Historical overview of Ramsey spectroscopy and its relevance on Time and Frequency Metrology

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Abstract. A brief overview of the historical evolution of the method of successive oscillatory fields developed by Norman Ramsey, and some different implementations of the decurrent methodology are presented. We use time and frequency standards, from Cs atomic beams to optical standards, as examples. The scientific progress and the technological implementation achieved through a partnership between USP-SC and INMETRO are shown on the characterization of each time and frequency standard.

1. Introduction
The 1989 Physics’ Nobel Prize was divided between Normal F. Ramsey, Hans G. Dehmelt and Wolfgang Paul. Ramsey was awarded "for the invention of the separated oscillatory fields method and its use in the hydrogen maser and other atomic clocks" [1].

Ramsey started to work on Rabi's group at Columbia University in 1937, right after the invention of molecular-beam magnetic resonance. The first experiments in the group were made with lithium fluoride molecules, hydrogen and deuterium. In fact, very good spectra were obtained from lithium fluoride molecules, but the results from hydrogen and deuterium were not as good. This problem was turned over to Ramsey, whereas other researchers in the group pursued, then, more promising and productive goals [2].

The Rabi's method setup employed a uniform magnetic field provided by a coil in which atoms and molecules pass through. When a “correct” frequency of a RF field is applied to the coil, the nuclear spin of an atom flips. The spectral line width in Rabi's resonance is a function of the time of flight throughout the coil, so the longer the time the sharper the line.

After joining Harvard University in 1947, Ramsey started to design his new molecular-beam magnetic resonance apparatus. He faced the challenge of maximizing the spectral resolution. One idea
was to making the RF coils as long as possible to obtain extremely uniform magnetic field, but it became extremely hard to achieve.

Inspired by the Michelson [3] stellar interferometer, that has almost twice the resolution of a perfect telescope with mirror diameter of the same length of the interferometer, Ramsey discovered that using two coils to apply an oscillatory field to the atoms, the resolution of the spectral lines would be twice the resolution obtained in Rabi's method [2,4,5].

In this article we will present a brief overview of the method of successive oscillatory fields developed by Norman Ramsey, its application on the main atomic clock models (Cesium Atomic Beam, Compact Atomic Clock, Fountain Clock, Hydrogen Maser and Optical Atomic Clock) for primary standards of time and frequency, and its importance on other physics experiments. As illustration, we present the progress of the scientific development and technology implementation on IFSC-USP and INMETRO.

2. The Ramsey Method

Ramsey developed a method, designed originally for measurements of nuclear magnetic moments, which can be applied for both atomic and molecular beams. In this methodology, instead of applying a coherent field over a period of time $T$, as in Rabi's method, the atoms or molecules are exposed to the field twice by a short period of time $\tau$, separated by a period $T$ with no field applied (Figure 1). The atoms are prepared on an initial state and enter the region of interaction on the time $t = 0$ and interact with a microwave radiation with frequency $\nu$ during the time interval $\tau$. In $t = \tau$ the atoms pass throughout a microwave free region and their quantum states evolve freely during the time interval $T$. Finally, on $t = \tau + T$, the atoms interact again with the microwave during a new period of time $\tau$. In each region of microwave field, the atoms feel a $\pi/2$ pulse. At the end of this process, with the right condition of frequency and of field power, that maximize the transition probability, the atoms must be at the state $|F = 4\rangle$. Parallel to the atomic beam direction, in the region of interaction, there is a static magnetic field (C-field) that permits to lift the degeneration of the hyperfine steady state. After that, the atoms pass through a detection system that measures the transition probability by an induction signal proportional to the number of atoms in the upper state.

The Full Width at Half Maximum (FWHM) of the interference pattern is $dn = 2T$. So, the longer the time spent by the atoms between the regions of interaction, the narrower the FWHM, improving the sensitivity.

![Figure 1. Schematic representation of Ramsey spectroscopy for Cesium Atomic Beam.](image)

The temporal separation between the oscillatory fields can be achieved by a spatial separation of the two regions of field interaction (e.g. Cesium Atomic Beam and Hydrogen MASER), or by the interaction with the field in the same spatial location, turning on and off the field (e.g. Compact...
Atomic Clock and Optical Atomic Clock), or making the atoms to pass twice the same region (e. g. Fountain Clock).

In this method of successive oscillatory fields the resonance line is about 1.7 times narrower than the Rabi’s methodology for the same length of the apparatus, the sharpness of the resonance is not affected by non-uniformities of the constant field, leading to an increase in precision by a factor of 20, it is more efficient at high frequencies, and the first-order Doppler shifts are eliminated. Notwithstanding those advantages, some precautions must be taken, e.g., the variation on the amplitude of the oscillating field may change the shape of the resonance line.

3. Primary Standards of Time and Frequency

The BIPM establishes the hyperfine transition $|6^2S_{1/2}, F=3, m_F=0>\rightarrow|6^2S_{1/2}, F=4, m_F=0>$ of the non-perturbed Cesium (Cs) fundamental state, at the microwave frequency region (9.192 631 770 GHz), as the definition of the second (Figure 2).

