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Atomic clocks: A brief history and current status of research in India

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Abstract. Frequency corresponding to the energy difference between designated levels of an atom provides precise reference for making a universally accurate clock. Since the middle of the 20th century till now, there have been tremendous efforts in the field of atomic clocks making time the most accurately measured physical quantity. National Physical Laboratory India (NPLI) is the nation's timekeeper and is developing an atomic fountain clock which will be a primary frequency standard. The fountain is currently operational and is at the stage of complete frequency evaluation. In this paper, a brief review on atomic time along with some of the recent results from the fountain clock will be discussed.

Keywords. Atomic physics; frequency standards; microwaves; diode lasers.

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1. A brief history of atomic time

1.1 *Time measurement*

Time is one of the seven fundamental physical quantities in the International System of Units (SI) [1]. Among all the physical quantities, time interval and frequency are the ones that can be measured with least uncertainty and best resolution. Continued research and advanced technology has made it possible to realize the SI second accurate to 16 places after the decimal point ($\approx 1 \times 10^{-16}$). The fundamental concepts necessary to understand all about time and frequency are well compiled in [2].

Precise time measurement is essential for a number of systems which includes global positioning systems, synchronization of telecommunication system, secure communications, measurement of fundamental physical constants and other units, detection of the gravitational waves, geodesy, deep space navigation, radio telescopes, air traffic control, stock exchange, space navigation, and satellite systems.

A clock is simply a combination of an oscillator and a counter. Highly stable periodic oscillations are produced by an oscillator and the number of oscillations are counted by a counter and displayed. The quality of an oscillator is determined by its degree of stability and accuracy and its capacity to remain immune to environmental changes. Any uncertainty or change in the frequency of the oscillator will result in a corresponding uncertainty or change in the timekeeping accuracy of the clock.

1.2 *Pre-atomic era*

Till the end of the middle ages, different instruments which were used as clocks included great movements of the Sun, Moon, and stars, stone structures like Stonehenge, sundials, water clocks, sand glasses and marked candles [3]. As the civilization progressed, a variety of modifications were made, but a major breakthrough in the history of clocks came with the discovery of the process of oscillation by the Italian researcher Galileo Galilei. Based on the principle of oscillation many clocks were discovered. There were mechanical clocks, quartz clocks and finally atomic clocks were discovered [4].

In the pre-atomic era, mechanical and electrical oscillators served as the laboratory standards for time interval and frequency measurements. The first practical pendulum clock based on the principles first outlined by Galileo Galilei was invented by the Dutch physicist Christian Huygens in 1656. Their daily error was of the order of 10 s, or a relative error of only 10^{-4}. Pendulum clocks were then succeeded by quartz crystal oscillators, which were based on the phenomenon of piezoelectricity discovered by P Curie in 1880. It resonated at a nearly constant frequency when an electric current was applied. The invention of quartz crystal clock by the American scientist Warren A Marrison in 1929 immediately brought an improvement by an order of magnitude.

However, quartz oscillators are not ideal frequency standards since their resonance frequency depends on the size and shape of the crystal; and no two crystals can be exactly alike. Atoms and molecules have resonances which are stable over time and space; each chemical element and compound absorbs and emits electromagnetic radiation at its own 'characteristic frequencies'. Therefore, as an unperturbed atomic transition is identical from atom to atom, a group of atomic oscillators generate the same frequency, unlike that of a group of quartz oscillators. Also, unlike all electrical or mechanical resonators, atoms do not wear out with time and position. Furthermore, the frequency of a quartz oscillator changes slowly over time due to aging, and can change more rapidly due to the effects of environmental factors, such as temperature, humidity, pressure, and vibration [5], whereas the atoms do not change their properties over time. Thus, the uncertainty of a quartz crystal oscillator cannot be reduced beyond 10^{-9}. But, still it is used in everyday applications like wrist watches, wall clocks etc. where such a high accuracy is not needed. These limitations of the quartz oscillators along with the quest for better accuracy led to the development of atomic oscillators.

1.3 *Atomic beam clock*

In an atomic clock the frequency standard is based on a transition between energy levels in a quantum system, for instance an atom, an ion, or a molecule. In the present day, the atomic clocks are the best means of measuring time. A comparison of the different frequency sources is made in [6]. The two-volume book on atomic clocks and atomic frequency standards by Vanier and Audoin has comprehensive knowledge of this field [7].

