



# The Quantum Physics of Atomic Frequency Standards

---

Recent Developments

Jacques Vanier • Cipriana Tomescu



CRC Press  
Taylor & Francis Group

# The Quantum Physics of Atomic Frequency Standards

---

## Recent Developments

Jacques Vanier

Université de Montréal, Montréal, Canada

Cipriana Tomescu

Université de Montréal, Montréal, Canada



**CRC Press**

Taylor & Francis Group

Boca Raton London New York

---

CRC Press is an imprint of the  
Taylor & Francis Group, an **informa** business

CRC Press  
Taylor & Francis Group  
6000 Broken Sound Parkway NW, Suite 300  
Boca Raton, FL 33487-2742

© 2016 by Taylor & Francis Group, LLC  
CRC Press is an imprint of Taylor & Francis Group, an Informa business

No claim to original U.S. Government works

Printed on acid-free paper  
Version Date: 20150617

International Standard Book Number-13: 978-1-4665-7695-7 (Hardback)

This book contains information obtained from authentic and highly regarded sources. Reasonable efforts have been made to publish reliable data and information, but the author and publisher cannot assume responsibility for the validity of all materials or the consequences of their use. The authors and publishers have attempted to trace the copyright holders of all material reproduced in this publication and apologize to copyright holders if permission to publish in this form has not been obtained. If any copyright material has not been acknowledged please write and let us know so we may rectify in any future reprint.

Except as permitted under U.S. Copyright Law, no part of this book may be reprinted, reproduced, transmitted, or utilized in any form by any electronic, mechanical, or other means, now known or hereafter invented, including photocopying, microfilming, and recording, or in any information storage or retrieval system, without written permission from the publishers.

For permission to photocopy or use material electronically from this work, please access [www.copyright.com](http://www.copyright.com) (<http://www.copyright.com/>) or contact the Copyright Clearance Center, Inc. (CCC), 222 Rosewood Drive, Danvers, MA 01923, 978-750-8400. CCC is a not-for-profit organization that provides licenses and registration for a variety of users. For organizations that have been granted a photocopy license by the CCC, a separate system of payment has been arranged.

**Trademark Notice:** Product or corporate names may be trademarks or registered trademarks, and are used only for identification and explanation without intent to infringe.

Visit the Taylor & Francis Web site at  
<http://www.taylorandfrancis.com>

and the CRC Press Web site at  
<http://www.crcpress.com>



Printed and bound in Great Britain by  
TJ International Ltd, Padstow, Cornwall

# **The Quantum Physics of Atomic Frequency Standards**

---

**Recent Developments**



---

# Preface

Volumes 1 and 2 of *The Quantum Physics of Atomic Frequency Standards*, henceforth referred to as *QPAFS* (1989), were written in the 1980s and were published in 1989. They covered, in some detail, work done up to 1987 on the development of atomic frequency standards. The text included a description of their development at that time, as well as a description of the research on the physics supporting that development. Since that time, the field has remained a very active part of the research program of many national laboratories and institutes. Work has remained intensive in many sectors connected to the refinement of classical frequency standards based on atoms such as rubidium (Rb), caesium (Cs), hydrogen (H), and selected ions in the microwave range, while new projects were started in connection to the realization of stable and accurate frequency standards in the optical range.

For example, intensive studies were made on the use of lasers in the optical pumping and cooling of Rb and Cs as well as on the development of a new type of standard based on the quantum-mechanical phenomenon called coherent population trapping (CPT). Regarding Cs and Rb, laser cooling of atoms has made possible the realization of an old dream in which a small blob of atoms, cooled in the microkelvin range, is projected upward at a slow speed in the gravitational field of the earth and the atoms fall back like water droplets in a fountain. In their path, the atoms are made to pass through a microwave cavity, and upon falling back after having spent their kinetic energy, pass through the same cavity, mimicking, with a single cavity, the classical double-arm Ramsey cavity approach. The system is called *atomic fountain*. Its advantage over the classical approach resides in the reduction of the width of the resonance hyperfine line by a factor of the order of 100 relative to that observed in the room temperature approach. The resulting line width is of the order of 1 Hz. Work has also continued on the development of smaller H masers, in particular in the development of passive devices and in the use of a new smaller so-called *magnetron cavity*. The advent of the solid-state laser in the form of the conventional edge-emitting type (GaAs) and vertical structure (VCSEL) has opened the door to a new approach in optical pumping for implementing smaller and more performing Rb and Cs cell frequency standards.

