

History of early atomic clocks

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Abstract

This review of the history of early atomic clocks includes early atomic beam magnetic resonance, methods of separated and successive oscillatory fields, microwave absorption, optical pumping and atomic masers.

1. Introduction

Ideas for atomic clocks and frequency standards have a long history. James Clerk Maxwell, in his 1873 *Treatise on Electricity and Magnetism* [1] wrote ‘A more universal unit of time might be found by taking the periodic time of vibration of the particular kind of light whose wave length is the unit of length.’ There was no way of realizing this excellent idea in 1873, since the periodic time of light was much too short to be measured, but beginning in 1937 a series of inventions and developments eventually made atomic frequency standards possible. Time and frequency measured with atomic clocks are now the most accurately measurable of all physical quantities.

2. Atomic beam magnetic resonance (caesium, Cs)

1937–1940. Atomic beam magnetic resonance frequency clocks were the first ones with high precision standards and they are still used for the international definition of the second and for many precision measurements. These clocks grew out of Rabi’s [2–5] molecular beam magnetic resonance method, which in turn was based on Dunoyer’s [4] 1911 pioneering molecular beam studies of neutral molecules at pressures sufficiently low for effects of collisions to be essentially negligible. Otto Stern [4] greatly improved molecular beam techniques and extended their applications by using inhomogeneous magnetic fields to deflect and separate molecules in different magnetic states, as in the famous experiment of Stern and Gerlach [4] that first demonstrated the significance of quantum mechanical space quantization. Rabi, discussing the changes in orientation states of an atom passing by successive static magnetic fields in different directions, wrote a seminal paper [2] entitled *Space quantization in a gyrating magnetic field*. This publication provided the fundamental theory for all later magnetic resonance experiments, but six months elapsed before Rabi [2–5], after a meeting with Gorter [4, 5], invented the molecular beam magnetic resonance method.

In this method, a beam of molecules is deflected by an inhomogeneous magnetic field and refocused by a second

magnetic field. An oscillatory magnetic field is applied in the intermediate region. If the frequency of the oscillator is equal to a Bohr frequency, $\nu_0 = (E_f - E_i)/h$, of the atom (where E_i and E_f correspond to the energy levels of an initial and a final state of the atom under investigation), the atom may make a transition to another state with different magnetic properties and the refocusing will fail, with a consequent reduction in the beam intensity. Therefore, the frequency at which there is the biggest change in beam intensity will be that for which $\nu = \nu_0$. Rabi *et al* [3] observed the first magnetic resonance in 1938, but the resonance frequency under investigation depended primarily on the interaction of a nuclear magnetic moment with an external magnetic field, which was too unstable for a precise atomic clock. The first magnetic resonance observations of internal molecular energies were made by Kellogg, Rabi, Ramsey and Zacharias [4] in 1939, and the first atomic hyperfine structure (HFS) transitions, changing the value of $F = I + J$, were observed in 1940 by Kusch, Millman and Rabi [4], including the Cs HFS transition that is now often used for atomic clocks.

1940–1945. The Columbia Molecular Beam Laboratory was closed during World War II. During this period, Rabi was asked to give the Richtmyer Lecture to the American Association of Physics Teachers and the American Physical Society, where he spoke about molecular beam resonance experiments, the possibility of atomic clocks and even the possibility of using them to test the gravitational red shift. William Lawrence wrote this lecture up in an article for the *New York Times* of 21 January 1945, making this article the first publication on atomic clocks in modern times.

Although an atomic clock could have been made in 1945, it would not have been better than the best pendulum or crystal standards of that time. Since L , the length of the oscillatory field region, must be less than $(l/2)\lambda$ for phase coherency, and the halfwidth, $\Delta\nu$, for the magnetic resonance [4] is $1.07\alpha/L$, the fractional halfwidth, $\Delta\nu/\nu$, of the resonance must satisfy

$$\frac{\Delta\nu}{\nu} > \frac{1.07(\alpha/\lambda)}{\nu} = \frac{1.07(2\alpha\nu/c)}{\nu} = \frac{2.14\alpha}{c}, \quad (1)$$

where α is the most probable velocity of that molecule in a volume of the gas and c is the velocity of light. (Note that the fractional resolution is independent of v .) For $\alpha = 200 \text{ m s}^{-1}$, $\Delta v/v > 2.14 \times 200/(3 \times 10^8) = 1.43 \times 10^{-6}$. Even if the resonance frequency of a molecular beam resonance apparatus had been located to within 1% of the resonance width, it would have been good to only 1.4 parts in 10^8 , which was no better than the best pendulum or crystal clocks of that time and so there was no immediate push to build an atomic clock before 1949.

