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1 Executive Summary

Introduction

Clocks with the highest accuracy are needed for major applications such as the realisation and dissemination of the SI second, the realisation of international atomic time, global navigation satellite systems, and the exploration of fundamental physical laws and phenomena. Better clocks improve these applications and make possible novel applications such as chronometric geodesy for earth science. Reaching the full potential and best exploitation of the new generation of optical clocks is hindered by the clock stability, i.e. by the speed at which measurements are done and the time needed to reach a given measurement uncertainty. The project improved ultra-stable laser technology and performance, with decreased uncertainties, and with this demonstrated state-of-the-art clock stability. The project demonstrated the use of quantum engineered states and methods, like for instance, non-destructive atomic detection, that can improve stability even further while offering other benefit and possibilities. Advances made by the project are also applicable to other atom sensors.

The Problem

The best atomic clocks today have an accuracy around 1 part in 10¹⁸ and short term instabilities of 10⁻¹⁵ over one second, improving with the square root of the averaging time. Accuracies in the low 10⁻¹⁸ range are expected in the near future. However, investigating physical phenomena, understanding associated shifts of the clock frequency and using clocks at the 10⁻¹⁸ level is hindered by the prohibitively long time needed to measure shifts and to compare clocks at this level. One limit to the clock stability comes from the ultra-stable laser which is used to interrogate the atomic transition. The impact of the laser is also related to the amount of dead time in the clock measurement cycle. Further to this is the quantum projection noise which impacts the detection of the atomic state. If an atomic ensemble are used, and not only a single atom or ion, one can make use of quantum entangled states between several atoms to pass this quantum projection noise limit and thus improve clock stability and performance. The project studied how to create and use such states so as to be beneficial to clocks and atomic sensors. Depending on the specific type of clock, or situation, this required us to develop novel technologies, devices and methods, to make the creation and use of quantum engineered states compatible with the many other requirements applicable to physical systems for clocks and atomic sensors.

The Solution

Among many possible axes of research to tackle the above problems, we investigated the following aspects. We investigated reducing the impact of thermal noise in ultra-stable optical resonators, by implementing long cavities and by developing crystalline coating technology. We also investigated reducing the impact of laser noise on clock and clock comparison stability by designing new interrogation protocols. We investigated non-destructive detection schemes for neutral atom clocks. We demonstrated novel clock interrogation protocols that use such non-destructive detection to mitigate the effect of laser noise. We studied how to bring such non-destructive detection into the quantum regime where the detection can be used to generate and use quantum entangled states. We studied other means to achieve these goals using interactions rather than non-destructive detection. In ion clocks, we investigated means and technologies to shift from single-ion to multi-ion clocks, and in this case, to generate and use quantum entangled states between multiple ions. The project has therefore demonstrated several new key enabling quantum engineering methods for improving optical clock stability and also accuracy in a variety of systems.

Impact

The project gave 23 scientific publications in high-ranking peer-reviewed journals. These publications report novel results, novel devices and proofs of concept that open many possibilities for further research, as well as developments and applications for a wide community working on clocks and atomic sensors.

The project led to new metrological capabilities within the NMIs of Europe, several of which have enhanced their operational measurement capability for optical frequency standards. New ultra-stable lasers with stateof-the-art stabilities were developed which in turn improved the stability of optical clocks at PTB. At LNE-SYRTE, a novel non-destructive detection method was implemented on a fully operational optical lattice clock. In the near future, this also will enable improvement of clock stability. Similarly, for the high yield manufacturing of micro-fabricated ion traps and the high fidelity interaction developed at NPL will facilitate



the development of multi-ion clocks as operational frequency standards. These improved capabilities are and will be used in coming optical fiber link inter laboratory comparisons which keep European laboratories at the forefront of the work toward a redefinition of the second based on optical frequency metrology.

The outcome of the project was largely disseminated at international conferences. A dedicated workshop was also organised to further augment the impact. Project members gave many lectures and conducted several other activities for the training of students, PhD students and early stage researchers.

The project contributed to the development of crystalline coating technology which supported the development of new innovative products and product lines. The project thereby contributed to the growth of a start-up company and already had and will continue to have direct economic impact. Knowledge gained by the project will feed the many collaborative R&D project that project partners have with industry (defence, space, etc.), creating further economic impact.

Over the long term, improvement of clocks and atomic sensors will positively impact existing applications of these devices and will enable novel applications, notably in earth science. Improvements in optical clocks will be key to the future redefinition of the second to the international system of units. The outcome of the project was and will be used to define international and European regulations and policies in this direction by the means of participation of several project members, both as individuals and as organisations, to committees of the CIPM, of the Joint Research Center, and of the Flagship Program on Quantum Technologies, etc.



2 Project context, rationale and objectives

Clocks with the highest stability and accuracy are needed for major applications such as the realisation and dissemination of the SI second, the realisation of international atomic time, global navigation satellite systems, and the exploration of fundamental physical laws and phenomena. Improvements in clock accuracy and stability will improve all these applications. Further, the increase of clock accuracy and stability in the low 10⁻¹⁸ range or better could also lead to new applications for clocks, for instance in geodesy and Earth science.

At the start of the project, the best atomic clocks had accuracy around 1 part in 10¹⁷ and short term instabilities of 10⁻¹⁵ over one second, improving with the square root of the averaging time. Accuracies in the low 10⁻¹⁸ range were expected in the near future. However, to practically demonstrate and benefit from such an unprecedented accuracy and to even evaluate the possible frequency shifts, with the current instability an averaging time of 10⁶ s (i.e. nearly two weeks) would be needed for a single measurement at 10⁻¹⁸ resolution. This prohibitively long time severely hindered the development and application of these clocks.

In state-of-the-art systems at the start of the project, the stability was limited by two main causes, depending on the type of system. One limitation was the standard quantum projection noise, which is proportional to the square root of the number of particles. The second limitation was the so-called Dick effect that arises due to combined effects of laser frequency noise and of large dead time in the probing sequence. The JRP addressed techniques with the potential to overcome these limitations by the introduction of quantummechanical entanglement to this field.

Beyond clocks the research undertaken within the JRP was also beneficial to several other atom-based sensors that can suffer the same type of limitations. This is notably the case for inertial sensors, such as accelerometers, gravimeters and gyrometers, based on atom interferometry. These sensors can be limited by the same standard quantum limit as optical clocks. They are also often limited by the noise caused by vibration, acceleration or rotation, which are reminiscent of the effect of the local oscillator noise affecting optical atomic clocks. The connection with this area of physics and metrology was made through the REG(IOTA) and this ensured that the needs of this field were considered in the JRP.

Similarly, the JRP addressed some of the needs that are shared with the field of quantum information processing and computation. The QIPC roadmap 2010 (<u>ftp://ftp.cordis.europa.eu/pub/fp7/ict/docs/fet-proactive/press-12_en.pdf</u>) stresses in section 4.4.1 "*Chip traps for quantum computing*" the need for "finding a trap technology that features small heating rates and long coherence times, and a trap design that allows for transport of the ions (along with their contained quantum information)". In section 1.1.4 it states "devices realising quantum algorithms with up to 10 qubits" as a 5 year goal. These statements show that the technology developments planned within this JRP were also highly relevant to the QIP community. The connection with this area of research was made through the REG(OAdW) and this ensured that the needs of this field were considered in the JRP.

The overall aim of the JRP was to identify, develop and implement methods that can improve the short term stability of optical atomic clocks beyond the limitation imposed by the standard quantum limit and the so-called "Dick effect" associated with the probe laser frequency noise. For that purpose, the JRP has defined the following specific objectives:

- Objective #1: To investigate the generation of entangled states using **interactions in neutral atom** systems. Two related approaches will be considered: one based on exciting the resolved interaction sidebands in lattice-confined atoms, and one based on spin dynamics in quantum gases.
- Objective #2: To investigate the generation of entangled states using **interactions in trapped-ion** systems. Initially quantum gates based on Coulomb and laser pulse interactions to generate these states will be used. Then states with small numbers of ions (N=2 to 6) yielding high coherence time and the possibility to cancel systematic shifts will be investigated.
- Objective #3: To investigate the generation of entangled states by performing **measurements in neutral atom** systems. Both the strong measurement regime and the weak measurement regime with fast feedback to the quantum system will be studied.
- Objective #4: To develop the **necessary technologies** for the above tasks. This includes low noise detection systems with a noise floor well below the standard quantum limit (see earlier), ion traps



with a low heating rate that can hold multiple ion coulomb crystals, hardware and software for fast feedback action on the quantum system, non-destructive detection systems, etc.