Since the 1990s, laser cooling has been studied extensively and aside from providing a means for realizing the fountain clock mentioned above, it has allowed the realization of clocks based on microwave transitions in ions such as mercury ( $\text{Hg}^+$ ), barium ( $\text{Ba}^+$ ), strontium ( $\text{Sr}^+$ ), and ytterbium ( $\text{Yb}^+$ ) confined within an electromagnetic trap.

On the other hand, intense work has been carried out in several laboratories in extending the work done at microwave frequencies to the optical frequency range. The gain in that approach relies mainly on the increase in the frequency of the atomic transitions involved, which provides for a line width similar to that obtained in the microwave range a resonance quality factor millions of times larger. Laser cooling has been applied successfully to such atoms as mercury (Hg), ytterbium (Yb), and strontium (Sr)

stored in optical lattice traps in order to reduce their thermal motion. Laser cooling has also been used in the mono-ion trap to implement optical frequency standards. In that case, a single ion, say  $\text{Sr}^+$  or  $\text{Yb}^+$ , is maintained in a Paul or Penning trap and its motion within the trap is damped by laser cooling. Clocks at optical frequencies have been implemented as laboratory units with unsurpassed accuracy and frequency stability reaching the  $10^{-16}$  to  $10^{-18}$  range. In both cases, the clock frequency is derived from a transition between the ground S state of the atom and an excited metastable state with a lifetime of the order of 1 second or more leading to a very narrow resonance line. The clock transition is detected by means of monitoring changes in the fluorescence level created by the cooling radiation when the clock transition is excited.

The large gap in frequency between the microwave and the optical range has always been an roadblock in the use of optical frequencies in various applications such as frequency standards or still high precision spectroscopy and fundamental research. The reason is mainly due to the fact that gaps between available optical frequencies for the realization of clocks are very large. It is extremely difficult to connect those frequencies to the microwave range. This connection is required because most of the applications are in the low frequency range of the spectrum and, furthermore, because the SI (International System of Units) definition of the second is based on a microwave hyperfine transition in Cs, in the X band. We have given in Volume 2 of *QPAFS* examples of the conventional method used to make that connection. That method comprises frequency- and phase-locking together banks of lasers with appropriate heterodyning in several steps in order to interconnect various optical frequencies to reach finally the microwave range. The connection has to be done over a large number of steps and involves tremendous investment of space and time to finally measure what very often happens to be just a single frequency. Such a task has been reduced considerably by the invention of the so-called *optical comb*, which comprises locking the repetition rate of a femtosecond laser to a stable atomic frequency standard of high spectral purity, such as an H maser referenced in frequency to a primary Cs atomic clock. When observed by means of a nonlinear optical fibre, the resulting laser spectrum consists of a spectrum of sharp lines, themselves called the *teeth of the comb*, which covers a frequency range of the order of 1 octave. Frequencies over a broad range are then measured essentially in a single step on an optical table, resulting in a considerable reduction in work and size as compared to the previous heterodyning technique, which required entire rooms filled with lasers.

This volume covers those subjects in some detail. It is divided into five chapters. Chapter 1 is an introduction, presenting a review of recent developments made on the improvement of conventional atomic frequency standards described in the two volumes of *QPAFS*. It highlights the main limitations of those frequency standards and the physical basis of those limitations and outlines the progress made during the last 25 years. Chapter 2 is a description of recent advances in atomic physics, theory and applications, that opened new avenues. Chapter 3 is concerned with research and development done in the development of new microwave frequency standards. Chapter 4 describes research and development done in the optical range to implement optical frequency standards based on new results in atomic physics as described in Chapter 2. Chapter 5 summarizes the results in frequency stability and accuracy

achieved with those new frequency standards and outlines selected applications. A short reflection is included giving some insight into future work.