On realizing the level of accuracy which can be obtained using an atomic standard of time, the existing astronomical definition of time based on Earth's rotation was changed to a very precise definition of the unit of time which is indispensable for science and technology. The SI second was defined in the 13th General Conference of Weights and Measures (CGPM) (1967/68, Resolution 1; CR, 103 and Metrologia, 1968, 4, 43) as: The second is the duration of 9,192,631,770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of the cesium 133 atom.

Cesium was chosen to define the second for a number of practical reasons. The perturbations on atomic energy levels are quite small in the low-density environment of a beam, and atomic beams of cesium are particularly easy to produce and detect. The frequency of the hyperfine transition that defines the second is relatively high and the linewidth can be made quite narrow by careful design. At the same time, the transition frequency is still low enough that it can be manipulated using standard microwave techniques and circuits [8]. Additionally, there is only one isotope (Cs-133) so that all atoms in the beam have the same hyperfine structure. The hyperfine splitting frequency of 9.19 GHz is relatively high compared to that of the other alkali atoms [9]. Cesium atoms are relatively heavy (133 a.m.u.), and they move at a relatively slow speed of about 130 m/s at room temperature allowing cesium atoms to stay in the interaction zone longer than hydrogen atoms, for example, which travel at a speed of about 1600 m/s at room temperature [10]. Moreover, cesium is non-poisonous and readily available. Again, cesium is one of the most easily laser-cooled and laser-manipulated elements, due to the fact that its level scheme is rather simple and that the 852-nm wavelength of its D2 line falls right into the range where silicon photodiodes are most efficient and high power, narrow linewidth laser diodes are available commercially [11]. Cesium is not the only atom that can be used as a reference for a frequency standard; transitions in rubidium and hydrogen are also commonly used. In order to relate their frequencies to the SI definition accurately, devices based on rubidium or hydrogen must be calibrated with respect to primary cesium-based devices.

The idea of using atomic resonance to measure time was proposed as early as the nineteenth century by Maxwell, Thomson and Tait in 1873 [12]. Many years later, in 1945, use of an atomic beam magnetic resonance as the basis of a clock was put forward by Rabi [13]. Ramsey and Silsbee in 1947 [14] invented the separated oscillatory field method for measuring the resonance of hydrogen molecular beam. This method improves the accuracy of the transition frequency determination. In 1949, the atomic timekeeping began with the construction of the first atomic clock. This standard, based on a resonance in the ammonia molecule, was constructed at the National Bureau of Standards (now National Institute of Standards and Technology) in the United States in a project led by Harold Lyons. The ammonia standard was quickly superseded by the cesium-beam frequency standard that forms the current basis for defining the second [15].

First ever Cs atomic clock based on the hyperfine fundamental transition of the cesium-133 atom was built by Louis Essen and J V L Parry at National Physical Laboratory

(NPL) in Teddington, England in June 1955, with a resonance width of 340 Hz and an accuracy of 1×10^{-9} [16]. The NPL device became the world's first cesium standard to be used on a regular basis for the calibration of secondary working frequency standards [17]. These are also known as thermal beam clocks. A thermal beam clock of beam of Cs atoms coming out of an oven, state selecting magnets, interaction with microwaves, and state interrogation at the end. It was also the first atomic beam frequency standard to operate in a 'closed loop' mode with an external microwave interrogation oscillator locked to the cesium hyperfine resonance using a servo control system along with the servo lock mechanism. The different types of atomic standards are discussed in [18,19]. After the realization of first thermal beam clock at NPL UK in 1955, major improvements were made in the design of the clock to increase the accuracy for the next 40 years. First commercial Cs clock named 'Atomichron' was manufactured in 1956 by a team led by Zacharias [20]. By 1966, HP started making the commercial Cs clocks [21]. The improved version of the commercial Cs clock HP 5071 with an accuracy of 1 in 10^{12} was later manufactured by Agilent and is now manufactured by Symmetricom Inc. The successful commercialization was led by the efforts from researchers at many laboratories around the world to improvise the clock design to increase the accuracy. Six different versions of thermal beam clocks named NBS-1 to NBS-6 were developed at NIST with successively increasing accuracy resulting from longer cavity lengths and velocity selective atomic selection. NIST-7, the last atomic beam standard developed at NIST, reached an accuracy of 5×10^{-15} due to the use of novel technique of optical pumping [22]. NIST-7 served as the primary standard till 1998 when it was replaced by the first fountain clock at NIST. High accuracy thermal beam clocks were developed at leading NMIs at Germany, France, UK, Canada etc. before Cs fountain took over as primary frequency standard in the late 1990s.

