 nm cavity-stabilized laser (Menlo Systems ORS) is measured. The fibre noise cancellation setup uses a general scheme based on a Michelson interferometer with compensation and end shifter AOMs. The fibre noise cancellation electronics is an ISI’s custom design based on an analogue I/Q demodulator, a loop filter realized by a digital signal processor (DSP), and a direct digital synthesizer (DDS) driving the compensation AOM. A DDS+PLL card for fibre links and complex electronics for Doppler induced phase noise compensation for fibre links has been developed. The DDS+PLL card incorporates a new microcontroller for a broader bandwidth of servo-loop control as well as the new written firmware for fibre link phase noise compensation.

The optical fiber link ISI and BEV-PTP has been benchmarked (comparison of two local optical reference cavities) and results have been submitted as a joint publication (ISI, BEV-PTP, TU-Wien). The fractional stability of 10^{-15} between the hydrogen masers was determined. The link provides reliable phase coherence with a fractional stability of 7×10^{-18} in 1000 s. Operation over 1 week was demonstrated.

BEV-PTP’s task has been to provide the reference laser for TU WIEN’s transportable laser system for direct nuclear excitation of Th-229. A transportable external cavity laser – ORS by Menlo Systems – was acquired and characterized. Additionally, BEV-PTP is providing traceability to the SI second (H-maser, Cs clock) via this reference laser for frequency measurements at TU WIEN. For this traceability chain an uncertainty budget was estimated.

PTB has contributed to the efforts directed versus ^{229}Th nuclear laser spectroscopy with the development of laser systems for the laser cooling and spectroscopy of electronic transitions of Th^{3+} ions and with a thorium ion trapping system. The cooperation of TU WIEN and PTB, together with partners at LMU Munich and at the University of Delaware has successfully obtained follow-up funding for joint work from a synergy grant of the European Research Council ERC. The objective has been successfully achieved.

5 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.

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