Such a text cannot be written without significant help from experts in the field. We wish to recognize the contribution and collaboration of many scientists. In particular, we wish to recognize the invaluable help of André Clairon, who has read the whole manuscript and helped in improving its exactness and completeness. We also show our gratitude to the following scientists who helped us through their encouragement, supplied original figures or material, and contributed by means of comments on various sections of the text: C. Affolderbach, A. Bauch, S. Bize, J. Camparo, C. Cohen-Tannoudji, E. De Clercq, A. Godone, D. Goujon, S. Guérandel, P. Laurent, T. Lee, S. Micalizio, G. Mileti, J. Morel, W.D. Phillips, P. Rochat, P. Thomann, R.F.C. Vessot, and S. Weyers.

**Jacques Vanier**

and

**Cipriana Tomescu**

*University of Montreal*





---

# Introduction

This book is about recent developments in the field of atomic frequency standards, developments that took place after the publication in 1989 of the first two volumes with the same title. Atomic frequency standards are systems providing an electrical signal at a cardinal frequency of, say, 10 MHz, a signal generated usually by a quartz crystal oscillator locked in phase or in frequency to a quantum transition inside an atom. The atom is selected for its properties such as easy detection of the particular quantum transition chosen and relative independence of its frequency of the environment. In early work, those conditions limited development around hydrogen and alkali atoms, which have transitions in the microwave range and could be manipulated easily as beams or atomic vapour with the techniques available at that time. Progress in the development of lasers and their stabilization extended that work to the optical range. A major task encountered in the early development of microwave standards has always been the elimination of Doppler effect. Atoms at room temperature travel at speeds of several hundred metres per second and, consequently, Doppler effect causes frequency shifts and line broadening of the resonance signal. This effect is generally eliminated by means of various storage techniques based on Dicke effect, or still beam techniques using the Ramsey double-arm cavity approach. These techniques are not well adapted to optical frequencies because of the shorter wavelengths involved. However, progress in the understanding of interactions between atoms and electromagnetic interactions has provided new means of reducing the velocity of atoms and reducing, if not eliminating, the constraints introduced by Doppler effect.

An atomic frequency standard that is operated continuously becomes an atomic clock. The operation is essentially a process of integration and the date set as the constant of integration provides the basis for implementing a timescale. This is the origin of atomic timescales, in particular the one maintained by the International Bureau of Weights and Measures. Various systems in operation have their own timescale, for example, the global positioning system (GPS) of the United States, the Russian Glonass system, the Chinese Beidou system, and the European Galileo systems under development, all playing an important role in navigation on or near the surface of the earth.

Although time is central to physics and is used in our day-to-day life, it is a concept that is difficult to grasp, let alone to define. We use it without questioning its origin and its exact nature. It is basic in physics for describing the dynamics of systems and ensembles of systems by means of equations that model the evolution of *objects* forming our universe. The concept is used as such without questioning much its exact nature and origin. In Newtonian mechanics, objects evolve in space and their behaviour is described by means of differential equations and functions of time and space. Both space and time are independent and in common language they are said to be absolute. In that context, time is not a function of space and space is not a function of time. However, in attempts to relate mechanics and electromagnetism by

space and time transformations, a difficulty arose. This is due to the finiteness and invariability of the speed of light, made explicit in Maxwell's equations, whatever the motion of the frame of reference in which it is generated and measured. In this context, with Einstein, Poincaré, Lorentz, Minkowski, and others, time and space become entangled and functions of each other. There is no such thing as an absolute space in which objects evolve in an absolute time framework, both independent of each other. Time and space form a single four-dimensional framework and cannot be treated independently. This concept forms the basis of the theory of relativity. This theory has been shown to be valid through multiple experiments and verifications to a level that raises its validity to a high degree. It should be pointed out that the most accurate verifications were done with atomic clocks, the instruments that are the content of this book. There is another question also often raised regarding the nature of time: Could it be discrete? If so what would be the size of its smallest quantity, the time quantum? Could it be that Planck's time is the smallest time entity? This is a totally unknown subject and appears to be a roadblock to in the development of a sustainable quantum theory that includes the concepts elaborated in the theory of general relativity.