1949–1950. The last scientific barrier to accurate atomic time standards was overcome in 1949 by Ramsey [6], who invented the methods of separated and successive oscillatory fields. In the separated oscillatory fields method, the oscillatory fields are coherently applied to the atom only as it enters and leaves the transition region. As discussed above, the need for coherency requires only that each separated oscillatory field region be shorter than half a wavelength, while the sharpness of the resonance is determined by the separation of the two separated oscillatory field regions, which can be many wavelengths apart. As a result, much narrower resonances are observed and much higher frequencies can be used. In 1950, Kolsky, Phipps, Ramsey and Silsbee [4] successfully used this method in molecular beam spectroscopy.

1952–1954. Sherwin, Lyons, McCracken and Kusch [4] started building an atomic Cs clock at NBS, but in 1953 this effort was unfortunately abandoned.

In 1954, Zacharias [4, 5] attempted to obtain an even narrower separated oscillatory field resonance in a ‘fountain’ experiment. Here, very slow atoms were allowed to travel upwards through an oscillatory field and then made to stop and fall under gravity, to pass again through the same oscillatory field about a second later. The resulting resonance should have been very narrow. Unfortunately, the experiment failed due to the number of very slow atoms being much smaller than the number expected due to their staying a long time in the non-equilibrium slit region, where they could be scattered to a higher velocity. Thirty-five years later, this experiment was successfully revived by Chu *et al* [7] with laser cooled atoms.

1955–1956. Zacharias, Holloway, McCoubrey and others at MIT and the National Company developed in 1955 [5] the first commercial atomic clock called the ‘Atomichron’, which used atomic Cs and the separated oscillatory fields resonance method. This clock required many ingenious engineering developments to obtain high reliability and a much longer operating life (years instead of hours). A number of Atomichrons were purchased by the US Army Signal Corps as well as by commercial organizations and used for research and frequency standards until they were replaced later by newer and better atomic clocks.

Beginning in 1955, Ramsey published a series of theoretical papers analysing possible errors in atomic clocks and the means by which these errors could be avoided or reduced. Also in 1955, Essen and Parry at the National Physical Laboratory, UK constructed and operated the first atomic clock that contributed to a national time and frequency standard. It was based on Cs and the separated oscillatory field method. They also, for the first time, measured the number

of Cs HFS oscillations in a second, which was at that time defined from astronomical observations. These measurements were later repeated by many observers until the International Committee on Weights and Measures in 1964 adopted the definition of the second as equal to 9 192 631 770 periods of the radiation corresponding to the transition between two specified levels of the ground state of a particular isotope of caesium, caesium-133. This definition disagrees with Essen and Parry’s first measurement by only seven parts in 100 million.

From 1956 onwards, many improvements in the Cs atomic clock were developed at universities, national laboratories and industries around the world. These improvements included better excitation, longer beams, reversed beams, reduced size and weight, improved cavities, optical pumping and laser state selection. The organizations contributing to these developments included NIST (formerly NBS), PTB, NRC, NPL, Bureau de l’Heure, the National Company, Bomac, Varian, Hewlett-Packard and Frequency & Time Systems. The contributing scientists and engineers included Mockler, Beehler, Barnes, Helwig, Wineland, Itano, Drullinger, Kartaschoff, De Marchi and others [8]. These and other developments will be described in subsequent papers in this issue.

3. Early microwave absorption and optical pumping

1934. In a great pioneering experiment in 1934, Cleeton and Williams [5] observed a single broad microwave absorption resonance in NH_3 corresponding to the tunnelling or inversion transition between states with the nitrogen atom being on one side or the other of the plane containing the three hydrogen atoms. Unfortunately, there was no follow-up for this first microwave absorption spectroscopy during the next 11 years.

1946–1947. After this 11 year gap and the development of microwave radar during World War II, there was a veritable explosion of microwave spectroscopy publications in 1946, with papers [5] by Bleaney, Penrose, Beringer, Townes, Dicke, Lamb, Becker, Autler, Strandberg, Dailey, Khyl, VanVleck, Wilson, Dakion, Good, Cole, Hershberger, Lambert, Watson, Roberts, Beers, Hill, Merrit and Walter. There were 79 published papers in 1947 and many more in subsequent years.

1948. In 1948, Lyons and others [5] at the NBS made a microwave clock based on the NH_3 inversion transition, but it was soon abandoned because of its instability.

