

Atoms and Ultra-Precise Measurement of Time

Christophe Salomon

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Introduction

In this Note, rather than giving an overview of the field of clocks and ultra-precise measurement of time, I relate the scientific context and results which have led to the attribution of this prestigious Balzan Prize to the author of this paper. First of all, it is a tremendous honor and a great joy for me, and I would like to express my warmest thanks to the General Prize committee and to the Balzan Foundation, whose commitment to Culture, Arts, and Science is truly remarkable.

This prize rewards a field of research that is very active internationally and has made spectacular progress in recent decades. Indeed, the precision of time measurement has improved by a factor of one billion over the last 70 years, reaching an error smaller than one second over the age of the universe, 13.7 billion years! This achievement is not the result of the work of a single person, but of the relentless, persevering work of an entire international scientific community in which the spirit of collaboration has always prevailed over the spirit of competition.

This is why I would like to sincerely thank all my colleagues, students, and postdoctoral researchers with whom I have had the privilege of working during my scientific career. In particular, I express my deep thanks to Claude Cohen-Tannoudji, Alain Aspect, Jean Dalibard, the late André Clairon, Philippe Laurent, Pierre Lemonde, Sébastien Bize, Giorgio Santarelli, Peter Wolf and the whole team at the Laboratoire Temps Espace at Paris Observatory. Without them, the results that led to this award could not have been achieved.

When talking about time in Bern, the name of Albert Einstein, who completely revolutionized our understanding of time, immediately comes to mind. It was here that Einstein developed the theory of special relativity between 1903 and 1905. From then on, time is no longer a universal quantity that flows uniformly at all points, as Newton had assumed. The three dimensions of space and time form a single,

inseparable block that is distorted by the surrounding matter. Thus, time flows differently on Earth, on a satellite, or on distant stars!

Today, accurate time measurements have become essential in many fields, such as navigation, the synchronization of transport and telecommunications networks, banking transactions, or satellite positioning. As an example, our cell phone constantly receives signals from atomic clocks onboard GPS or GALILEO satellites orbiting the Earth at an altitude of 20.000 kms. These signals provide every individual with their position within a few meters' accuracy anywhere on the globe. This represents an incredible service to billions of individuals and to our modern society, a service which is, in addition, free of charge.

Academic Career Path

My activity on clocks, precision measurements, laser frequency stabilization, and laser cooled atoms started during my PhD under the supervision of Christian Bordé (1980-1984) and my post-doctoral stay in the group of Nobel laureate John Hall at JILA, University of Colorado, in 1984-1985 [Salomon 1988]. My PhD work on high resolution sub-Doppler laser spectroscopy and Ramsey fringes in the mid-infrared has pushed the resonance linewidth on room temperature molecules down to the kilohertz level, with a resolving power $\nu/\delta\nu = 3 \cdot 10^{10}$. This work has revealed novel hyperfine structures and symmetries in spherical top molecules such as SF₆ or OsO₄. The spectral resolution was limited by the transit time of room temperature molecules across the 11 cm diameter laser beam in an 18-meter-long saturated absorption cell with 1 m diameter. The demonstration of the slowing of an atomic beam by W. Phillips and H. Metcalf at the beginning of the 1980s at NIST (USA) [Phillips 1982] convinced me that controlling the motion of atoms or molecules was a clear way forward for improving laser spectroscopy and precision measurements. At that time, in the mid-80s, experimentally complex atomic beam machines and dye laser systems were used to manipulate the atoms. Nevertheless, at JILA, I was thrilled to slow down and cool a sodium atomic beam to very low velocities by radiation pressure.

In the fall of 1985, I was invited to join the newly created laser cooling group at the École Normale Supérieure (ENS) to establish an experimental activity together Jean Dalibard and Nobel laureates Claude Cohen-Tannoudji and Alain Aspect. Apart from visiting scientist positions in Germany and in the USA, I remained at

Laboratoire Kastler Brossel (École Normale Supérieure, ENS-Paris), first as a CNRS junior researcher, and later as CNRS research director.

Laser Cooling

In the mid-1980s diode laser systems operating in the near infrared were just beginning to appear on the market (even though with very limited laser power, in the 5-10 mW range and with poor spectral purity). By their compactness, and low cost, diode lasers were expected to bring substantial simplicity to the experimental set-ups aiming at cooling and trapping atoms. The ENS group and I then decided to work with cesium atoms, the atoms in use in atomic clocks since 1967; their optical resonance line is at a wavelength of 852 nm. The first goal of the group was to investigate the laser cooling of cesium. This goal was reached beyond expectations as the lower limit for laser cooling was not set by the commonly accepted Doppler limit but by a Sisyphus cooling mechanism theoretically identified by Jean Dalibard and Claude Cohen-Tannoudji [Dalibard 1988] and independently by S. Chu at Stanford [Chu 1988]. In this sub-Doppler cooling process, the atom is continuously forced to climb up potential hills, losing energy very efficiently, as in the Greek myth of Sisyphus. In [Salomon 1990] my collaborators and I carried out a detailed test of the Sisyphus cooling theory with cesium atoms in optical molasses and could achieve a temperature of only 2 microKelvin, two orders of magnitude below the Doppler cooling limit, and the lowest ever temperature achieved at the time. The cesium atoms were viscously confined in the so-called «optical molasses» created by three pairs of counterpropagating cooling laser beams in a very low residual pressure vacuum chamber.