- Objective #5: To **characterize these entangled states**, notably in terms of the degree of quantum correlation and coherence lifetime.
- Objective #6: To **design and implement protocols** where the use of the above methods can improve the spectroscopy and the clock stability. The design will include consideration of the potential impact, positive or negative, of these protocols on accuracy.
- Objective #7: In one or several of the systems under study, the **spectroscopy and stability beyond that imposed by the Dick effect** for the same system will be demonstrated. With transportable grade probe lasers, improvement of the stability by a factor of 10 or more beyond the Dick limit is expected.
- Objective #8: In one or several of the systems under study, **spectroscopy and stability beyond that imposed by the standard quantum limit** will be demonstrated, for the same system in the same configuration. This is the more exploratory part of the JRP and therefore the more difficult to quantify. A gain of 3 or more in stability is possible, which corresponds to a shortening by 10 of measurement duration to reach the same resolution.



3 Research results

The research was conducted on a diverse array of system types and through a range of experiments. In this section, the work is described for each type of system experiment. At the beginning of each description there is an explanation of how the work connects to the high level goals of the project and to the transversal objectives stated in Section 2.

3.1 Ion trap microfabrication

As described previously in Section 2, developing novel technologies is required for several studies of entangled state generation planned in this JRP. It is generically gathered under objective #4 of the project. This objective applies to the case of studies based on trapped ions. The work reported in this Section 3.1 focused on technologies to grant availability of micro-fabricated traps that can be produced in quantities and that can hold ions with minimal perturbation of their state of motion. This work was undertaken by NPL.

We have developed high yield parallel fabrication of micro-fabricated ion traps on 4 inch wafers. We have developed methods to test and characterize such traps, and assess their suitability for optical clocks. More specifically, the following aspects were studied:

Fabrication

- Parallel micro-fabrication process now developed properly for 4-inch wafers (in partnership with niche-volume foundry)
- Automated electronic packaging is provided by packaging company
- Recent round: 2 wafers processed, each with 36 chips, and 80% geometric yield achieved.
- 20 chips packaged and a further 30 available for packaging
- Another round of fabrication and packaging is in progress: expect 50 trap chips with yield >90%, due June 2017

Electrical test

- Electronic testing on a set of 10 packaged chips from wafer 1: breakdown voltage varied but a value up to 500V at ~15 MHz was achievable. Limits likely due to Si residue from dicing process.
- Wafer 2: chips diced with care to make them free of Si residue and awaiting electrical test.
- Next generation wafers (in progress will have no need for dicing, so residue problem eliminated).

lon test

- One of the ten fabricated traps operates up to 300 V amplitude at frequencies from 8 to 23 MHz with no breakdown.
- Initially operating parameters for ions are 16.4 MHz and 150 V amplitude.
- Loading ions in dedicated zone and transporting them to experimental zone works well
- Preliminary investigation of motional heating rates with the Doppler recooling method showed that after 1 minute of uncooled operation there was no sign of heating. Result should be consistent with <=100 quanta/s for 88Sr+ at 400 kHz axial and 2.2 MHz radial motional frequencies.
- More sensitive measures using optical spectroscopy of S-D transition are in progress.
- Measurements using sideband cooled ions will be used thereafter.
- Lifetimes look to be excellent, but a more thorough test is required.

3.2 Toolbox for ion qubit manipulation

As described previously in Section 2, developing novel technologies is required for several studies of entangled state generation planned in this JRP. It is generically gathered under objective #4 of the project. This objective applies to the case of studies based on trapped ions. The work reported in this Section 3.2



focused on technologies to create high fidelity, long term stable interaction between pulsed laser beams and trapped ions, as prerequisite for more complex laser-ion interaction sequences needed to prepare entangled states in these systems. Another objective, objective #2 of the JRP, is to investigate generation of entangled states using interactions in trapped-ion systems. Another aspect of the work reported in this Section 3.2 involved implementing simple forms of single ion and string-of-ions operations which are building blocks of such schemes. The aim is to evaluate and optimize the fidelity of such gates. This work was undertaken by NPL.

We have developed and tested methods for high reliability qubit manipulation aimed at multi-ion clocks. This includes:

- An agile digital frequency synthesizer
- A laser beam pointing and power stabilisation system
- Putting together and using these two technologies, we have studied their impact on single qubit manipulation fidelity
- Ion string splitting/shuttling whose reliability was tested and optimised.

3.3 Non-destructive detection of lattice trapped strontium atoms

As described in Section 2, one possible path to generating entangled states useful for clocks relies upon non-destructive atomic detection. The objective #3 is to investigate this approach in neutral atom systems. The work reported in this Section 3.3 focused on the case of optical lattice clocks. It started with developing a novel technique for non-destructive detection adapted to such clocks, namely lattice trapped strontium atoms. As this technique is generically applicable to other atoms and to a variety of other experiments and other situations, this contributes to objective #4 of the JRP. Then, the technique was applied and studied on atomic samples. This work is undertaken by OBSPARIS and LNE.

The work reported in this Section 3.3 relates to the experimental realisation of a new detection scheme for measuring the transition probability in optical lattice clocks. Usually, the transition probability in neutral atom clocks is deduced from a measurement of the number of atoms in the fundamental clock state by observing the fluorescence emitted by the atoms under the influence of a bright resonant laser beam. The main drawback of these fluorescence schemes is that atoms scatter photons in all directions, making it hard to collect all the available information, or to amplify the emitted information with an optical resonator. The aim of this work is thus to demonstrate a new detection scheme that make use of the dispersive properties of the probed atoms. For this, a weak and focused off-resonance laser beam is sent on the probed atoms, and an interferometric system is implemented to measure the phase shift induced by the atoms of the laser beam. In such dispersive detection schemes, all the information resulting from the light-atom interaction is gathered in a small-volume Gaussian laser mode, that is completely and optimally (with respect to the photon shot noise limit) sampled by the interferometric detection. Because of this, they offer many advantages for optical lattice clocks:

- The signal-to-noise ratio is enhanced, making the detection noise negligible with respect to other noise sources that may limit the frequency stability (quantum projection noise, Dick effect).
- This signal-to-noise ratio can be further amplified with an optical resonator (cavity). In this configuration, the multiple round trips in the cavity enhance the atom-light interaction, yielding an improvement on the order of the square root of the cavity finesse.
- The power sent to the atoms can be small enough for the atoms to scatter less than ~ 100 photons, in which case they stay trapped in the optical lattice-clock. This "classical" non-destructivity regime can be used to reduce the Dick effect currently limiting the frequency stability of optical lattice clocks.
- In optical lattice clocks, the atoms are confined in a small region of space, making it possible to reach high optical depths, suitable to reach a detection noise in the "quantum regime", *i.e.* smaller than the quantum projection noise, for a number of scattered photons per atom less than one. With these parameters, the quantum coherence of atoms is partly preserved in the detection process, making it possible to implement spin-squeezing protocols to demonstrate a frequency stability



exceeding the quantum projection noise limit. Reaching a signal-to-noise ratio suitable for these protocols was therefore a major objective of the project.

The research developed to fulfil these objectives consists in the implementation, in an actual optical lattice clock system, of a cavity-assisted, non-destructive detection scheme. The goal was to reach a signal-to-noise ratio in the quantum regime. For this, a new vacuum system for one of the Sr optical lattice clocks available at LNE-SYRTE has been designed and assembled. It comprises a bi-chromatic optical cavity, used for both trapping cold strontium atoms and performing the non-destructive detection. The high finesse of the cavity (F = 16000) at the detection wavelength yields a theoretical shot noise limited sensitivity of 3 atoms for a single scattered photon. Putting together a metrological-grade clock with such a cavity-based scheme requires **solving several challenges:**

- The wavelengths at which the cavity is operated are constrained: the trapping laser has to be tuned to the "magic wavelength" required for optical lattice clocks, and the detection laser has to be tuned close to the ¹S₀ ¹P₁ transition. This implies finding strategies to realize a homogeneous coupling between the atoms and the cavity and to locate an operable length for the cavity.
- The geometry of the cavity has to be compatible with the required geometry of a high accuracy optical clock. In particular, the cavity has to leave space for all the various laser beams used to manipulate and probe the atoms. Also, the PZT actuators used to tune the length of the cavity have to be efficiently shielded in order to avoid the systematic frequency shift associated with an external static electric field. This results in a cavity that has an extended mechanical support which is prone to vibrations.
- The detection scheme has to be robust enough to allow for operational measurements of the clock over extended periods of time, and not just for a short proof-of-principle experiment.

To address these challenges, we demonstrated **a novel heterodyne optical protocol** to measure the intracavity phase shift induced by the atoms. It consists in measuring the reflection by the cavity of a phase modulated signal whose second-order side-bands are both injected in consecutive modes of the cavity. It combines several critical advantages:

- It provides for an error signal to lock the probe laser frequency on the cavity, in order that the probe laser is always injected, without requiring an independent laser, as in many cavity schemes. This error signal is then processed by a fast digital PID based on a FPGA. The design of such a fast system was required to accommodate with the large excursions of the cavity resonances due to the cavity geometry and finesse.
- Another output of the heterodyne scheme provides the atomic signal. Using parity properties of the atomic dispersion and of the phase modulation, this signal only contains the phase shift due to atoms and not the phase shift due to residual technical noise (cavity length fluctuations or laser frequency noise), making the detection noise immune.
- Finally, the two second order side-bands injected in the cavity form a set of two lattices out of phase, resulting in a homogeneous laser power along the cavity axis. This ensure that all the atoms are equally coupled to the cavity, and hence equally contribute to the signal. In addition to the experimental implementation of the detection scheme, we also developed a theoretical analysis of the shot noise limit that shows that the detection optimally collects the cavity-enhanced information delivered by the atoms.

