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RESULTS IN OPERATION, RESEARCH, AND DEVELOPMENT OF ATOMIC  
CLOCKS AT THE NATIONAL BUREAU OF STANDARDS

ERGEBNISSE BEIM BETRIEB, DER FORSCHUNG UND ENTWICKLUNG VON  
ATOMUHREN IM NATIONAL BUREAU OF STANDARDS

RÉSULTATS CHEZ LE PROJET, LE RECHERCHE ET LE DEVELOPPEMENT  
DES ÉTALONS ATOMIQUES DANS LA NATIONAL BUREAU OF STANDARDS

Summary:

Since the last CIC, significant progress has been made in various laboratories leading to new capabilities and new future uses of atomic clocks. This paper summarizes the contributions of the National Bureau of Standards to these developments and tries to forecast future developments:

Two primary cesium standards, NBS-4 and NBS-5, are now in operational use with an accuracy of near  $1 \times 10^{-13}$ , and with a stability of better than  $1 \times 10^{-14}$  (sample time of 3 hours). The concept and feasibility of a passive hydrogen storage device has been demonstrated leading to the projection of long term stabilities (hours to weeks) of at least  $1 \times 10^{-14}$  for these devices. Studies of different electronic systems as well as wall and magnetic field effects promises significant improvements in the accuracy of hydrogen standards. Methane stabilized helium-neon laser were operated and demonstrated excellent frequency stabilities, and research on a methane beam system is in progress. Novel noise studies of quartz crystal resonators confirmed that quartz crystal oscillators with short-term stabilities of the order of  $10^{-13}$  (1 second) are possible, an important fact in its own right and of significance to the development of atomic clocks of extreme short-term stability.

## Zusammenfassung:

Seit dem letzten CIC sind bemerkenswerte Fortschritte in mehreren Laboratorien erzielt worden, die neue Meßmöglichkeiten und neuartige zukünftige Anwendungen von Atomuhren erlauben. Dieser Bericht gibt einen Überblick über die Beiträge des National Bureau of Standards zu dieser Entwicklung und versucht zukünftige Entwicklungen abzuschätzen:

Gegenwärtig sind zwei primäre Cäsiumnormale in Betrieb mit einer Genauigkeit von annähernd  $1 \times 10^{-13}$  und mit einer Stabilität von besser als  $1 \times 10^{-14}$  (Meßzeit: 3 Stunden). Das Konzept und die Realisierbarkeit einer passiven Wasserstoff-Speicher-Apparatur ist demonstriert worden, welches Grund zu der Erwartung gibt, daß solche Geräte Langzeitstabilitäten von wenigstens  $1 \times 10^{-14}$  aufweisen werden. Studien verschiedener elektronischer Systeme, sowie Untersuchungen von Einflüssen der Wände und des Magnetfeldes versprechen bedeutende Verbesserungen in der Genauigkeit von Wasserstoffnormalen. Methan-stabilisierte Helium-Neon-Laser wurden mit hervorragender Frequenzstabilität betrieben, und Untersuchungen einer Methanstrahl-Apparatur werden gegenwärtig durchgeführt. Neuartige Untersuchungen des Rauschens von Quarzkristall-Resonatoren bestätigten, daß Quarzkristalloszillatoren mit Kurzzeitstabilitäten von einer Größenordnung  $10^{-13}$  (1 Sekunde) möglich sind; dies ist eine bedeutende Möglichkeit an sich und darüberhinaus wichtig für die Entwicklung von Atomuhren mit extremer Kurzzeitstabilität.

## Résumé:

Depuis le dernier CIC, d'importants progrès ont été faits dans divers laboratoires, conduisant à nouvelles possibilités et de nouveaux futurs usages des étalons atomiques. Cet imprimé résume les contributions faites par le National Bureau of Standards à ces développements et essaye de prédire les développements à venir.