Cesium Atomic Fountain Clock

With a thermal velocity of 1 cm/s, when turning off the molasses beams, the 2 μ K cloud of cesium atoms fell under the effect of gravity with a very small expansion, almost like a stone! Following an early proposal by J. Zacharias in the mid 1950's at MIT, my long-term collaborator and friend André Clairon from the French national metrology institute at Paris Observatory and I then created at ENS the first cesium atomic fountain [Clairon 1991].

From optical molasses, laser cooled atoms were launched upwards through a microwave cavity exciting the cesium hyperfine clock transition at 9.192 GHz. Atoms passing through the cavity on their way up and on their way down produce interference fringes; their period

scales as the inverse of the travel time that the atoms spend above the microwave cavity and is called «Ramsey fringes». Such resonances bear the name of its inventor, Norman Ramsey, who received the Nobel Prize in 1989 for introducing this powerful tool in quantum physics which is still universally used today. In our first fountain, the coherent interaction time of 250 milliseconds provided Ramsey fringes with a resonance width of 2 Hz, two orders of magnitude narrower than in conventional atomic beam cesium clocks. Furthermore, the inferred frequency stability from the signal to noise ratio was at least as good as in the best atomic beam primary cesium clocks developed in metrology institutes. This work clearly demonstrated the interest of cooling and controlling the motion of atoms for clocks and precision measurements. Our 1991 paper opened the way to the realization of operational primary cesium standards in fountain geometries.

Primary Frequency Standards and International Atomic Time

The first cesium fountain operating truly as a primary frequency standard with all systematics effects evaluated and corrected was achieved at Paris Observatory in 1995, followed later by several international metrology institutes worldwide including PTB, NPL, NIST, NICT, METAS, KRISS, and NIM.

In 1997, I initiated a very fruitful collaboration with a team from the University of Western Australia led by D. Blair. The team was a worldwide specialist of ultra-stable cryogenic oscillators. Using such a device as the interrogation oscillator for the Paris cesium fountain, we were able to show for the first time that a cold atom fountain could operate at the fundamental quantum noise limit up to several million atoms [Santarelli 1999, Bize 2005]. This noise is the fundamental noise induced by the measurement process in quantum mechanics. For N uncorrelated detected atoms, the signal to noise ratio S/N scales as $N^{1/2}$. This is the standard quantum limit. Following this work, advances in recent years with quantum correlated atoms are now able to beat this limit and approach the Heisenberg limit $S/N \sim N$ but with limited atom numbers or coherence time so far. This field of quantum metrology and quantum sensing with correlated particles is extremely active today.

Over 15 fountain devices are today in operation in National Metrology Institutes around the world. They are permanently compared by GNSS satellite methods (or optical fibers on the ground) and they serve to steer the TAI (Temps Atomique International) and UTC (Universal Coordinated Time) computed by the Bureau International des Poids et

Mesures, (BIPM). The accuracy of cesium fountain devices approaches 1-2 parts in 10^{16} , meaning that the SI second, the unit of time intervals, is realized with this uncertainty. This is equivalent to an error that is less than one second over 300 million years! To reach this level of accuracy, we have developed a set of original methods to evaluate the various systematic frequency shifts that affect the cesium hyperfine resonance (magnetic field, cold collision shift, blackbody radiation shift, cavity pulling...) [Sortais 2000, Pereira dos Santos, 2002, Bize 2005].

Atomic fountains are not restricted to time measurements. They are also used to realize atom interferometers which are extremely sensitive to external forces such as gravity acceleration, or rotations [Atom interferometry 2002]. Atom interferometers are now used for Earth gravity mapping and for inertial navigation systems. The French Exail company now sells commercial quantum gravimeters which have been largely inspired by the design of Paris Observatory cold atom fountains.