The experimental demonstration of this heterodyne protocol enabled us to detect lattice trapped atoms in the SrB optical lattice clock, as displayed in Figure 3.3.1. It reaches a resolution of 3.7 atoms for 38 scattered photons per atom, which is in the "classical non-destructivity regime". Minor technical improvements will enable us to inject less power in the cavity, and reach the "quantum non-destructivity regime". Technical details about the detection can be found in [Vallet2017].





Figure 3.3.1: Atomic signal acquired with the nondestructive detection.

In conclusion, we have built a novel detection scheme for optical lattice clocks, which shows a very high signal-to-noise ratio, compatible with the generation of spin-squeezed states for quantum metrology. This detection will allow both for classical enhancement of the frequency stability (through the reduction of the Dick effect allowed by the atom recycling and short loading allowed by this detection), and quantum enhancement of the frequency stability beyond the quantum projection noise limit.

Research results are the following:

- Experimental realization of a novel detection scheme to measure the transition probability in optical lattice clocks with unprecedented sensitivity
- Investigation of a novel heterodyne optical system to homogeneously probe atoms in a linear cavity.
- Development of a system in principle able to produce quantum engineered states of the atoms in optical lattice clocks

3.4 Non-destructive detection of trapped rubidium atoms

The work reported here relates to the same high level objectives #3 and #4 as in the previous Section 3.3. Here, instead of strontium and optical transition, the system under study is based on rubidium and the microwave transition. In this case, non-destructive detection directly addressing the atom number difference between two states, rather than the atom number of a single state is possible. The measure quantity is more readily matched to usage in clocks. It is a good model system to test new schemes. And at the same time, rubidium is widely used in commercial clocks and in atomic inertial sensors. In the work reported in this Section 3.4, such detection is developed and thoroughly characterised in terms of trade-off between detection sensitivity and detection destructivity. Also, technologies for fast actuation on the atomic system based on the information of the non-destructive detection was developed. The work was undertaken by REG(IOTA).

In this experimental setup, cold atomic ensembles of rubidium 87 are optically trapped in a four mirrors cavity in a bow-tie configuration, where they show a lifetime of more than 10 s. The atomic clouds are coherently manipulated using microwave pulses resonant with the hyperfine transition at 6.8 GHz; in that way it is possible for example to induce Rabi oscillations between the F=1 and F=2 hyperfine levels of the $5^2S_{1/2}$ electronic level. The common approach to measure the system evolution is to shine the microwave radiation for a given interval of time ΔT , measure the phase of the oscillation in terms of the population unbalance between the two hyperfine states, and repeat the sequence with a new atomic cloud for different ΔT so as to



plot the fringe. The method is clearly time consuming. We adopted a different approach, based on a nondemolition measurement of the population unbalance. To this aim we realize a heterodyne probe, which uses a carrier beam at 780 nm phase modulated so as to generate two frequency sidebands. The carrier is frequency referenced to the D2 transition of rubidium, is such a way that each of the two sidebands dispersively probes the population of one of the two levels on which the atomic system evolves. The population unbalance signal is obtained by demodulating the optical beat generated by the three frequency components on a fast photodiode. In this way we could monitor in real-time the evolution of an atomic system, undergoing Rabi oscillations or a Ramsey interrogation, causing only a very limited and tunable destructivity of the atomic coherence at each interrogation.

With the target of implementing measurement-and-correction schemes, we considered the problem of optimizing the non-destructive detection on the trapped Rb atoms, which translates into quantifying and maximizing the ratio between the information obtained with each measurement and the destructivity it causes. To this aim we realized a series of measurements where the atomic ensemble in a well-defined initial state is perturbed by noise; the non-destructive probe tries then to measure the effect of such noise; finally a correction is applied to regain the initial atomic state. The noise is implemented as a microwave pulse that changes the initial spin direction in an unknown direction and/or angle. The efficiency of the noise protection protocol is characterized versus the strength of the probe. In this way it was possible to reveal the competition between extracting more information from the atomic system and preserving its coherence. A very weak detection leaves the atomic state unperturbed, but does not help to choose the right steering direction to recover its initial state. On the other hand, a stronger pulse fully characterizes the atomic states after the noise action, but destroys as well the quantum resource represented by the atomic coherence. The protocol efficiency shows a maximum in between, as a result of the trade-off between the two competing effects, as shown in [Vanderbruggen2012]. The behaviour of the measurement-and-correction scheme was then characterized with respect to several parameters of the experiment, such as the number of atoms in the cold ensemble, and the properties of the noise applied to the atoms [Vanderbruggen2014], obtaining a very good agreement with what expected theoretically.

In these measurements the evaluation of the noise effect and the successive correction is done via a microcontroller with embedded data converters: it reads out the probe signal and sets the correction microwave pulse via RF switches and phase shifters on the signal at 6.8 GHz. The low resolution of the data converters is at the basis of the rather high level of technical noise of the trade-off measurement. A FPGA based feedback system has been later implemented to increase the performance of the measurement-andcorrection loop. The commercial system (Red Pitaya) uses a Xilinx ZC7Z010 system-on-chip (SoC), connected to fast, high resolution data converters.

3.5 Improving non-destructive atomic detection with squeezed light

As described in Section 2, one of the high level objective #6 of the JRP was to design and implement protocols that can improve atomic spectroscopy, clock and atomic sensors. In non-destructive detection schemes, like the ones described in Sections 3.3 and 3.4, laser light crossing the atomic sample is probed to gain information on the atomic state. This light detection itself has quantum fluctuations that limit the sensitivity and the potential of the method. Looking forward to the future, one can ask how to improve on this limit. In the work reported in this Section 3.5, this question was investigated theoretically. Specifically, we studied whether and how light with tailored quantum properties can give a benefit to non-destructive atomic detection schemes. The work reported in this Section 3.5 was undertaken by INRIM, with inputs from OBSPARIS and REG(IOTA) on parameters and configurations relevant for this study.

The aim is to propose a quantum strategy affordable by current technology, overcoming the photon shot noise due to intrinsic quantum fluctuations of optical probe, for non-destructive measurement (phase shift measurement) of atomic clocks.

We considered first the use of a squeezed beam together with a strong classical coherent beam injected into the input ports of an interferometer. It has been recently demonstrated that this strategy allows the reaching of the fundamental bound to the phase sensitivity in case of non-negligible decoherence and losses (losing some photons is unavoidable). The configuration is similar to the one of the quantum enhanced gravitational wave interferometer demonstrated by LIGO consortium. Only one port of the interferometer is read out and homodyne detection is not necessary to enjoy quadrature squeezing benefit.



In order to build a general model describing the interaction of atoms with quantum light in the interferometer we consider the interdependence of the photon losses and the phase shift by Kramers-Kronig-like relation. The specific value of the parameters and the details of such a model has been provided by the INRIM time-frequency group and by the OBSPARIS. We have introduced in the model either the parameter of the Ytterbium blue line or the Strontium blue line.

The fact that losses and the phase shift are in general related (for example both depend on the detuning from the clock transition and the number of atoms) on the one side, and the fact that losses (and unbalancing of the arm) play a detrimental effect on the quantum enhancement make the research of the optimal detuning an interesting and unexplored problem. We have introduced all these effects in a simulation. The result is that the uncertainty in the determination of the number of atoms by the measurement of the phase shift can be reduced by more than 3 times using current technology (more than 13 dB of squeezing is currently available).

Furthermore we explored the applicability of a novel scheme based on the covariance measurement between two different and independent interferometers both interacting with the atomic ensemble. In principle this scheme could allow to suppress noise from independent sources (included shot noise). With the use of quantum correlation (like the so called two-mode squeezing) this effect should be stronger. However after long calculation we have shown that in realistic condition this setup cannot provide significant advantage in the uncertainty reduction with respect to the single interferometer injected with single-mode squeezed beam described previously.

3.6 Using interactions to create spin-squeezed states in rubidium condensate

Objective #1 of the JRP is to generate entangled states using interactions in neutral atom systems. In this Section 3.6, we describe the work undertaken in rubidium Bose-Einstein condensate. In this system, trapped atoms are cooled to ultralow temperatures where the many atoms in the sample tend to condensate into the same single quantum state. Such systems support strong interactions between atoms which can be controlled so as to create quantum entangled states of many atoms. The aim of the work is to design a control sequence yielding states with the highest metrological relevance and to characterise and quantify their usefulness (objective #5). This work was undertaken by REG(LUH).