![Figure 2. Cesium Energy Levels for D2 transition at 852 nm.](image)

A primary standard of time and frequency possess the best possible accuracy and stability and it finds applications on local representation of time scale, telecommunications, base stations for satellites, geo positioning, among others, emphasizing the economic impact of the methodology developed by Ramsey, which gave rise to the atomic clock technology.

3.1. Cesium Atomic Beam

Most of the standard atomic clocks in operation around the world are commercial Cs atomic beam clocks employing magnetic selection, notwithstanding the facts that their metrological characterization are not so easy, and that there are some difficulties to correct all uncertainty factors. The assembling of an optically pumped Cs beam clock is a good alternative that allows a complete metrological characterization of the effects that can affect its operation and of the correction factors. Thus, an accurate and stable operation can be achieved. The main differences between the magnetic selected and optically pumped beam clocks are related to the way the atoms of Cs are prepared on the quantum state $F = |3\rangle$ and how the transition probability to $|F = 4\rangle$ is measured.

3.1.1 Optically Pumped Atomic Beam. The atoms of Cs that emerge from a heated oven with velocities of about 200 m/s on the states $|F = 3\rangle$ and $|F = 4\rangle$, are collimated by a nozzle. The atoms are optically prepared to obtain a population inversion, putting the most part on the $|F = 4\rangle$ state. They pass then through spatially separated oscillatory fields (figure 1) and the Ramsey spectroscopy is
performed, obtaining the interference pattern shown in figure 3. Locking the microwave frequency at the maximum of the interference pattern allows operating it as a primary frequency standard (or a local “atomic clock”). The Ramsey fringe shown in figure 3 was obtained with a primary frequency standard developed at the Physics Institute of São Carlos [6] since 1998. This standard was allocated to Inmetro at the end of 2013 [7] to be commissioned as a national primary standard.

Figure 3. Ramsey fringe for cesium atomic beam.

3.2. Compact Atomic Clock
The compact atomic clock is an evolution of the thermal beam clock. It uses cold atoms trapped by laser to interrogate the clock transition, improving its performance.

Figure 4 shows a setup of the compact atomic clock. Three-axis laser beams and a magnetic field trap the atoms (magneto-optical trap) and cool them down at the center of the setup. The atoms are prepared on the state $|F = 3\rangle$ in the same way described at Cs beam section. In sequence, the magnetic field is turned off and the atoms are exposed to the microwave oscillatory field for a time period $\tau$, the microwave oscillatory field is turned off for a time period $T$ and the atoms are exposed again to the microwave oscillatory field to a new period of time $\tau$ as described on section 2.

Figure 4. Compact atomic clock setup.
The main advantage of this setup is its compact size compared to the size of the thermal beam clock, allowing its use at base stations, satellites and for high performance signal dissemination.

Figure 5 presents the Ramsey interferometry pattern, provided by a compact clock, measured at São Carlos laboratory [8]. It is possible to observe an improvement on the spectral resolution when it is compared to that of the Cs beam.

![Ramsey fringe obtained with a compact atomic clock](image)

**Figure 5.** Ramsey fringe obtained with a compact atomic clock

3.3. Fountain Atomic Clock

The fountain atomic clock also uses cold atoms to measure the hyperfine (clock) transition [9]. The process of trapping the atoms is similar to that of the method described for the compact clock, the difference being that it follows a ballistic trajectory concept introduced by Zacharias [5].

After preparing the atoms on the state |F = 3⟩, the cold atoms are launched up vertically with a few meters per second velocity and pass through a region of oscillatory microwave field, for the first time, in a time period \( \tau \). The gravitational force slows the atoms down until they fall back and, after a time period \( T \), the atoms pass again at the same region of oscillatory microwave field by another time period \( \tau \) (see figure 6). The fringe shown in the figure was obtained in São Carlos [9].
Figure 6. Fountain atomic clock setup and Ramsey fringe.

It is interesting to note that, due to the different constructive characteristics, the time interval $T$ between the oscillatory microwave fields is bigger at Fountain clock, followed by Compact Clock and then by the Atomic Cs Beam and thus the spectral resolution and accuracy are better at the same order.

4. Other Frequency Standards

4.1. Hydrogen MASER

The microwave amplification by stimulated emission of radiation, whose acronym is MASER, has been built using various atomic or molecular species in order to perform high-resolution microwave spectroscopic investigations. The Hydrogen MASER has achieved the most widespread use, its stability being superior to that of Cesium (Cs) clocks for periods from about 10 s to a day.

The atomic hydrogen MASER uses a transition between two ground state levels $|F = 1, m_F = 0\rangle$ and $|F = 0, m_F = 0\rangle$ of atomic hydrogen, with a frequency separation of 1 420 405 752 Hz (1.42 GHz). The atomic hydrogen (H) is produced in an intense electrical discharge from the molecular hydrogen gas ($\text{H}_2$). The atoms emerge from the source into an evacuated region and enter a state-selecting magnet. The $|F = 1\rangle$ state will be focused onto the small aperture of the storage cell, whereas the $|F = 0\rangle$ state is defocused. When these atoms are exposed to microwave radiation at the hyperfine frequency, more atoms are stimulated to go from the higher-energy state to the lower one. Thus, this device is an amplifier [4,10].