Rubidium Fountain Clocks

1995 was the year of the discovery of Bose-Einstein Condensation (BEC) in a dilute gas of rubidium atoms, for which E. Cornell, W. Ketterle and Carl Wieman were awarded the Nobel Prize in 2001. In this search, the collisional properties of rubidium 87 atoms were measured, and it was shown that they were much more favorable for BEC than for cesium atoms, which display an anomalously large collision cross-section at low temperature. The frequency shift due to cold collisions in cesium clocks is energy dependent and large. It represents the main limitation to the stability and accuracy of fountain primary frequency standards. In 1996, André Clairon and I proposed building a rubidium fountain that could display better stability and accuracy than cesium ones. The first Rubidium cold atom fountain was built at Paris Observatory [Bize1998] simultaneously with K. Gibble's group at Penn State University. As predicted, the rubidium fountain indeed displayed a very small collision shift, which was disentangled from cavity pulling, and about 30 times smaller than in cesium [Sortais 2000]. This enables one to operate the fountain with a larger number of atoms hence an improved frequency stability, while preserving a low collisional frequency shift. For this reason, the ground master clock running the American GPS satellite navigation system at USNO is based today on a collection of 5 rubidium fountains. Since 2004, the rubidium hyperfine transition is also recommended by BIPM as a

secondary representation of the SI second and is routinely used in TAI computation.

Fundamental Physics Tests

A second advantage of a rubidium fountain is that it enables one to perform new tests in fundamental physics. The first example is a search for variations of some fundamental constants of physics, such as the fine structure constant, α , that characterizes the strength of the electromagnetic interaction, or the ratio of electron mass to proton mass. Most unification theories which attempt to unify gravity with the Standard Model of particle physics do predict the possibility of changing fundamental constants or the existence of new particles such as dilatons or axions. By measuring the ratio of the hyperfine frequencies of rubidium 87 and cesium 133 over a period of 5 years, we were able to establish a new, much stricter upper bound on the temporal drift of this ratio, at the level of $7 \cdot 10^{-16}$ per year [Marion 2003]. Since then, measurements have been extended over an 18-year period in a dual Rb-Cs fountain by Jocelyne Guéna, Sebastien Bize and Peter Wolf at Paris Observatory [Hess 2016]. This research has been particularly active in the last two decades with important advances, in particular at NIST, USA, Paris Observatory, PTB in Germany and Riken in Japan involving also optical clocks and gaining a further two orders of magnitude in sensitivity. The current limit on α changes is lower than $2.5 \cdot 10^{-19}/\text{year}$ [Filzinger 2023].

Bloch Oscillations, Atom Interferometry, and the Measurement of the Fine Structure Constant

When pushing the limits of laser cooling with Raman transitions into the nanokelvin range at ENS, my collaborators and I have reached a regime where atoms could be cooled to a temperature below the single photon energy divided by the Boltzmann constant. Quantum mechanics tells us that the atom's de Broglie wavelength becomes on the order of – or larger than – the wavelength of the cooling light. In this regime, a fully quantum mechanical treatment of the atomic motion should be used, in similarity with the motion of electrons in a periodic crystal. When prepared in the fundamental band of a periodic potential and submitted to a constant external force (an electric field for electrons), particles (atoms or electrons) should display an oscillatory motion instead of a drift as in standard electrical conduction. This purely quantum effect is extremely hard to observe in solid state materials and

our group made the first direct observation of Bloch oscillations with cold atoms [Ben Dahan 1996]. Since this observation, the method has been developed in various applications such as the measurement of Berry curvature in topological lattices, or as a precision accelerator [Peik 1997]. Another important application of Bloch oscillations is a high precision measurement of the fine structure constant α . This is provided through the measurement of the single photon recoil when an atom emits or absorbs a single photon. In 2020, the Cladé/Guellati group at LKB-ENS used Bloch oscillations combined with Ramsey spectroscopy on cold rubidium atoms to obtain the most precise measurement to date of the fine structure constant [Morel 2020].

PHARAO: a Cold Atom Clock in Microgravity

Realizing that gravity was a major perturbation for cesium atoms at microkelvin temperatures, I proposed in 1991 to the French Space Agency CNES to explore the possibility of realizing an atomic clock with cold atoms in orbit where the gravity acceleration is vastly reduced. First demonstration experiments onboard jet plane Zero g parabolic flights were made in 1992 [Lounis 1992], followed by the realization of a cold cesium clock prototype tested in parabolic flights in 1997 [Laurent 1998]. A very substantial instrumental effort has been made to build a rugged, reliable and transportable cesium clock capable of supporting the 2g-0g-2g acceleration sequences occurring in the jet plane.

This cesium clock has been subsequently transformed into a mobile fountain (FOM) serving as a transportable primary cesium standard. FOM was transported several times to the Max Planck Institute for Quantum Optics in Garching (DE) in the group of Professor T.W. Hänsch. A direct frequency measurement of the hydrogen 1s-2s transition frequency was made using the newly proposed frequency comb technique of T.W. Hänsch, which linked the optical frequency domain to the microwave domain [Niering 2004]. The 2005 Nobel Prize was awarded to T.W. Hänsch and J.L. Hall for this invention. Two subsequent transports of FOM to Garching in 2004 and 2011 enabled the Paris/Garching group to improve the 1s-2s hydrogen absolute frequency to $4.2 \cdot 10^{-15}$ in relative value, as well as to refine limits on drifts of fundamental constants [Fischer 2004, Parthey 2011]. These high precision measurements in the simplest of the atom, hydrogen, represent a stringent test of the quantum electrodynamics (QED) theory.