Atom interferometric measurements are fundamentally limited by the Standard Quantum Limit (SQL). A measurement precision beyond the SQL requires the generation and application of entangled states. Entangled states of ultracold atoms have been created with Quantum Non-Demolition measurements, Cavity backaction, and Kerr-like collisional interaction. These methods allowed to create spin-squeezed samples, where the relevant spin uncertainty was reduced to below the shot noise. Spin dynamics in Bose-Einstein condensates is an alternative method for the generation of entangled states, which was pioneered by REG(LUH). Within QESOCAS, the method was supposed to be further advanced and possible applications towards the measurement of time were to be explored.

The metrological gain of two different quantum states was studied: Twin Fock states and spin-squeezed states. The creation of entangled states in different modes of the external potential was model by a Bogoliubov analysis [Scherer2013]. It was shown that spin dynamics creates a so-called two-mode squeezed vacuum states, which is a superposition of Twin-Fock states. Within QESOCAS, we could demonstrate that the created Twin-Fock states contain many-particle entanglement [Lücke2014]. A generalized spin squeezing parameter of 11.4 dB was obtained, the largest value at the time for any atomic system. Furthermore, the metrological gain was estimated from the measurement results [Apellaniz2015].

A main objective of REG(LUH) was the generation and application of spin-squeezed states which have not been generated with spin dynamics before. In 2015, we were able to push the novel method of atomic homodyning down to the single-particle level [Peise2015]. This offered new possibilities to study the created entangled states. Atomic homodyning was applied to the two-mode squeezed vacuum state and allowed for the demonstration of Einstein-Podolsky-Rosen entanglement [Peise2015a]. The homodyning data were exploited to reconstruct the full density matrix of the many-particle state. The created two-mode squeezed vacuum state had a fidelity of 78%.



In summary, spin dynamics was shown to offer the reliable production of squeezed vacuum states. Squeezed vacuum states present a valuable resource to improve the metrological sensitivity of atom interferometers, most prominently the frequency sensitivity of atomic clocks.

3.7 Improvement of atomic clock with squeezed vacuum

The work reported in this Section 3.7 extends the work described in Section 3.6 and logically addresses objective #6 and objective #8 on the same system. Namely, protocols making use of entangled states of a rubidium condensate studied in Section 3.6 are designed and implemented (objective #6), here with the aim to make improvements in detection of noise and clock stability. Finally, a full prototypical clock sequence showing below standard quantum limit operation is demonstrated (objective #8). The work reported in this Section 3.6 was undertaken by REG(LUH).

The created two-mode squeezed vacuum states described in section 3.6 are however not directly applicable for interferometric applications, because the squeezed observables are two-mode quadratures of two modes, in our case F=1 m_F =±1. A possibility to exploit the two-mode squeezed vacuum for metrology is a basis change to the symmetric and asymmetric superposition of these states. In this new basis, the two-mode squeezed vacuum is converted to two independent single-mode squeezed vacuum states.

Single-mode squeezed vacuum is a very useful non-classical state for interferometry beyond the standard quantum limit (SQL). In a standard atom interferometric measurement, a fixed ensemble of atoms is prepared in one of two relevant states, leaving the second state initially empty. A first beam splitter operation transfers the atoms to a superposition of the two states. Here, they acquire a phase difference, which is read out by a second beam splitter operation and a final measurement of the number of atoms in the two states. The noise in the output signal, giving rise to the SQL, can be described as the amplified quantum noise of the initially empty (vacuum) input state. A possibility to reduce the measurement uncertainty below the SQL is to replace the vacuum input by a squeezed vacuum input state. This method has been successfully implemented in optical interferometry, including the operation of large gravitational wave detectors with a sensitivity beyond the SQL. It has however not been demonstrated in the field of atom interferometry.

Within QESOCAS, we have demonstrated an implementation of a prototypical microwave clock which can exploit the metrological gain of the generated squeezed vacuum states [Kruse2016]. A standard microwave clock drives transitions between the two hyperfine states that do not experience a first-order Zeeman shift, in the case of Rubidium F=1, $m_F=0$ and F=2, $m_F=0$. In our implementation, the lower clock state was replaced by the symmetric superposition of F=1, $m_F=\pm 1$. The symmetric state has also not first-order Zeeman shift and can be populated with a squeezed-vacuum state, as described above. For the clock operation, a combination of radio-frequency and microwave pulses couples the symmetric state to the upper clock state F=2, $m_F=0$. We demonstrated the implementation of the described clock measurement with an initially empty symmetric state and recorded Ramsey fringes as a function of the applied microwave frequency.

In a second step, we created a single-mode squeezed vacuum in the symmetric input state prior the microwave clock operation. On mid-fringe position, where the number of atoms are equally distributed between the two output states, we recorded a number fluctuation below the shot noise level. The most relevant metrological observable, the phase estimation uncertainty, was obtained from the measurement of the number fluctuations and the slope (or contrast) of the fringes. The result was 2 dB below the SQL for a total number of 10,000 atoms. The data allowed for an estimation of the Fisher information of the atomic resource and for a reconstruction of the Wigner function of the single-mode squeezed vacuum state. In addition, the Allan variance was recorded to quantify the frequency dependence of the technical noise sources.

In summary, the operation of a prototypical microwave clock beyond the Standard Quantum Limit was demonstrated. The experimental results proved the applicability of squeezed vacuum for quantum-enhanced atom interferometry. Interestingly the clock signal of 10,000 atoms in one input state was improved by squeezed vacuum with a mean atom number of only 0.75 atoms. The results point the way towards the implementation of entangled many-particle states in state-of-the-art atomic clocks.



3.8 Atomic phase locking and stability improvement from non-destructive detection in a trapped rubidium clock

The work reported in this Section 3.8 extends the work described in Section 3.4 and logically addresses objective #6 and objective #7 on the same system. The non-destructive detection of trapped rubidium atoms is used to implement a novel clock operation protocol (objective #6) where multiple successive weak non-destructive detections are used to gain information without interrupting the probing of the clock transition. A clock sequence making use of this information shows improved clock stability beyond the limit imposed by the oscillator frequency noise (objective #7). This work was undertaken by REG(IOTA).

The measurement-and-correction scheme developed to monitor non-destructively the evolution of the population unbalance of an atomic ensemble during a Rabi oscillation could be straightforwardly modified for a clock configuration. In this case, the evolution of interest is the relative phase between the classical oscillator and the fundamental oscillatory signal associated to a quantum superposition state. In a microwave clock, which is the case we studied, the classical oscillation is provided by a stable quartz oscillator; in an optical clock the classical oscillation is given by a laser locked on an ultra-stable optical cavity. In both cases, the clock stability is ultimately limited by the quality of the classical oscillator used to interrogate the quantum oscillation; this is due to the very long coherence time demonstrated for the atomic sample commonly adopted to implement atomic clocks. The limitation is set by the measurement process, which determines not the differential phase between the two signals but its projection in terms of a population unbalance. Inverting the relation to obtain the phase from the projection is ambiguous whenever the angle is comparable or bigger than the inversion region, namely the interval $[-\pi/2:\pi/2]$. In such case, phase wraps determine a readout error which affects the clock performance. The standard approach to improve the stability of a clock consists then in improving the quality of the local oscillator, to allow a longer interrogation interval without phase wraps. Our approach consists in measuring the phase drift at a safe time without destroying the atomic coherence, which is then available to continue the interrogation in a time correlated manner. On the basis of the measured phase drift, a correction phase shift is applied to maintain the relative phase within the inversion region for the next interrogation. The measurement-and-correction cycle can be repeated several times; in a last cycle the differential phase is measured precisely, and the total phase evolution over the extended interrogation time is computed by summing the last measurement and all the applied phase shifts. In this way one obtains a much longer effective interrogation time, and hence a proportionally higher sensitivity. The protocol has been proposed and experimentally demonstrated in [Kohlhaas2015].

We first demonstrated that our coherence preserving probe can be used to follow the evolution of the relative phase between the local oscillator and the atomic quantum state. To this aim we applied a frequency shift of 100 Hz to the LO after the beam splitter that prepares the quantum superposition state; successive weak measurements of the relative phase projection revealed a periodic oscillatory behavior with a period of 10 ms, resulting from the induced precession of the relative phase. The system has been then used in closed loop, first to correct disturbances applied to the LO, like periodic phase jumps or frequency drift. The correction was applied via a digitally controlled phase shifter acting on the LO phase. In a final set of experiments, the measurement-and-correction protocol was implemented in an atomic clock sequence. Instead of interrogating the atoms in a standard Ramsey fashion ($\pi/2$ – free precession – $\pi/2$ – destructive measurement of phase – application of frequency correction to the LO), we adopted a sequence with several successive Ramsey intervals, with coherence preserving measurements and application of phase corrections in between. In this way we could demonstrate a longer effective interrogation interval, and hence an increased sensitivity of the clock. More specifically, we demonstrated an enhancement of the sensitivity with the number N of successive probe intervals higher than $N^{1/2}$. This result demonstrates that successive measurements are at least partially correlated, which is due to the fact that the same coherent state is used during the sequence, without re-initialization of its phase.