Deux étalons principaux de césium, NBS-4 et NBS-5 sont maintenant en opération avec une précision de près de  $1 \cdot 10^{-13}$ , et avec une stabilité de plus de  $1 \cdot 10^{-14}$  (échantillon de temps de 3 heures). Le concept et la possibilité d'un système passif à stockage d'hydrogène ont été démontrés pour arriver à la projection de stabilités à long terme (allant d'heures à semaines) d'au moins  $1 \cdot 10^{-14}$  pour ces systèmes. Des études de différents systèmes électroniques autant que des effets de parois et de champ magnétique promettent des améliorations importantes dans le domaine de la précision des étalons d'hydrogène. Des lasers d'hélium-neon stabilisé au méthane ont été mis en opération et ont montré des stabilités de fréquences excellentes; la recherche d'un système à jet de méthane se poursuit. De nouvelles études de résonateurs de cristal à quartz ont confirmé que des oscillateurs de cristal à quartz avec des stabilités à court termes de l'ordre de  $10^{-13}$  (une seconde) sont possible; un fait important par lui-même et par sa signification dans le développement des étalons atomiques d'excellente stabilité à court terme.

## Introduction

In the area of frequency and time, the Time and Frequency Division of the National Bureau of Standards (NBS) is charged with the realization of the unit of time, the second. Research and development of atomic frequency and time standards at NBS dates back to the very beginning of this field. In 1948 the first "atomic" clock, an ammonia absorption device, was completed and operated [1]. Research on cesium standards also began at about this time leading to an operating device in 1952 [2]. It was, however, not until 1958, that cesium clocks were used in connection with the NBS time services. A succession of cesium devices served in this capacity; they were designated NBS-I, NBS-II and NBS-III [3, 4].

In 1966 theoretical and experimental work started on our present two operational standards, NBS-4 and NBS-5. NBS-5 was completed in 1972 followed by a first preliminary evaluation in January 1973. NBS-4 was completed in 1973 and was first evaluated in August 1973. Both of these primary standards have served in many calibrations of the NBS Atomic Time Scale as well as in measurements of the frequency of the International Atomic Time Scale (TAI).

Work on promising other devices since the last CIC in 1969 includes hydrogen maser oscillators and passive hydrogen storage devices. The maser oscillator program translated slowly into the present passive hydrogen storage program starting in 1970.

Also in 1970, work was initiated on methane stabilized helium-neon lasers supplementing ongoing research at JILA\*. The work concentrates on a methane molecular beam system.

In 1973 experimental studies of the noise of quartz crystal resonators were started which led to important new results with significant bearing on future atomic clocks and infrared/visible frequency synthesis.

In the following, the results obtained in these four areas will be presented in more detail.

### Cesium Standards:

NBS-4 (fig. 1) has been used on a continuous basis for six calibrations of the NBS atomic time scale and serves currently on a monthly to bi-monthly basis for this purpose. NBS-5 (fig. 2) has served in six calibrations of the NBS atomic time scale and is being modified with the intent to further increase its stability, and to render possible an operational calibration capability on a continuous service basis.

The stabilities of NBS-4 and NBS-5 were measured against each other and against other high performance crystal and cesium oscillators. These measurements show that NBS-4 can be characterized by  $\sigma_y = 1.5 \times 10^{-12} \tau^{-1/2}$  and NBS-5 can be described by  $\sigma_y = 0.85 \times 10^{-12} \tau^{-1/2}$ . In a comparison between NBS-4 and NBS-5, a flicker "floor" of  $9 \times 10^{-15}$  was reached.

\*Joint Institute for Laboratory Astrophysics. The NBS Laboratory Astrophysics Division is part of JILA.

The accuracy evaluation of NBS-4 and NBS-5 included--for each device separately and independently--the check and measurement of the magnetic field (4.8 A/m typical), magnetic field inhomogeneities (<1% peak-to-peak), microwave excitation spectrum (noise and sidebands down better than 50 dB), servo system related shifts, etc. [5]. The most significant accuracy limitation, the cavity phase shift, was checked and measured using three different methods [5, 6, 7, 8, 9]: (a) reversal of the beam direction (used in NBS-5), (b) measurement of the velocity distribution followed by a frequency shift experiment

Figure 1.  
NBS-4.