Although we may feel somewhat uncomfortable in the context of such questions, time remains the most basic concept in physics, is fundamental, and is the quantity that is measured with the greatest precision. Current atomic clocks can commonly keep time to an accuracy of 1 s in a million years, or in other words are stable to better than 1  $\mu$ s in a year. For example, the timescale generated by the GPS satellites for navigation, based on atomic frequency standards on satellites and on ground, is stable after appropriate processing and filtering to about 1 ns/day. On the other hand, on the basis of our inability to measure time by astronomical means with such accuracy, it was decided in 1967 to replace the astronomical definition of the second by one in terms of a particular atomic hyperfine transition in the Cs atom. The frequency of that transition is set at 9,192,631,770 Hz. Furthermore, since now the speed of light is defined exactly as 299,792,458 m/s, providing at the same time a definition of the metre, the mechanical units of the SI become essentially determined by the basic time unit, the second. The concept of unifying all SI units in terms of a single quantity goes further due to the Josephson effect phenomenon, which relates voltage to frequency in a most fundamental expression,  $2e/h$ , involving only fundamental constants. This is the subject that will be described in Chapter 5.

From this discussion, it is evident that time plays a most important role in physics and technology and the realization of the highest accuracy and precision of the SI unit, the second, has remained one of the most active preoccupations of several laboratories and institutes over the past 50 years. Starting with tremendous improvements in the realization of the second within the microwave range, work has extended to the optical range with proven increase in frequency stability and accuracy by several orders of magnitude. These achievements were possible mainly through a better understanding of the interactions between electromagnetic radiation and atoms, providing a means of altering the properties of atoms. This book is about those improvements that have taken place mainly during the past 25 years, on the realization of stable and accurate frequency standards.

---

# Authors

**Jacques Vanier** completed his undergraduate studies in physics at the University of Montreal, Québec, Canada, before moving to McGill University to undertake his graduate studies. During his career he has worked in various industries (Varian, Hewlett-Packard); taught physics; and carried out research at Laval University, Montreal, Québec, Canada, and has also been an active member of the National Research Council of Canada, in Ottawa, Ontario, Canada. His research work is oriented towards the understanding and the application of the quantum electronics phenomena and he has been a consultant for several companies engaged in the development of atomic clocks. Jacques has also been very active on the academic circuit, giving lectures and presenting at numerous conferences in universities, national institutes, and summer schools around the world. He has written more than 120 journal articles and proceedings papers and is the author of review articles and books on masers, lasers, and atomic clocks. His book *The Quantum Physics of Atomic Frequency Standards*, written with C. Audoin, is recognized as a main reference in the field. He is the author of *The Universe: A Challenge to the Mind* published by Imperial College Press/World Scientific. Jacques is a fellow of the Royal Society of Canada, the American Physical Society, and the Institute of Electrical and Electronic Engineers. He has received several awards for his contributions to the field of measurement science. He is currently an adjunct professor in the Physics Department, University of Montreal, Québec, Canada.



**Cipriana Tomescu** completed her studies in physics at the University of Bucharest, Romania, where she obtained her PhD degree.

From 1982 to 2004, she was a researcher at the National Institute of Laser Physics, Plasma and Radiation, Bucharest, Romania. In the early years of her employment, she participated in the construction of H masers used by the Bucharest Observatory, the Institute of Metrology, and the Faculty of Physics. During the period 1996–2004, she was laboratory director. During the period 1992–2006, she also worked in various national laboratories, in particular, Paris Observatory, LNE-SYRTE, France; Neuchâtel Observatory, Switzerland; and Communication Research Laboratory, Japan. At those locations, she contributed to the development of advanced state-of-the-art atomic frequency standards, such as Rb and Cs fountains using atom trapping techniques and laser atom cooling. From 2008 to 2012, she worked at the University of Liege, IPNAS, and at Gillam-Fei. She was responsible for the implementation of the first H maser realized in Belgium under



Plan Marshall: SKYWIN-TELECOM. She is the author of numerous publications in scientific journals and conference proceedings and she has been invited to make presentations at numerous symposia, universities, and national institutes. In 1985, she received the D. Hurmuzescu Prize of the Romanian Academy for work on the physics of the H maser. She is currently an invited researcher in the Physics Department of the University of Montreal, Québec, Canada.