We finally proposed a scheme to implement our protocol in the state-of-the-art lattice clocks, which are limited exactly by the quality of the LO. The protocol uses two atomic clocks operated at the same time, of which one allows the tracking and correction of the LO phase evolution via periodic coherence preserving measurements, and the second is devoted to limit the incidence of systematic effects. Recently a similar configuration based on two atomic systems was adopted at NIST to demonstrate an enhanced sensitivity in a spectroscopic measurement (Nature Photonics **11**, 48–52 (2017)).



3.9 Improved clock stability thanks to an ultra-stable lasers based on a long cavity

Lasers with excellent frequency stability are key to optical clocks. Better ultra-stable lasers yield better clock stability and ultimately better clocks. Equally in line with the goals of this JRP, lowering limitations of spectroscopy due to laser noise makes it easier to observe other limits, in particular the standard quantum limit, and to study the means to overcome them, here with quantum engineering. This Section 3.9 reports on the development of technology (objective #4) for improved ultra-stable lasers. The approach is here based on a room temperature long cavity. The resulting ultra-stable laser is characterized with a strontium lattice clock setup and shown to give excellent clock stability. The work was undertaken by PTB.

The application of advanced interrogation methods based on entangled or squeezed states offers considerably improved stability of quantum sensors like cocks. However, they benefit only if the quantum noise, in particular the quantum projection noise (QPN), is the dominant noise contribution. The detailed investigation of all noise processes contributing to the clock noise is therefore an important step to bring today's optical clocks to a performance level where they can actually be improved by squeezing or entanglement.

Hereby, the frequency noise of the interrogation laser plays an important role. The non-continuous interrogation of the atoms in optical clocks causes the Dick effect, which dominated at the start of the project the noise in lattice clocks even for very small numbers of atoms that were interrogated. Therefore, we implemented an improved interrogation laser with substantially reduced frequency noise in a Sr lattice clock.

The interrogation laser is pre-stabilized to a 48 cm long ULE glass resonator [Häfner2015]. The laser system shows a before not achieved fractional frequency noise floor of below 10^{-16} (see Fig. 3.10.1 in Sec. 3.10). With this laser system it was possible to increase the operational interrogation time to about 0.6 s increasing the line quality factor to about 3×10^{15} .

Using this laser, the instability of the clock was significantly reduced. The investigations were performed by a self-comparison of two independent, interleaved stabilization cycles on the same apparatus since no second clock with sufficient stability was available as reference [Al-Masoudi2015]. The observed instability is shown in Fig. 3.9.1.



Figure 3.9.1: Observed instability of the difference between two interleaved stabilizations with 0.64 s interrogation pulses (circles). The lines show the instabilities according to the noise model: full line $4.7 \times 10^{-16}/r^{1/2}$ for the interleaved; instability of $1.6 \times 10^{-16}/r^{1/2}$ (dashed line) inferred for single-cycle clock operation.

We have investigated different noise contributions to our Sr lattice clock, namely the QPN, detection noise and Dick effect-induced noise [Al-Masoudi2015]. The detection quality was found to be good enough to allow for QPN-limited detection at a few hundredths of atoms. A full noise model including a calculation of the Dick effect based on the noise characteristics of the interrogation laser shows very good agreement with the observed data (Fig. 3.9.1). With this model, we can infer a clock instability in normal operation mode of $1.6 \times 10^{-16} / \tau^{1/2}$, which was among the best found for any clock at the time. The noise model reveals however, that the Dick effect still dominates the clock instability even at small atom numbers and with the excellent



interrogation laser. Thus, we conclude that noise caused by the Dick effect has to be reduced before lattice clocks under realistic operation conditions will benefit from sub-QPN interrogation methods. This could either be achieved by quasi dead time free operation or improved interrogation lasers (see Sec.3.10 and 3.12). With a noise floor at few 10⁻¹⁷ and interrogation times beyond one second, this may become realistic.

3.10 Second generation cryogenic silicon resonator

As in Section 3.9, the work reported in this Section 3.10 addresses technology development (objective #4) relating to lasers with extreme frequency stability. Here, a more advanced solution is investigated, based on a single-crystalline cavity operated at cryogenic temperatures. This approach has the potential for much improved stability. A second generation system is developed and characterised against a first generation system and the long cavity of Section 3.9 Frequency noise properties and laser linewidth are investigated. The stability of the second generation system is inferred from the comparison of these three different lasers. It gives stability beyond that of any other laser. The work was undertaken at PTB.

Because of the Dick effect, laser frequency noise usually limits the instability of neutral-atom clocks above the value given by the quantum noise. Thus the improvement of the laser sources is an important step towards exploiting stabilities at the quantum level and subsequently even the below the standard quantum level by entangled states.

After years of improvement, technical noise levels of ultra-stable cavity-stabilized lasers have now been reduced to a level where the fundamental thermal Brownian fluctuations of the cavity constituents, most noticeable the mirror coatings, limit the frequency stability. One way to reduce this type of noise is operation a cryogenic temperature. In the setup at PTB we employ single-crystalline silicon for the cavity spacer and the mirror substrates. Silicon is a very promising material, as it has a low mechanical loss and thus low thermal noise and it also shows a zero thermal expansion coefficient at a temperature of 124 K.

At the beginning of QESOCAS we had already set up a 1.5 µm laser system based on a 21 cm long silicon cavity operated a 124 K that reached an instability of 1×10^{-16} , which was still away from the thermal noise level. During the course of QESOCAS we have rebuilt the cavity setup (Si2), improving on the vibration and thermal isolation, the cavity finesse and the optics [Matei2016]. To enable an efficient characterization and optimization in addition a nearly identical second independent cryogenic system was set up (Si3). Here the technical noise sources could be reduced to well below the thermal noise level. In a three-cornered hat comparison of the two Si based systems and the 698 nm laser system of the strontium lattice clock that is based on a 48-cm-long room temperature cavity (Section 3.9) we could infer a new record low instabilities expressed as modified Allan deviation mod $\sigma_y = 4 \times 10^{-17}$ (see Fig. 3.10.1 left) for both silicon systems.



Fig 3.10.1: Left: Individual Modified Allan deviations from a three-cornered hat comparison of Si2 (squares), Si3 (circles) and the 698 nm ULE laser (diamonds). Right: FFT spectrum of the beat note between lasers Si2 and Si3 (frequency resolution 5 mHz).



The beat frequency between the two silicon systems was mixed down to a few kHz to analyze the spectrum with a Fast Fourier Transform (FFT) analyzer. We have obtained linewidth of the beat of below 10 mHz, indicating individual linewidths of the lasers as low as 5 mHz [Matei2017]. From the recorded phase fluctuations, we were able to predict practical coherence times for different atomic interrogation schemes in optical clocks.

The 698 nm strontium clock laser is compared to the silicon based laser via a fs-frequency comb. It provides a real-time "virtual beat" at a frequency $m_{Si}/m_{Sr} \times v_{Sr} - v_{Si}$ (m_{Si} and m_{Sr} are the integer mode numbers of the comb at both frequencies v_{Sr} and v_{Si}) that will allow to phase-lock the 698 nm clock laser to the Si cavities and make their high stability available for improved operation of the strontium lattice clock.

3.11 Tailored probing sequence for lattice-clock assisted ion interrogation

To date, optical clocks based on ions are single ion clocks where detection is at the quantum limit. For this single ion clock, increasing the interrogation time is the only way to improve stability. One way to achieve this is to improve ultra-stable lasers, which is the approach followed in Section 3.9 and 3.10 in line with objective #4. Here, another approach is studied along the line of objective #6. The idea is to use an optical lattice clock to gain information in real-time about the laser frequency noise in order to enable extend interrogation time in the ion clock, beyond the limit imposed by the laser noise. One can describe this scheme as lattice-clock assisted ion clock. With this idea in mind, a specific probing sequence for the lattice clock was devised, and optimized. This work was undertaken at PTB.

The stability of optical atomic clocks is mainly limited by the ultra-stable clock laser used for interrogation: In optical lattice clocks with the large available signal-to-noise ratio (SNR) mostly the Dick-effect, i.e. the aliasing of frequency fluctuations limits the instability. In single ion clocks, the quantum projection noise limits the SNR and the stability, however, if the interrogation time can be increased, still the quantum-limited stability would improve as the square root of the interrogation time. With corresponding natural linewidth in the clock ions of below 10 mHz (for Al⁺, Yb⁺ octupole) interrogation times of more than 10 second could be employed, which is far beyond the coherence time of today's best lasers.

To overcome this limitation, we developed a novel interrogation scheme that will allow building a compound clock to enable probe times longer than the coherence time of the clock laser. The proposed technique utilizes a correlated interrogation sequence of two atomic clocks with one lattice clock laser to resolve the phase ambiguity apparent in Ramsey interrogation of the second clock beyond the coherence limit of the interrogation laser. This leads to an improved stability of the frequency delivered by the compound clock. In addition, the frequency ratio of the optical clocks used in this compound approach is arbitrary.