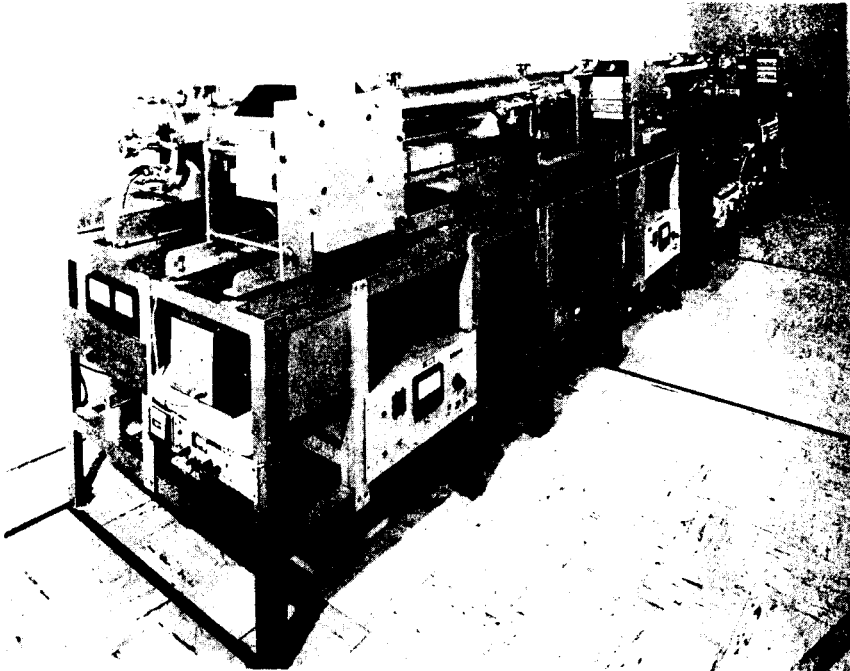
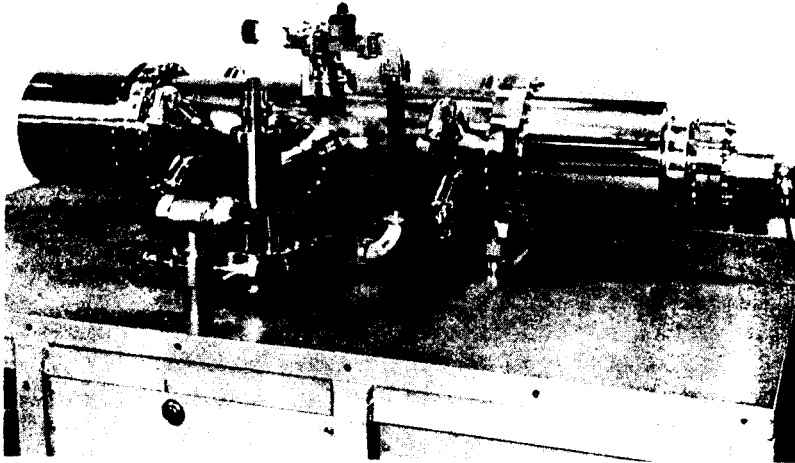


Figure 2.  
NBS-5.

where the power of the microwave excitation was suitably changed (used for NBS-4 and NBS-5), and (c) by a frequency shift experiment using different atomic velocities which were selected by pulsed operation of the microwave excitation (used for NBS-5). The second order Doppler effect correction was obtained for NBS-4 and NBS-5 from the velocity distribution of the atomic beam which was determined from pulsed excitation as well as from an analysis of the Ramsey pattern.

All of the above different methods led to a satisfying agreement within the independently assigned uncertainties. For an individual and independent full accuracy evaluation, one sigma uncertainties of  $2 \times 10^{-13}$  for NBS-5 and  $3 \times 10^{-13}$  for NBS-4 were achieved (fig. 3). By using a series of such evaluations, the memory of each resting with the NBS atomic time scale, an accuracy of close to  $1 \times 10^{-13}$  was achieved. The reproducibility of either NBS-4 or NBS-5 is estimated at better than  $1 \times 10^{-13}$ .