1.4 *Cesium fountain clock*

In 1990s the techniques of laser cooling and trapping were demonstrated and explained by Chu, Cohen-Tannoudji and Phillips [23–25]. These techniques significantly reduced the Doppler effect, which limited the performance of the thermal beam clocks. The concept of a cesium fountain was introduced in the 1950s by Jerrold Zacharias [26]. He planned to build a cesium beam clock vertically with one Ramsey interaction zone. Slow atoms from the cesium oven would traverse the microwave interaction zone while travelling upward, reverse their velocity under the influence of gravity, and traverse the same microwave interaction zone while travelling downward. The two interactions with the microwaves reproduced Ramsey's two-pulse interaction scheme, and a ballistic flight travelling only a metre upwards would give interaction times approaching one second instead of the 10 ms typical of thermal beam clocks up to that time. However, Zacharias' idea was successfully implemented by Steven Chu and coworkers at Stanford in the late 1980s with the incorporation of the laser cooling technique [27] resulting in the first working Cs atomic fountain clock. Finally, the first atomic fountain primary frequency standard, initially proposed by Zacharias was operated at BNM-LPTF (now LNE-SYRTE) in Paris in 1995 [28]. A fractional frequency uncertainty of 10^{-15} was obtained. Thus, the concept of fountain increased the interaction time of atoms with microwaves which was limiting the accuracy of thermal beam clocks. Since 1995, the microwave fountain clocks have been improved but mainly limited by the quantum fluctuations.

The success of the first primary frequency standard atomic fountain inspired other researchers in the metrology laboratories around the world to build laser-cooled cesium fountain clocks. The international atomic time (TAI) is the weighted average of time kept by atomic clocks all over the world. Cesium fountains, being the primary standards and most accurate clocks, get higher weight. Therefore, it is essential to increase the number of national primary frequency standards to contribute to the maintenance of the global time-scale. Currently there are ten Cs atomic fountain clocks that are contributing to the international atomic time [29]. These include: IEN-CSF1 (Italy) [30], NICT-CSF1 (Japan) [31], NIST-F1 (USA) [32], NMIJ-F1 (Japan) [33], NPL-CSF2 (UK) [34], PTB-CSF1 (Germany) [35], PTB-CSF2 (Germany) [36], SYRTE-FO1 (France) , SYRTE-FOM (France) [37] and SYRTE-FO2 (France) [38]. Apart from these, cesium atomic fountains are being developed by India [39], Russia [40], Mexico [41], China [42] and Korea [43].

1.5 *Future frequency standards*

Present atomic frequency standards are based on microwave transitions. These are likely to be replaced with frequency standards based on a forbidden transition in the optical frequency region. With similar linewidth as for microwave frequency standards, standards with optical transition have much higher resonance frequency (10^{15}) and thus can be accurate to 10^{-18} level. Intensive research is being carried out at leading NMIs on optical frequency standards based on single trapped ion or neutral atoms trapped in optical lattices [44–47]. At NPLI also, an optical frequency standard based on single trapped ytterbium ion is being developed [48]. It is expected that optical frequency standards will replace the Cs fountain as primary frequency standards in the next few years.

2. Cesium fountain clock at NPLI

At CSIR-NPL, development of India's first ever Cs fountain clock started only a few years ago [49]. The aim of the activity was to build an ultra-precise primary frequency standard for the first time in the country with a relative uncertainty at about 1×10^{-15} (such an atomic clock, when operated continuously, will neither loose nor gain a second in about a million years). Achieving this level of accuracy requires a remarkable combination of technological innovations in precision lasers, vacuum technology, magnetic shielding and advanced optical, electronic and mechanical systems besides control over the atomic and optical environment. The fountain is now fully operational and is at the stage of complete frequency evaluation. A brief description of the fountain and some preliminary results are discussed in the following sections.

2.1 *Description of NPLI atomic fountain*

In a Cs fountain clock, Cs atoms are cooled and launched up, passed through a microwave cavity on the way up and down and are probed for their state in the detection region. The

NPLI fountain has a (0, 0, 1) geometry of the magneto-optical trap (MOT) for cooling and launching operations [50]. The atoms are first loaded and cooled in MOT followed by further cooling in optical molasses (OM). They are launched using moving molasses and cooled further with polarization gradient cooling. The NPLI fountain clock has three major parts: physics package, optical set-up, electronics module. The detailed description about the various techniques related to clock operation is documented in [51–57]. The optical set-up and physics package of the Cs fountain clock (India-CsF1) developed at CSIR-NPL are shown in figure 1.