By using a sequence of multiple Ramsey-like pulses, we can design a sensitivity function that is approximately constant, but less than one (Fig. 3.11.1). Thus the atomic signal at the end of the sequence depends unambiguously on the average frequency during the full interrogation time. This knowledge of the true average frequency can then be used to resolve the 2π phase ambiguity that appears in a Ramsey interrogation on the second clock at in interrogation time beyond the laser's coherence time. For this scheme to work it is not even necessary that both clocks operate at the same optical frequency. It is only necessary to perform a phase-coherent lock between the two interrogation lasers e.g. via a femtosecond frequency comb.





Fig. 3.11.1: Left: Sensitivity function for a multiple-pulse Ramsey sequence (red) in comparison to the one of a Rabi interrogation. Right: Trace of the Bloch vector on the Bloch sphere with a detuning $\Delta = 0.4/T_{dark}$.

As we had only one clock available, we have tested the scheme using PTB's strontium optical lattice clock in a sequential fashion. We could show, that the phase obtained with a standard Ramsey sequence could be unwrapped using the signal from a sequential multi-Ramsey interrogation of duration T_{Rams} , when the laser was detuned by more than one Ramsey fringe period $\Delta > 1/T_{Rams}$. In a real compound clock, both interrogation would be performed in parallel with independent clock.

3.12 Development of crystalline coatings for ultra-stable lasers

The work reported in this Section 3.12 deals with the technology development (objective #4) of better ultrastable lasers. It is established that one of the limits to the stability of such lasers comes from mechanical thermal noise in the coatings of mirrors used in ultra-stable cavities to which lasers are stabilized. Using crystalline materials from coatings can reduce this mechanical thermal noise drastically. Such technology can have a large impact on ultra-stable lasers and clocks with an extreme level of performance. The work reported here is aimed at developing several aspects of this key technology to adapt to a diversity of situations such different wavelengths, different types of substrate and also cryogenic temperature. The yield and control of the manufacturing process was another key issue considered in this work. The work was undertaken by REG(XMS) and PTB.

In the course of this effort REG(XMS) has made significant progress in the development and optimization of substrate-transferred crystalline coatings for ultra-stable optical reference cavities. In the end we met and even exceeded the technical targets as outlined in our original proposal. The only negative point in this endeavor is the inability to make a direct thermal noise measurement within the timeframe of the project, though this work has continued beyond the closing date of June 2016.

Highlights of our research efforts include:

1) The development of a robust and repeatable manufacturing process for AlGaAs-on-Si end mirrors for cryogenic optical reference cavities

2) Demonstration of <5 ppm of excess optical losses for 1550 nm crystalline coatings on Si (with a maximum finesse exceeding 300,000 in our best optics)

3) Completion of all XMS(REG)-related deliverables, with the single exception being the thermal noise measurements to be performed by PTB



4) Continued growth of our start-up company (from 2 people at the start of the effort to 15 staff today), as well as immediate commercialization of all technologies developed during the course of this project

At the outset of this project on February 1, 2014, REG(XMS) consisted of two staff members, namely Prof. Markus Aspelmeyer and Dr. Garrett Cole of the University of Vienna. Our crystalline coating technology had first been demonstrated in 2013 (Nat. Phot. 7, 644 (2013)), and while we had proven the low-Brownian noise properties of our mirrors at room temperature, the optical quality of mirrors produced with our substratetransfer coating technique still had room for improvement. Early optics showed losses at the ~15-20 ppm level with a strong position dependence on the optical quality. With the support of the funding supplied by the QESOCAS project we spent 18 months optimizing our manufacturing process and improving our inspection techniques in order to minimize the optical losses in our single-crystal interference coatings. Over that period our company expanded greatly, adding an additional office in Santa Barbara, CA, USA and developing customized tooling dedicated to the manufacturing of high quality crystalline coatings. The most significant advancement developed during this project was the realization of our "flipped" coating process, whereby we take the epitaxial multilayer, remove it from the original growth substrate and bond it to the final substrate face down, with the top of the mirror surface now being the backside of the originally deposited multilayer. Utilizing this improved manufacturing process, we were able to significantly reduce the limiting optical scatter in our coatings. In parallel, we pursued improved epitaxial growth processes in order to reduce the background impurity levels in the mirror structure. Modifications included an increased deposition rate (to minimize the incorporation of dopants from the growth chamber) as well as a slight increase in the growth temperature to 650-700 °C (vs 600 °C for our original process). Ultimately, we were able to reduce the total excess losses to less than 5 ppm, with absorption at the sub-ppm level and optical scatter losses as low as 3 ppm in the best optics, enabling a finesse exceeding 300,000 at 1550 nm.

With the optical quality reaching a level on par with state-of-the-art ion-beam sputtered coatings, we then turned our attention to developing a repeatable process for transferring our epitaxial GaAs/AlGaAs multilayers onto single-crystal silicon substrates. Our initial attempts, made shortly before the kick-off of this project, had been successful at room temperature, though some optics exhibited delamination upon cooling to cryogenic temperatures. Thus, in order to avoid failure of any AlGaAs-on-Si optics in cooling, we undertook a dedicated study to significantly enhance the bond strength of our coatings. Working in this direction we developed customized tooling to reduce handling of the delicate components during cleaning and bonding (in order to minimize defects that tend to lead to interfacial failure) and we also explored various post-bond annealing processes to further enhance the interfacial energy between the GaAs and underlying Si. Ultimately, we were able to realize bond strengths sufficient for cryogenic cycling to liquid ⁴He temperatures below 10 K. This was more than enough for successful cooling to the target temperature of this effort, which was the first zero-crossing in the thermal expansion of single-crystal silicon at 124 K.

With the optical quality and bond strength improved, and with a robust transfer process optimized for singlecrystal Si substrates identified, we then produced the final physical deliverables for this effort, namely optimized crystalline GaAs/AlGaAs interference coatings on bulk Si substrates that were delivered to our partners at PTB. These optics have now been thermally cycled and shown to be robust against the thermal strain evolved in cooling to a base temperature as low as 100 K. Even more excitingly, these optics have demonstrated a maximum finesse of approximately 400,000 at 1542 nm, more than sufficient for suppressing residual amplitude modulation in the planned cryogenic cavity-stabilized laser system. Our growing team at CMS, now up to 15 staff members, is very happy to have been given the opportunity to participate in this exciting project and we are of course very pleased with the scientific advances realized. Since this research grant kicked-off in early 2014, we have expanded our product lines to include low-loss mid-infrared optics, which directly exploit the Si coating process explored in this project. Furthermore, the enhanced bond strength required to survive cryogenic cooling has also enabled us to produce a new product line of high thermal conductivity and low optical loss mirrors for high-power laser systems. Here, the high bond strength we can realize allows the optics to survive the extreme operating environment that they are subject to.



3.13 Characterizing spatial variations of frequency shifts by correlating Ramsey experiments in long ion strings

As mentioned already in Section 3.1, using entanglement to improve ion clocks requires traps with improved characteristics that can support multi-ion operation. One key characteristic of such traps is the spatial homogeneity of frequency shifts across the string of ions. In the work described in this Section 3.13, a method for efficient measurement of trap inhomogeneity is demonstrated for up to 20 ions. This can be used as a generic characterisation and optimisation method to support and qualify novel ion traps for quantum metrology (objective #4). The work is also contributing to objective #2 as it relies upon and tests parallel coherent laser excitation and detection of a string of ions. This work was undertaken by REG(OAdW).

Optical atomic clocks probing multiple ions crystallized into an ion string require a precise spatial characterization of sources of frequency shifts over spatial distances of a few tens of micrometers. Previous experiments demonstrated that the dominant sources of noise limiting the coherence time are typically strongly correlated on the different ions. This opens the prospect of probing spurious frequencies shifts with probe times vastly exceeding the single-particle coherence time by measurements of two-particle coherences.

For the case of two ions, the most direct approach yielding the optimum signal consists in preparing the particles in an entangled state $|\phi(0)\rangle = (|e\rangle |g\rangle - |g\rangle|e\rangle)/\sqrt{2}$, letting the state evolve for a certain time τ . If the transition frequencies differ on the two ions, the state $|\phi(\tau)\rangle$ will oscillate between singlet and triplet states at frequency equal to the ions' frequency difference. This oscillation is revealed by a measurement of a two-ion coherence, i.e. a measurement of the parity along a direction in the equatorial plane of the Bloch sphere.