---

# Contents

Preface..... xiii  
Introduction..... xvii  
Authors..... xix

**Chapter 1** Microwave Atomic Frequency Standards: Review and Recent Developments ..... 1

1.1 Classical Atomic Frequency Standards..... 2

1.1.1 Cs Beam Frequency Standard ..... 2

1.1.1.1 Description of the Approach Using Magnetic State Selection ..... 3

1.1.1.2 Review of Frequency Shifts and Accuracy .... 7

1.1.1.3 Frequency Stability of the Cs Beam Standard ..... 15

1.1.1.4 Recent Accomplishments ..... 16

1.1.2 Hydrogen Maser ..... 33

1.1.2.1 Active Hydrogen Maser ..... 33

1.1.2.2 Passive Hydrogen Maser ..... 48

1.1.2.3 Frequency Stability of the Hydrogen Maser ..... 53

1.1.2.4 State of the Art of Recent Developments and Realizations..... 57

1.1.3 Optically Pumped Rb Frequency Standards ..... 69

1.1.3.1 General Description..... 69

1.1.3.2 State-of-the-Art Development ..... 71

1.2 Other Atomic Microwave Frequency Standards ..... 82

1.2.1  $^{199}\text{Hg}^+$  Ion Frequency Standard ..... 83

1.2.1.1 General Description..... 83

1.2.1.2 Frequency Shifts ..... 85

1.2.1.3 Linear Trap ..... 88

1.2.2 Other Ions in a Paul Trap ..... 90

1.2.2.1  $^{171}\text{Yb}^+$  and  $^{173}\text{Yb}^+$  Ion Microwave Frequency Standards ..... 91

1.2.2.2  $^{201}\text{Hg}^+$  Ion Microwave Frequency Standard ..... 92

1.3 On the Limits of Classical Microwave Atomic Frequency Standards ..... 93

Appendix 1.A: Formula for Second-Order Doppler Shift..... 94

Appendix 1.B: Phase Shift between the Arms of Ramsey Cavity ..... 95

Appendix 1.C: Square Wave Frequency Modulation and Frequency Shifts.....	95
Appendix 1.D: Ring Cavity Phase Shift .....	97
Appendix 1.E: Magnetron Cavity .....	98

<b>Chapter 2</b>	<b>Recent Advances in Atomic Physics That Have Impact on Atomic Frequency Standards Development .....</b>	<b>101</b>
2.1	Solid-State Diode Laser.....	102
2.1.1	Basic Principle of Operation of a Laser Diode.....	102
2.1.2	Basic Characteristics of the Semiconductor Laser Diode .....	105
2.1.3	Types of Laser Diodes .....	106
2.1.4	Other Types of Lasers Used in Special Situations.....	108
2.2	Control of Wavelength and Spectral Width of Laser Diodes .....	109
2.2.1	Line Width Reduction .....	109
2.2.1.1	Simple Optical Feedback.....	109
2.2.1.2	Extended Cavity Approach.....	109
2.2.1.3	Feedback from High- $Q$ Optical Cavities ...	112
2.2.1.4	Electrical Feedback .....	112
2.2.1.5	Other Approaches .....	112
2.2.1.6	Locking the Laser to an Ultra-Stable Cavity.....	113
2.2.2	Laser Frequency Stabilization Using an Atomic Resonance Line .....	116
2.2.2.1	Locking the Laser Frequency to Linear Optical Absorption .....	116
2.2.2.2	Locking the Laser Frequency to Saturated Absorption.....	117
2.3	Laser Optical Pumping.....	119
2.3.1	Rate Equations .....	120
2.3.2	Field Equation and Coherence .....	122
2.4	Coherent Population Trapping.....	127
2.4.1	Physics of the CPT Phenomenon .....	129
2.4.2	Basic Equations .....	131
2.5	Laser Cooling of Atoms .....	136
2.5.1	Atom–Radiation Interaction.....	138
2.5.1.1	Effect of a Photon on Atom External Properties: Semi-Classical Approach.....	138
2.5.1.2	Quantum Mechanical Approach.....	143
2.5.2	Effect of Fluctuations in Laser Cooling and Its Limit .....	158
2.5.3	Cooling below Doppler Limit: Sisyphus Cooling ....	160
2.5.3.1	Physics of Sisyphus Cooling.....	160
2.5.3.2	Capture Velocity .....	164

2.5.3.3	Friction Coefficient .....	165
2.5.3.4	Cooling Limit Temperature .....	166
2.5.3.5	Recoil Limit.....	166
2.5.3.6	Sub-Recoil Cooling .....	167
2.5.4	Magneto-Optical Trap.....	167
2.5.5	Other Experimental Techniques in Laser Cooling and Trapping.....	170
2.5.5.1	Laser Atom-Slowing Using a Frequency Swept Laser System: Chirp Laser Slowing.....	171
2.5.5.2	Laser Atom-Slowing Using Zeeman Effect: Zeeman Slower .....	173
2.5.5.3	2D Magneto-Optical Trap .....	177
2.5.5.4	Isotropic Cooling .....	180
2.5.5.5	Optical Lattice Approach .....	183
	Appendix 2.A: Laser Cooling—Energy Considerations.....	189