The physics package contains two ion pumps, the cooling and trapping chamber and the fluorescence detection region, the flight tube, microwave cavities and three layers of magnetic shields. The entire cooling and interaction region is ultrahigh vacuum with an overall vacuum of about 6×10^{-10} Torr.

An elaborate optical set-up is needed to generate precise frequency- and intensity-controlled set of six cooling and two detection beams. An extended cavity diode (Master laser) laser (ECDL) is frequency-locked using saturated absorption spectroscopy. The locked laser output is amplified using a tapered amplifier and then carefully split in six-cooling (four horizontal and two vertical) and two detection beams. The frequency and intensity of all the beams are controlled by acousto-optic modulators (AOM). In addition, home-built fast mechanical shutters are used to eliminate residual scattering of resonant light during the Ramsey interrogation time. Another frequency-locked ECDL (re-pump laser) is used to pump the atoms which fall out of the cyclic cooling transition due to spontaneous emissions. A small quantity of re-pump light is added to one of the

Figure 1. Optical set-up and physics package of the cesium fountain clock developed at CSIR-NPL.

cooling and one detection beams. The eight laser beams are precisely and efficiently coupled to single-mode polarization maintaining fibres (PMFs) which transport the laser beams from the optics table to the cooling and detection chamber in the physics package. At the physics package, thin diverging beams coming out of the fibres are expanded and collimated using indigenously designed beam expanders in order to launch broad beams inside the cooling chamber. The atoms are cooled in the intersection region of the six cooling laser beams. The magnetic coils in anti-Helmholtz configuration are wound up and down on the neck of the cooling chamber to provide position-dependent magnetic trapping force at the centre of the trap. It is possible to cool 10^7 Cs atoms at 6 μK temperature in cloud size of 4 mm diameter. Once the atoms are cooled and trapped, they are launched up in moving molasses by detuning the frequencies of the vertical laser beams. The atoms then interact with microwaves in the cavity twice and then are detected in the detection zone using fluorescence detection.

Apart from the elaborate optical set-up and complicated mechanical design of the physics package, the electronics module is extremely complicated and critical for the complete operation of the fountain. Most of the electronics used for fountain operation has been indigenously developed in the group. The three main components of the module are fountain sequence controller, precision microwave synthesizer and data acquisition system. The entire fountain operation is a sequenced operation with at least ten differently timed subprocesses (e.g. cooling, molasses, launch, post-cooling, state selection, blow away, detection etc.) happening automatically within 1.5 s duration of one fountain cycle. The entire timing sequence is controlled by the sequence controller consisting of autonomous microcontroller and frequency generation cards. The microcontrollers are programmed in such a way that they receive the input variables from front-end user interface program on a PC interfaced with the sequence controller and send the control signals at calculated times to AOMs, shutters etc. The entire sequence controller was designed and assembled in-house.

The microwave synthesizer is a high resolution, precisely tunable (in μHz range) and extremely stable microwave source ideal for such high-precision experiments. The data acquisition and processing system handles high-speed data transfer using a data acquisition card (DAQ) and real-time processing of the data on a LabVIEW platform. Raw fluorescence signals from the detection region are acquired by a high-speed DAQ and processed in the LabVIEW program to estimate transition probability. The LabVIEW program operates in two modes: (I) Ramsey fringe scan and (II) frequency locking and analysis.

2.2 *Results*

At present, the fountain is fully operational and we have got the preliminary results, viz. Ramsey fringes, C-field mapping, frequency locking, stability analysis and estimation of systematic shits. Major work has been done to optimize all the operational parameters and to improve the signal-to-noise ratio in the detected signals. We are now able to observe Ramsey fringes with more than 90% contrast as shown in figure 2.