For the case of N>2 ions, this approach is no longer optimal, as any ion that is maximally entangled with another ion, cannot be entangled with any other ions of the string. Therefore, a superior approach consists in preparing an initial state in which each ion is prepared in an equal superposition (|g> + |e>)/ $\sqrt{2}$ and following the prescription given above for all pairs of ions in parallel. Correlated noise acting on the string will decohere any two-ion reduced state into a mixed state that has a fidelity of 50% with a maximally entangled Bell state and which contains genuine quantum correlations that can be characterized by quantum discord. In the absence of strong enough noise sources, the state can be actively dephased by randomizing the phase of the measurement $\pi/2$ pulses with respect to the preparation pulses. Compared to the case of a maximally entangled state, the resulting contrast is reduced by 50%, but each ion can be correlated with every other ion in the string instead of a single one. Moreover, the protocol is attractive from the point of view that it does not require the capability of deterministically entangling ions by laser pulses using protocols requiring ground-state cooling the ion crystal.

We have implemented this protocol with up to 20 calcium ions forming a linear ion crystal of about 100 μ m length in order to investigate the shifts of the S_{1/2} to D_{5/2} quadrupole transition caused by the electric quadrupole shift, linear magnetic field gradients and ac-Zeeman shifts induced by the rf-fields confining the ions.



Figure 3.13.1: Measurement of magnetic field inhomogeneity by correlation measurements on a twenty-ion string



The left panel of the figure shows the parity oscillations obtained by correlating the state of ion 1 with all of the other ions in a 20-ion string for an investigation of the magnetic field inhomogeneity causing a spatially varying Zeeman shift. Measurements on different Zeeman transitions and variation of external control fields enable disentangling the various sources giving rise to the measured shifts. The correlation technique has enabled measuring shifts with sub-Hertz resolution using probe times of up to 300 ms in an experiment where the single-ion coherence on some of the transitions was shorter by an order of magnitude. This makes the technique a powerful tool for precision characterization of spatial frequency shift variations and demonstrates the power of harnessing quantum correlations for precision measurements.

3.14 N-ion multipartite entanglement generation: Mapping phononic excitations onto electronic excitations

Objective #2 and objective #5 of the JRP are respectively to devise and implement entangled states in trapped ion systems and to characterize these states in terms of degree of correlation and coherence lifetime. This Section 3.14 report such study. The novel scheme investigated to generate entangled states is based on first generating an entangle state of motion in the string of ions, making use of strong coupling provided by the coulomb interaction between the electric charge carried by each ion. Then, a design laser excitation maps all at once and smoothly the entanglement of motion into entanglement of the internal state of ions, the metrologically useful entanglement. The lifetime of this state is measured, highlighting the importance of traps with low heating rate to reach long lifetimes. This work was undertaken by REG(OAdW).

Entangled states of atoms are of interest for quantum metrology as some of these states have been shown to provide an enhanced sensitivity for parameter estimation as compared to the case of unentangled atoms. One such class are Dicke states, i.e. states in which a fixed number n of excitations is coherently delocalized over the ensemble of N atoms, as was recently demonstrated for various experimental settings (R. Krischek et al., PRL 107, 080504 (2011), I. Apellaniz, NJP 17, 083027 (2015)).

Given this background, we set out to create N-ion multipartite entanglement by creating symmetric Dicke states of N ions, i.e. states that are symmetric under any permutation of the ion numbering with N/2 excitations coherently delocalized over the ion string. Our approach to creating these states was to first prepare the string's collective center-of-mass (COM) motion in a Fock state with N/2 phononic excitations, and to use a laser pulse to convert these phononic excitations into delocalized electronic excitations by rapid adiabatic passages on the first vibrational sideband of the electronic transition. While the generation of motional Fock states is a standard technique in trapped-ion experiments, the creation of such Dicke states by rapid adiabatic passages has never been demonstrated before.

In our experiment, we cooled an 8-ion string to the ground state of the COM motion and used a focused laser beam to add n phonons to the mode while preparing all calcium ions in the S1/2 state. We then swept the frequency over the red vibrational sideband using a Gaussian intensity envelope in order to realize the adiabatic passage mapping phonons to electronic excitations in the D5/2 state as illustrated in panels (a) and (b) of Figure 3.14.1. By global spin rotations prior to a fluorescence measurements detecting the S1/2, we could measure the populations of any desired spin projection operator as shown in (c). As expected, most of the population resides indeed in the state with four excitations and the odd-state populations of the x- and y-operator are suppressed. Further measurements allowed a characterization of the state by reconstructing its Wigner function shown in (d) (following the prescription laid out in NJP 13, 065019 (2011)).

Based on our experimental data, we could confirm that the created state has a generalized spin squeezing parameter $\xi_{gen}^2 = 0.37 < 1$ (for a definition see PRL 112, 155304 (2014)) and thus is entangled. The state is also detected as being non-separable by other entanglement witnesses.





Figure 3.14.1: Creation and analysis of 8-ion symmetric Dicke states. (a) States coupled by the rapid adiabatic passage (RAP). (b) Mapping of phononic to electronic excitations by the RAP. (c) Measured populations of an 8-ion Dicke state in the x-, y- and z-basis. (d) Reconstructed Wigner function.

The experimental results demonstrate that rapid adiabatic sideband passages for mapping Fock to Dicke states is a promising technique for creating metrologically useful multi-partite entangled states. The current results also highlight the need for carrying out such experiments in ion traps with low vibrational heating rates in order to avoid a diffusion of phonon populations during the mapping step by randomly fluctuating electric fields at the motional frequency. Our current experiments have been affected by this noise source, as is evident from the z-basis population distribution shown in (c) which have prevented us so far from demonstrating the superiority of entangled states over separable states. We expect, however, to overcome this problem by repeating the experiments in an improved trap similar to the one employed in another Innsbruck experiment, which features heating rates that are more than an order of magnitude lower than in the present one.

3.15 Key results and conclusions

The key results and conclusions of the research undertaken are as follows:

- We have improved technologies and methods that will enable optical clocks with multiple ions using quantum entangled states. In particular, we have developed high yield parallel fabrication of micro-fabricated ion traps on 4 inch wafers. We have developed methods to test and characterize such traps, and assess their suitability for optical clocks.
- We have experimentally demonstrated non-destructive detections of trapped atoms for clocks, based on the dispersive effect of the atomic sample on a probe light.
 - We have developed a novel detection scheme to measure the transition probability in optical lattice clocks with unprecedented sensitivity. We have investigated a novel heterodyne optical system to homogeneously probe atoms in a linear cavity. We have shown that this system is in principle able to produce quantum engineered states of atoms in optical lattice clocks.
 - We have demonstrated the non-destructive detection of the hyperfine population difference of trapped rubidium atoms. We have studied factors that influence the trade-off between destructivity and sensitivity of the detection. We have optimized this detection scheme for its applications to clocks. We have developed control systems capable of fast acquisition to the information delivered by this detection and of fast actions on the atomic system based upon this information.
- We have theoretically investigated whether squeezed light could improve such non-destructive detection schemes. Taking into account realistic conditions of implementation, especially, optical losses, we estimated that significant enhancement of the sensitivity of such detection is possible.
- Using the non-destructive detection of hyperfine population difference of trapped atoms, we have experimentally demonstrated a novel scheme where the phase difference between the atomic coherence and the interrogation oscillator is tracked without destroying the atomic sample. We have



shown that this scheme could be used to extend the interrogation of the atomic sample beyond the coherence time of the interrogation oscillator. We have experimentally shown that, in turn, thanks to the extended interrogation duration, the clock stability was improved.

- We have proposed and theoretically studied a scheme where an optical lattice clock is used to extend the interrogation time of an optical single ion clock beyond the coherence time of the interrogation laser. As in the previous result, such scheme provide a mean to improve, for a given ultra-stable laser, the stability of the ion clock.
- We have developed novel ultra-stable lasers. Room temperature laser based on a long 48 cm cavity at 698 nm was designed, implemented and characterized against two cryogenic silicon cavities. State-of-the-art thermal fractional frequency noise floors of 6x10⁻¹⁷ and 4x10⁻¹⁷, where demonstrated. With the room temperature laser, state-of-the-art fractional frequency stability of 1.6x10⁻¹⁶ at one second was demonstrated on a strontium optical lattice clock.
- We have pushed crystalline coating technology that is key to further improve such ultra-stable lasers. Repeatable, high yield manufacturing of AlGaAs crystalline mirror coating with low optical losses was developed (best finesse above 300000). Process to transfer of such coatings onto crystalline silicon substrate was demonstrated, that can withstand operation at cryogenic temperature. This is a key enabling technology for next generation ultra-stable lasers.
- In a rubidium Bose Einstein condensate experiment, we have shown that interactions and spin dynamics offer the reliable production of squeezed vacuum states. We have characterized this squeezed vacuum. We have designed a scheme of a prototypical microwave clock were such states was used to demonstrate stability beyond the standard quantum limit, thereby experimentally proving the applicability of squeezed vacuum for quantum-enhanced atom interferometry.
- In multiple ion trap, with strings of up to 20 ions, we have demonstrated a protocol using 2 ion correlation Ramsey spectroscopy to perform precise characterization of systematic shift inhomogeneity across the ion string. This is an important capability for enabling highly accurate multiple ion clocks. A second key result is the demonstration of a novel promising scheme for generating entangled state of multiple ions, by rapid adiabatic transfer from motional Fock state to metrologically useful Dicke state. Limits of the scheme were analyzed and ways forward to higher level of entanglement defined, highlighting the need for trap with lower heating rates.