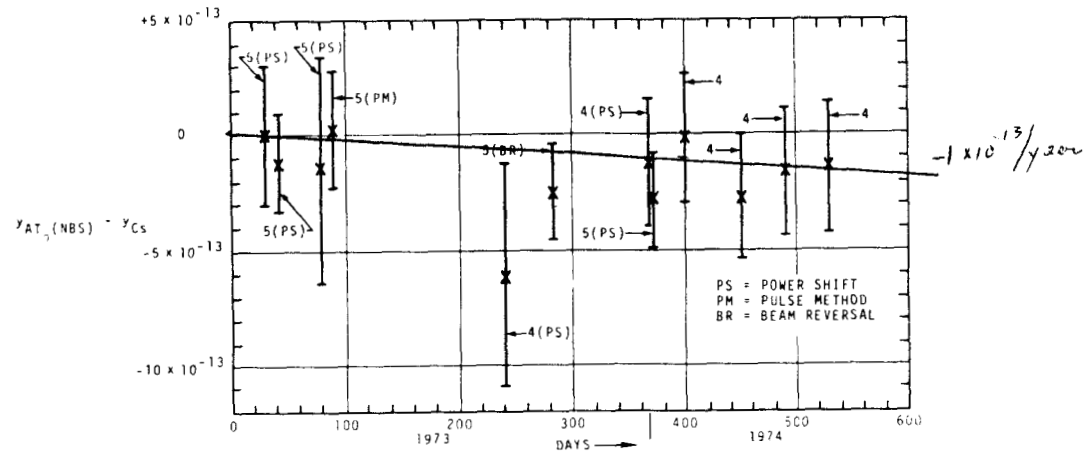


Figure 3. Fractional frequency of the NBS time scale AT (NBS) as measured by NBS-4 (labeled "4") and NBS-5 (labeled "5") during 1973/74. Error bars are 1σ accuracy estimates.

Measurements of TAI:

The total of 13 individual calibrations of the NBS atomic time scale since January 1973 using NBS-4 and NBS-5 has led to corresponding information on TAI. The relationship between TAI and our time scale is known via Loran-C and occasional portable clock comparisons. Between January 1973 and September 1973, a change in TAI of  $\Delta(y_{TAI} - y_{Cs}) \approx -3 \times 10^{-13}$  was noted. Since September 1973, the rate of TAI has not changed; for this time period we assign a value of

$$y_{TAI} - y_{Cs} = +10 \times 10^{-13}.$$

This value includes the gravitational correction for the altitude of Boulder (general relativity) of  $1.8 \times 10^{-13}$ . The one sigma uncertainty is  $1.6 \times 10^{-13}$  which includes the Loran-C measurement uncertainty.

## Hydrogen Standards:

In 1969 and 1970 a series of experiments in cooperation with Harvard University and the Smithsonian Astrophysical Observatory led to a new value for the unperturbed hydrogen hyperfine frequency (clock transition). This value was significantly smaller than most previous measurements [10]. Since then, several laboratories in various countries [11, 12, 13, 14, 15, 16] have re-measured the hydrogen frequency. A summary of the results is shown in Table 1. The weighted mean of all values of Table 1 is 1420405751.7696 Hz, a value which is very close to the original NBS value of 1970.

TABLE 1 INTERNATIONAL AGREEMENT...H MASER OSC (SINCE 1970)  
(MEAS. IN TERMS OF Cs)

	DATE OF PUB	1420405751 HZ PLUS	1 SIGMA
NBS, U.S.	1970	0.769 Hz	$1.7 \times 10^{-12}$
NBS/SAO/HARVARD, U.S.	1970	0.767 Hz	$1.4 \times 10^{-12}$
NRC, CANADA	1971	0.770 Hz	$2.1 \times 10^{-12}$
NASA-GSFC, U.S.*	1972	0.775 Hz	$2.2 \times 10^{-12}$
NPL, U.K.	1973	0.766 Hz	$2.1 \times 10^{-12}$
HARVARD, U.S. ("BIG BOX")	1974	0.768 Hz	$1.4 \times 10^{-12}$
LHA, FRANCE*	1974	0.770 Hz	$2.1 \times 10^{-12}$
RRL, JAPAN	1974	0.773 Hz	$3.5 \times 10^{-12}$