Hydrogen MASER has also been operated with the microwave power confined in two small cavities that function as separated oscillatory field devices (Ramsey’s method). The atoms that are stimulated to emit radiation move randomly into and out of these cavities with oscillatory fields and spend the intermediate time in the large container with no such fields. Due to the large size of the storage box, there are longer flight times and less frequent wall collisions, so the resonances are narrower and the wall shifts are smaller than for a normal hydrogen MASER (with just microwave cavity).

4.2. Optical Atomic Clock

The natural evolution of these atomic clocks points to the most promised measurements in the optical range frequencies (THz), instead of using microwave excited transitions (GHz) range. Even at this newfangled range, Ramsey method can be of further assistance. Single laser pulse techniques are similar to Rabi spectroscopy and multiple pulses techniques can be related to Ramsey spectroscopy. Multi pulse Ramsey spectroscopy offers several ways to significantly manipulate some frequency shifts present in the excitation, interrogation or detection systems. The most promising method is called Hyper Ramsey spectroscopy scheme [11]. It is based on time-separated pulses that can have different durations, frequencies and phases. The pulses manipulate the induced frequency shifts of the spectroscopic signals due to interference effects. The signature Ramsey fringes are present in Hyper Ramsey fringes under some conditions, and the capability of reducing frequency shifts is very important for uncertainty characterization of high precision optical clocks. Inmetro and Physics Institute of São Carlos have established a cooperation agreement to develop this class of frequency standards based on Sr (Strontium) atoms [12].

5. Conclusion

The field of time and frequency standards received, and continues to receive, important contributions from the Ramsey method. This method turned to be fundamental to the evolution of the atomic clock apparatuses. Brazil is now in the route to construct an infrastructure to build, operate and maintain primary frequency standards that should be, in a near future, capable of contributing to the TAI of the BIPM. It is interesting to note that, due to the different constructive characteristics, the time $T$ between the oscillatory microwave fields is, at Fountain clocks, bigger than the Compact Clock and the Atomic...
Cs Beam Clock. Mainly due to these characteristics, its spectral resolution and its accuracy are better than these last clocks.

References
[1] “The Nobel Prize in Physics 1989”. Nobelpri ze.org. Nobel Media AB 2014. Web. 24 Jul 2015. <http://www.nobelprize.org/nobel_prizes/physics/laureates/1989/>
[2] Kleppner, D. “Norman Ramsey and his method”. Phys. Today, 66, 1, 25 (2013)
[3] Michelson, A. A., and Pease, F. G., “Measurement of the diameter of Alpha-Orionis by the interferometer”, Astrophys. J. 53, 249–259(1921).
[4] Ramsey, N., “The method of successive oscillatory fields”. Phys. Today 66, 1, 36 (2013)
[5] Ramsey, Norman F., “Molecular Beams”. Oxford University, 1990.
[6] Bebeachibuli, A. “Relógio Atômico a Feixe Efusivo de Césio: Estudo Da Estabilidade e da Acurácia Como Função do Deslocamento Da Frequência Atômica Devido ao Efeito Zeeman de Segunda Ordem, ao Cavity Pulling e ao Rabi Pulling”. M.Sc. dissertation, São Paulo: USP, 2003.
[7] Tarelho, L.V.G.; Garcia, G.A.; Baratto, A.C.; de Souza, R.S.L.; de Martin Junior, J.; Muller, S.T.; Magalhães, D.V.; Bebeachibuli, A.; Bagnato, V.S.; Rovera, G.D.; “Joint Effort to Commissioning a Thermal Cesium Beam with Optical Pumping as Primary Frequency Standard to Brazilian NMI”, Annals of 29th Conference on Precise Electromagnetic Measurements (CPEM), Rio de Janeiro, Brazil, 2014
[8] Muller, S T, “Padrão de frequência compacto”, DSc. Thesis, São Carlos, 2010
[9] Alves, R F, “CHAFARIZ ATÔMICO DE Cs 133”, M.Sc. Dissertation, São Carlos, 2012
[10] Riehle, F., “Frequency standards basics and applications”, Wiley-VCH, Weinheim, 2004
[11] Yudin, V.I.; Taichenachev, A.V.; Oates, C. W.; Barber, Z. W.; Lemke, N. D.; Ludlow, A. D.; Sterr, U.; Lisdat, Ch.; and Riehle, F., “Hyper-Ramsey spectroscopy of optical clock transitions”, Physical Review A 82, 011804(R) (2010).
[12] Magalhães, D V; Müller, S T; Pechoneri, R D; de Martin Junior, J; Bueno, C; Bagnato, V S; Rodriguez, S.A., Courteille, P.W.; Paiva R.R.; Garcia G.A.; Tarelho L.V.G.; Amaral M.M.; Baratto, A.C.; Souza, R.S.L; Atomic Frequency Standards in Brazil -Research and ongoing applications, Annals of XXXIII Encontro de Físicos do Norte e do Nordeste, Natal, Brazil, 2015.