In conclusion, we have demonstrated several new key enabling quantum engineering methods for improving optical clock stability and also accuracy in a variety of systems. In several case, these methods can give a benefit both in a quantum regime, to go beyond the standard quantum limit, and in a classical regime, for instance, to make a better use to existing ultra-stable oscillators. We have also improved key technologies for future ultra-stable oscillators. We have developed state-of-the-art ultra-stable lasers and use them to demonstrate clock stability close to 10⁻¹⁶ at one second. Several of these achievements relied upon joining strengths and capabilities between partners. Exchange of ideas between partners was also key to several novel concepts demonstrated within the project.

The research met its targets and achieved almost all of the objectives. Prototypical experiments were made for all objectives with one of the systems under study or more. Development of high yield procurement and manufacturing of micro-fabricated ion traps suffered significant delays but made significant progress. Early tests bodes well for their use in clocks and quantum metrology and work in this direction continued after the formal end of the JRP.

4 Actual and potential impact

4.1 Metrological capabilities

The QESOCAS project led to new metrological capabilities. New ultra-stable lasers with state-of-theart stabilities were developed which in turn improved the stability of optical clocks at PTB. At LNE-SYRTE, a novel non-destructive detection method was implemented on a fully operational optical lattice clock. In the near future, this also will enable improvement of clock stability. Similarly, for the high yield manufacturing of micro-fabricated ion traps and the high fidelity interaction developed at NPL will facilitate the development of multi-ion clocks as operational frequency standards. These



improved capabilities are and will be used in coming optical fiber link inter laboratory comparisons which keep European laboratories at the forefront of the work toward a redefinition of the second based on optical frequency metrology.

4.2 Scientific impact

The project gave 23 publications in high-ranking peer-reviewed scientific journals. More are submitted or in reparation. 33 oral presentations were given at international conferences and workshops, including several invited talks. 19 posters related to the project were presented at international conferences and workshops. All this yielding broad and public dissemination of the outcome of the project.

The project demonstrated several novel concepts and methods which are and will further be used to enhance the sensitivity of quantum sensors such as optical clocks and atom interferometers. Several of these concepts are in principle applicable to other atomic sensors such a magnetometers. Some of these concepts and methods may also contribute to the broader field of quantum technologies. This is for instance the case of low heating rate, wafer-based micro-fabricated ion traps, of entangled atomic system preparation scheme.

Methods and concepts developed within the QESOCAS project have and will further improve optical clocks, starting with clocks developed and operated at NMIs for frequency and time references. For example, PTB have state-of-the-art clock stability thanks to QESOCAS, OBSPARIS has a clock with unmatched ultra-sensitive detection thanks to QESOCAS. This will in turn impact scientific applications of these clocks. It will enhanced their capability to test and investigate fundamental laws of physics, to uncover physics beyond the standard model and to search for dark matter.

As sensors of gravity, better atom interferometers and better optical clocks will impact geodesy and Earth science. Concepts and methods developed with the QESOCAS project will help reaching the ultimate limits of these quantum sensors in term of sensitivity to gravity. They will also offer new possibilities for revisiting the trade-off between sensitivity and transportability. For instance, with a non-destructive detection, one may obtain the same clock stability with a simplified, more transportable and more affordable ultra-stable laser.

4.3 Continuation of this research

Research undertaken within this project will continue being highly relevant and timely. In particular, it is connected to the new Future and Emerging Technology Flagship on Quantum Technologies which is being started by the European Commission. Outcomes of the QESOCAS project will feed new activities under the quantum sensing & quantum metrology pillar of this program. Clocks and atomic sensors are explicitly identified as important areas of applications for Quantum Technologies, where readiness for transfer toward industry is high. Further, advanced optical time and frequency metrology is a potential resource for other pillars of the Flagship program such as quantum telecommunication or computation.

4.4 Dissemination activities

In addition to the already mentioned general and usual dissemination via scientific publications and communications, a final international workshop was organized, jointly with the EMRP JRP ITOC project, in order to disseminate the outcome of QESOCAS. The workshop took place in association with the European Time and Frequency Forum (April 2016, University of York, UK) which is one of the most important international conferences in time and frequency metrology. As a consequence, the workshop could attract many participants from companies, from stakeholders and organizations acting in field of time and frequency metrology, and from laboratories and metrology institutes outside Europe, like for instance, NIST (USA), NRC (Canada), NICT & NMIJ (Japan). Further information on the workshop can be found at this link: http://www.eftf2016.org/satellite-workshop.html.

4.5 Capacity building and training

As part of the project, 14 training courses were provided, often in international or European schools or events. Many of these schools or events were specifically oriented toward students, PhD students and early stage researchers. In particular, the QESOCAS project co-organised a school together with the



Initial Training Network FACT (Future Atomic Clock Technology). Several partners of QESOCAS gave lectures at this school. Similarly, QESOCAS partners gave lectures at another school organised by another European Initial Training Network QTEA (Quantum Sensor Technologies and Applications). Also broadly reaching students, PhD students and early stage researchers, QESOCAS partners gave 29 others presentations at seminars/workshops in many countries, both in Europe and outside, several of them as invited speakers.

Seven PhD thesis were completed whilst working on the QESOCAS project. Four other PhD students contributed to the project as part of their PhD.

At QESOCAS project meetings, these PhD students, and also other early stage researchers, were involved as participants or as contributors, giving them opportunity to learn about European organizations in their field, to visit laboratories and more generally to get familiar with European projects.

4.6 Contribution to committees and regulations

Many partners of QESOCAS are members of scientific committees associated with the main international conferences in time and frequency metrology, such as the European Frequency and Time Forum, the International Frequency Control Symposium and of the Conference on Precision Electromagnetic Measurements. Results of QESOCAS contributed to defining some orientations and programs of these conferences, for example in proposing topics and invited speakers that enhance connections and promote exchange of ideas between time and frequency metrology and quantum engineering communities.

Several partners of QESOCAS participated, making use of the results of QESOCAS, in a meeting organised by the Joint Research Center (JRC) of the European Commission. They contributed to the elaboration of *JRC Science for Policy Report: The impact of quantum technologies on the EU's future policies*. Likewise, QESOCAS partners, making use of the results of QESOCAS, participated in meetings and contributed both at the national and European level to the elaboration of roadmaps for the Future and Emerging Technology Flagship on Quantum Technologies which is being started by the European Commission.

Several partners of QESOCAS are members of several working groups and consultative committees of the Conférence Internationale des Poids et Mesures (CIPM). Within these committees, they disseminate the result of QESOCAS when appropriate and make use of results of the project to contribute defining recommendations made to the CIPM. The committees where QESOCAS partners are members are the Consultative Committee for Time and Frequency (CCTF) of the CIPM and the following working groups (WG): WG on Primary and Secondary Frequency Standards, Joint Consultative Committee on Length & CCTF WG on Frequency Standards, WG on Advanced Time and Frequency Transfer, WG on International Atomic Time.

4.7 Socio-economic impact

Shortly before the beginning of the project, the start-up company Crystalline Mirror Solutions GmbH was founded. It became a partner of the project as REG(XMS). For the need of the project, the company improved its processes for manufacturing high quality crystalline mirror coatings. The characteristics of this technology was thereby enhanced in terms of optical losses and in terms of bonding strength to several substrates. With these improvements the number of applications of the technology was augmented and the company expanded its product lines. In particular, it enabled applications of these coatings to the new spectral range (mid-infrared). It enabled applications in a cryogenic environment. It enabled other applications in extreme conditions such as high power laser systems. At the start of the project, the company was only composed of the 2 founders of the company. Today, the company has grown to 15 staff members. The research undertaken within the QESOCAS project had direct and immediate impact on innovation and creation of new economic activity.

On the long term, the work undertaken within the QESOCAS project will translate into increased levels of performance of operational optical clocks and atomic sensors. Such future optical clocks will be



used to the redefine the second of the international system of units and to improve timescale and timekeeping. Such timescale, like International Atomic Time (TAI) or time reference of Global Navigation Satellite Systems, are essential globally available resource and infrastructure to society. More accurate optical clocks will also yield a new application, named chronometric geodesy, were clocks are used to a sensor of the gravitational potential and as a tool of better knowledge of earth's gravitation for the benefit of earth science, of exploration and management of underground resources, better understanding of the earth climate, etc. Likewise, improved atomic sensors will improve earth science and its applications.

5 Website address and contact details

The address of the project public website is <u>https://syrte.obspm.fr/jrpexI01/</u>.

Enquiries about the project should be addressed in the first instance to the project coordinator, Sébastien Bize for LNE-SYRTE (<u>sebastien.bize@obspm.fr</u>).



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