\* WALL SHIFT CORRECTION NOT INDEPENDENT

Hydrogen storage in a passively operating device was proposed [17] and experimentally tried at NBS beginning in 1970 [18, 19]. Figure 4 shows a picture of the apparatus used for these experiments, and figure 5 depicts the storage bulb assembly. In a passive device, the cavity which contains the storage bulb, can have a rather low quality factor. The cavity is driven by an interrogating signal which is derived from a crystal oscillator; the oscillator is locked to the hydrogen resonance by means of a feedback servo. The necessary signal containing the hydrogen resonance information can be acquired in several different ways: (a) detection of the signal which was amplified by the hydrogen resonance (classical maser amplifier); (b) detection of the dispersion of the hydrogen resonance, i.e., of the phase-shift due to the presence of resonating hydrogen atoms; (c) detection of hydrogen atoms after they left the cavity (hydrogen storage beam). We tried method (c) with some encouraging results [18]; however, the need for an efficient, low background hydrogen detector became apparent. Such a detector is not yet available. Thus, the principal advantage of this detection mode, the excellent frequency stability coupled with negligible cavity pulling [18, 20], could not fully be realized. We concentrated on method (b) and actually operated a complete frequency standard system [18, 19]. In dispersion detection no frequency modulation is needed as is shown

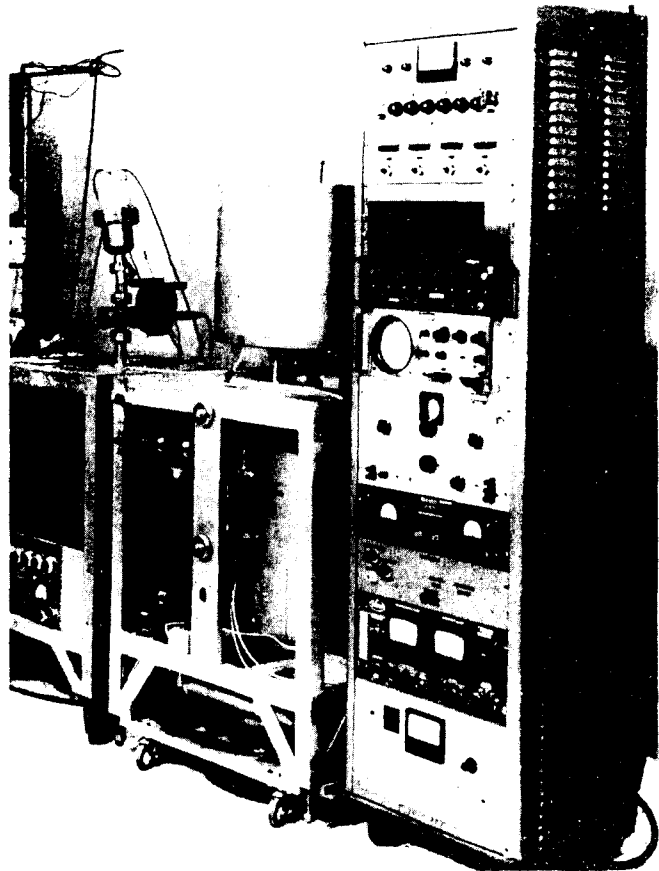


Figure 4. Hydrogen beam systems.

Foreground: H-storage beam standard;  
background: H-beam detection experiment.

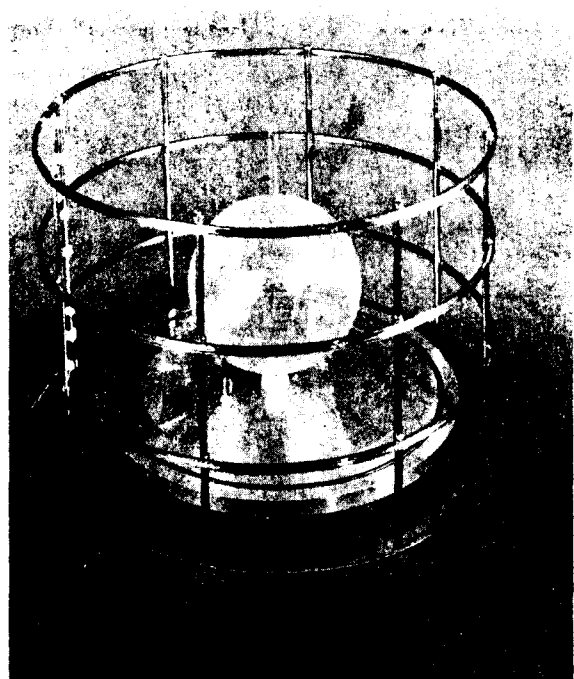


Figure 5.