1949. In 1949, Kastler and Brossel [5] developed the powerful technique of optical pumping, in which the occupancy of certain states could be enhanced by inducing transitions with light. Much stronger magnetic resonance and microwave absorption signals could then be obtained. Unfortunately, the optical pumping radiation also induces a light shift of the resonance frequency, a serious disadvantage for a frequency standard, but means were eventually developed by Cohen-Tannoudji and others to reduce or control these light shifts.

Since 1953. Dicke, Kastler and others [5] introduced the use of inert buffer gases such as He to reduce the first-order Doppler shifts by reducing the atom's average velocity and to reduce the numbers of wall collisions. They also used cells coated with various waxy substances to diminish the frequency shifts caused by wall collisions.

1956. In 1956, Dehmelt [5] increased the sensitivity of optically pumped cells by the process of 'shelving', in which a lower frequency clock transition is detected by changed optical fluorescence, depending on whether one of the clock states is occupied.

1958. In 1958, Bender, Dehmelt, Robinson, Dicke and others [5] further increased the sensitivity and accuracy of optically pumped Rb clocks by using hyperfine filtering to reduce the effects of unwanted spectral lines.

Since 1960. Many engineering improvements have been made in optically pumped Rb clocks, and some optically pumped Rb frequency standards were qualified by NASA for use in space missions. Some of the institutions participating in this work were Efratom, ITT, Varian, GTC, FRK, STL, General Radio, FRK, EG&G and Frequency & Time Systems, and participating scientists and engineers included Ardit, Carver, Andres, Bell, Bloom, Stralemeyer, Jechert and Riley.

Optically pumped Rb cells are now highly stable, with an Allan variance $\sigma_y(\tau)$ of 5×10^{-14} for periods of 10^5 s and 5×10^{-13} for 1000 s periods. Such optically pumped Rb clocks are used for many purposes because of their high stability, lower cost and reliability. They are, for example, extensively used in the GPS satellites.

4. Atomic masers (hydrogen and rubidium, Rb)

1954. In 1954, Gordon Zeiger and Townes [5] made the world's first maser, which used electric dipole moments and the tunnelling or inversion transition of NH_3 . At that time it was thought that a maser could not be made using the atomic HFS because of the much smaller dipole moments and transition energies.

1958. In 1958, Kleppner, Ramsey and Fjelstadt [5], in a pioneering experiment on trapped atoms, obtained sharp resonances with Cs atoms trapped in an evacuated bottle long enough for the atoms to undergo several hundred wall collisions during the resonance period. They then realized that atomic hydrogen could probably be stored longer, but the sensitivity of hydrogen detection at that time was very poor, and so they calculated the possibility of the hydrogen resonance being detected by its effect on the radiation field. Due to the narrowness of the resonance and the long storage time, they found that the detection should be easy and that the device could even be operated as an atomic maser.

1960. In 1960, Goldenberg, Kleppner and Ramsey [9] constructed and successfully operated the first atomic

hydrogen maser. Hydrogen atoms in the higher $F = 1$ hyperfine state are focused by the six-pole magnet to continuously load atoms in the high energy HFS state into a Teflon-lined quartz bottle in a tuned cavity. The system spontaneously oscillates as a maser and emits a highly stable signal that can be used as a clock or frequency standard.

1963–1967. In 1963, Bender [5,9] pointed out that spin exchange collisions might reduce the accuracy of the hydrogen maser. But Crampton [5], in a series of papers, showed that if the maser were tuned to make the frequency independent of the line resonance width, the spin exchange shift would be exactly balanced by the cavity pulling effect. Since 1967, many variations and improvements have been made in hydrogen masers by scientists throughout the world, including Vessot, Kleppner, Ramsey, Crampton, Peters, Vanier, Levine, Cutler, Berg, Reinhardt, Audoin, Grivet, Vanier, Kartaschoff, Brenner and Zitzewitz, as will be discussed in other papers in this issue.

The hydrogen maser has the advantage of being a free running atomic oscillator with high stability, narrow bandwidth, low noise and relatively high output power. Its Allan variance, $\sigma_y(\tau)$, is 5×10^{-16} for periods of 1000 s. These characteristics make hydrogen masers particularly useful in radio-astronomy with very long baseline interferometers. The high power and stability of hydrogen masers have also made them particularly useful as flywheel oscillators for much lower powered devices, such as laser cooled atomic frequency standards. Many national standards laboratories use the hydrogen maser for broadcasting time signals even though the maser frequency may be steered by other standards. The hydrogen maser has also been used in various tests of the special and general theories of relativity including the gravitational red shift.

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