ACES/PHARAO: Atomic Clock Ensemble in Space

Following the demonstration of a cesium clock in parabolic flights, and answering a call for flight opportunities on the International Space Station (ISS), together with the Paris Observatory team, in 1997 I proposed a mission concept called ACES (Atomic Clock Ensemble in Space) to the European Space Agency, ESA, and the French Space Agency, CNES [Spallicci 1997]. The cold atom clock PHARAO is joined by a Space Hydrogen Maser and two time transfer systems, from space to ground, one in the microwave domain, one in the optical domain using short laser pulses of light. A precise orbit determination, provided by an onboard GNSS receiver, and an onboard computer complement the payload.

After 25 years of advanced technical developments in European industry, France (PHARAO clock), Switzerland (Space Hydrogen Maser, SHM), Germany (Time transfer system in the microwave domain, MWL), and the Czech Republic (Laser link, ELT), the ACES payload has been launched towards the ISS on April 21, 2025 by a Space X Falcon rocket. This long development period stems from the fact that building space qualified equipment with low mass, low power and sufficient reliability at the state of the Art in Time and Frequency metrology requires innovative instrumentation and testing as well as sufficient funding [Salomon 2001, Laurent 2006, Cacciapuoti 2009, Laurent 2015, Laurent 2020, Cacciapuoti 2020].

Since April 28, 2025, the Atomic Clock Ensemble in Space (ACES) hardware assembled by Airbus as a main contractor to ESA is installed on the European Columbus module of the ISS on an external platform pointing towards the Earth. ACES is run by the European Space Agency. The French Space agency, CNES, hosts the ACES control center in Toulouse and the mission data repository which is accessed by the ACES Science team. The main data analysis center is located at LTE under the responsibility of Peter Wolf. During the commissioning phase, the flight instruments have been switched on and characterized. The cold cesium clock has already produced Ramsey resonances 4 times narrower than in cesium fountains on Earth with a 2 second coherence time. The microwave time transfer system begins to produce meaningful time comparisons between PHARAO and ground clocks and the precision of the ISS orbit determination is better than 1 meter.

ACES/PHARAO Scientific Objectives

Once all instruments are well characterized, the ACES mission will enter at the beginning of 2026 the Science phase, which will last 2 and ½ years. The scientific objectives include the demonstration of a cold cesium primary frequency standard in micro-gravity with increased coherence time and with $2 \cdot 10^{-16}$ relative accuracy, a 2ppm test of Einstein's gravitational redshift (one order of magnitude gain over present tests), and a search for time variations of fundamental constants and for light dark matter. With its advanced time transfer systems offering one to two orders of magnitude gain over GPS-based distant clock comparisons, ACES will also create a global comparison network of ultra-precise clocks, involving both microwave and optical frequency standards. This is particularly important in the context of the future redefinition of the second of the International System of Units which will be based on optical clocks which now surpass microwave clocks by a factor 100 [Bromley 2018, Aeppli 2024]. Intercontinental frequency comparisons at the 10^{-17} - 10^{-18} level will provide sufficient confidence for supporting the new definition with atomic clocks operating in the visible or UV part of the electromagnetic spectrum.

The ACES mission involves an international collaboration of scientists from the main metrology institutes worldwide: LNE at Paris Observatory (FR), PTB (DE), Wetzell Observatory (DE), NPL (UK), NIST (USA), JPL(USA), NICT (JP), RIKEN(JP), INRIM (IT), and METAS (CH). They are located in 7 different countries, 5 in Europe, 1 in Japan and 2 in the USA. With their advanced microwave and optical clocks, they will all contribute to the scientific objectives of the mission by comparing their timing signals to that emitted by the PHARAO space clock. In addition, a number of laser ranging stations around the globe will perform high accuracy time transfer experiments between the space clock and ground clocks via picosecond pulses of laser light.

Summary

In conclusion, thanks to this scientific journey, I have had the opportunity to participate in a dynamic quest to achieve ever greater precision in the measurement of time. Given the rapid progress made in this field of research over the past few decades, one may wonder whether this pace of improvement will continue in the decades to come. The 20th century saw the birth of two highly successful theories, general relativity and quantum mechanics. However, general relativity is a classical theory that does not easily unify with quantum mechanics.

At a certain level, this unification could impose a fundamental limit on the precision of time interval measurements. Exploring this limit is a strong motivation for conducting new experiments at the boundary between these two theories.

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