The 5 MHz crystal oscillator is locked to the central Ramsey fringe using square wave frequency modulation. This 5 MHz locked to the atomic transition is compared against

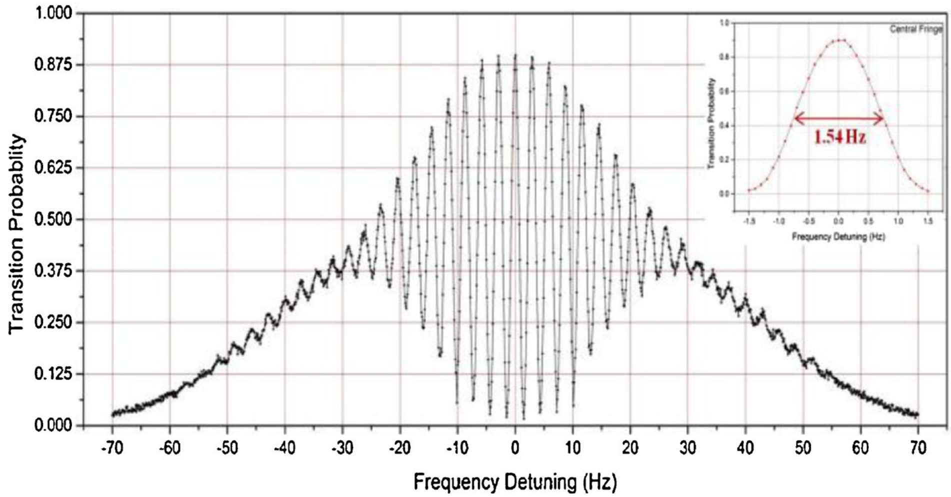

Figure 2. Ramsey fringes with more than 90% contrast corresponding to a toss height of 65.4 cm and Ramsey time of 325 ms. Inset: Enlarged central fringe.

5 MHz from H-maser. The Allan deviation of the frequency difference between the fountain and the H-maser is shown in figure 3. The noise averages down as 6.32×10^{-13} $\tau^{-1/2}$ and hits few parts in 10^{15} at one day averaging.

The fountain is currently at the stage of complete frequency evaluation. It is possible to run the fountain continuously for days without major interruptions provided the

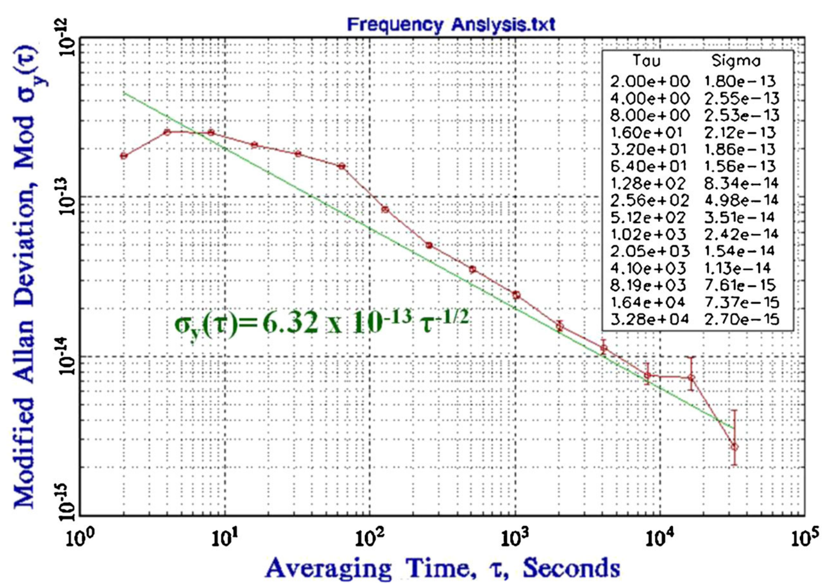

Figure 3. Allan deviation of the frequency difference between the fountain and the H-maser as a function of the averaging time.

temperature of the room is stable. Initial estimation of various systematic shifts to the fountain frequency has been done. A complete frequency evaluation and comparison with fountains in other laboratories such at PTB, Germany is being planned.

Apart from the evaluation of the first Cs fountain, a second generation Cs fountain with special design features to have improved stability and accuracy is being developed at NPLI.

3. Summary

A brief review on atomic clocks along with their current status of research has been presented in this paper. At NPLI, India's first primary frequency standard namely, a Cs fountain clock, has been developed and is operational. Microwave interrogation of the atomic cloud in order to observe Ramsey fringes has been performed and frequency stability of the fountain with respect to a hydrogen-maser has been estimated. We are in the process of optimizing the parameters and planning to do complete evaluation of the fountain frequency for systematic and statistical shifts in the next few months. Once approved by BIPM, India will enter the elite club of only 6–7 countries in the world to have a primary Cs fountain frequency standard. NPLI is also developing a second Cs fountain with better accuracy and an optical frequency standard based on single trapped ytterbium ion.

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