**Chapter 3** Microwave Frequency Standards Using New Physics ..... 191

3.1	Cs Beam Frequency Standard .....	192
3.1.1	Optically Pumped Cs Beam Frequency Standard.....	192
3.1.1.1	General Description.....	192
3.1.1.2	Frequency Shifts and Accuracy .....	194
3.1.1.3	Experimental Determination of Those Shifts.....	197
3.1.1.4	Frequency Stability .....	198
3.1.1.5	Field Application .....	200
3.1.2	CPT Approach in a Beam.....	200
3.1.2.1	General Description.....	200
3.1.2.2	Analysis .....	201
3.1.2.3	Experimental Results.....	206
3.1.3	Classical Cs Beam Standard Using Beam Cooling.....	208
3.2	Atomic Fountain Approach .....	210
3.2.1	In Search of a Solution .....	210
3.2.2	General Description of the Cs Fountain.....	211
3.2.3	Functioning of the Cs Fountain.....	213
3.2.3.1	Formation of the Cooled Atomic Cloud: Zone A .....	213
3.2.3.2	Preparation of the Atoms: Zone B.....	217
3.2.3.3	Interrogation Region: Zone C.....	218
3.2.3.4	Free Motion: Zone D .....	218
3.2.3.5	Detection Region: Zone E.....	218
3.2.4	Physical Construction of the Cs Fountain.....	219
3.2.4.1	Vacuum Chamber .....	219
3.2.4.2	Microwave Cavity .....	220



3.2.4.3	Magnetic Field .....	221
3.2.4.4	Temperature Control .....	221
3.2.4.5	Capture and Selection Zone .....	221
3.2.4.6	Detection Zone .....	221
3.2.4.7	Supporting Systems .....	221
3.2.4.8	Advantages and Disadvantages of a Pulsed Fountain .....	222
3.2.5	Frequency Stability of the Cs Fountain .....	223
3.2.5.1	Photon Shot Noise .....	224
3.2.5.2	Quantum Projection Noise .....	225
3.2.5.3	Electronic Noise .....	225
3.2.5.4	Reference Oscillator Noise: Dicke Effect ...	225
3.2.6	Rubidium and Dual Species Fountain Clock .....	226
3.2.7	Frequency Shifts and Biases Present in the Fountain .....	229
3.2.7.1	Second-Order Zeeman Shift .....	230
3.2.7.2	Black Body Radiation Shift .....	232
3.2.7.3	Collision Shift .....	237
3.2.7.4	Cavity Phase Shift .....	240
3.2.7.5	Cavity Pulling .....	242
3.2.7.6	Microwave Spectral Purity .....	247
3.2.7.7	Microwave Leakage .....	247
3.2.7.8	Relativistic Effects .....	248
3.2.7.9	Other Shifts .....	249
3.2.7.10	Conclusion on Frequency Shifts and Accuracy .....	250
3.2.8	An Alternative Cold Caesium Frequency Standard: The Continuous Fountain .....	251
3.2.8.1	Light Trap .....	252
3.2.8.2	Interrogation Zone, Microwave Cavity .....	253
3.2.8.3	Preliminary Results .....	255
3.2.9	Cold Atom PHARAO Cs Space Clock .....	257
3.3	Isotropic Cooling Approach .....	258
3.3.1	External Cavity Approach: CHARLI .....	258
3.3.2	Approach Integrating Reflecting Sphere and Microwave Cavity: HORACE .....	260
3.3.3	Different HORACE Approach .....	261
3.4	Room Temperature Rb Standard Approach Using Laser Optical Pumping .....	262
3.4.1	Contrast, Line Width, and Light Shift .....	263
3.4.2	Effect of Laser Radiation Beam Shape .....	272
3.4.3	Expectations Relative to Short-Term Frequency Stability .....	273
3.4.4	Review of Experimental Results on Signal Size, Line Width, and Frequency Stability .....	273