Hydrogen storage bulb.  
Magnetic field coils and bottom  
parts of cavity and magnetic  
shields are visible.

in figure 6. This simplifies the servo system provided reasonable care is taken to assure adequate phase-stability of the critical electronic circuits. Phase offsets and drifts can be removed to first order by a modulation of the hydrogen resonance: Figure 6 depicts beam intensity modulation as an example. Another method used relies on quenching of the hydrogen resonance by injection of the low frequency Zeeman resonance frequency.

The basic advantages of the passive approach are: (a) A much greater freedom to choose and vary the operational parameters as compared to the maser oscillator since no oscillation threshold conditions must be met before a measurement can be performed. This may prove to be of importance in the evaluation of accuracy limitations, such as spin-exchange effects, wall shift, etc. (b) A significant reduction in cavity pulling due to the possibility of operating with very low cavity quality factors. In terms of temperature stabilization, the requirements are no longer dictated by cavity pulling but by the second order Doppler effect with  $1.4 \times 10^{-13}$  per degree kelvin. The potential for achieving excellent long-term stability and/or relaxed engineering constraints with regard to cavity stability is evident.

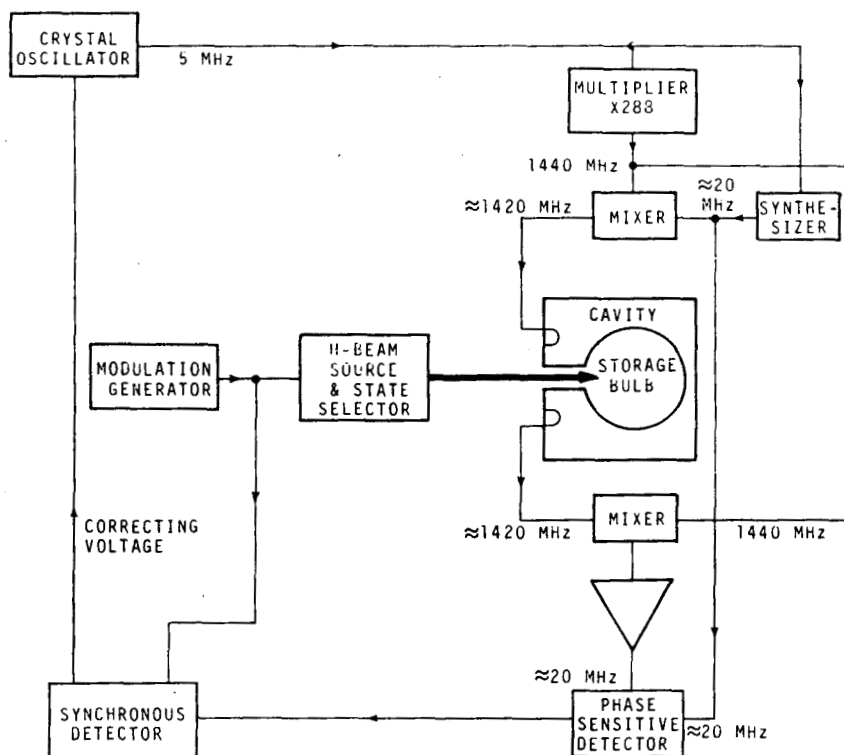


Figure 6. Block diagram of the dispersion lock technique.

### Methane Standards:

Pioneering work [21] in a different division of NBS stimulated us to initiate a supplementing study. Stability results reaching down to  $1 \times 10^{-13}$  at 10 second averaging time were achieved [22]. Since then, work progressed on a methane beam system with the intent to allow quantitative studies of the photon recoil accuracy limitation [23, 24], of the possibility of an interferometric Ramsey structure featuring very sharp resonances [25], and of various other physical effects relating to the stability and accuracy of methane stabilized helium-neon lasers.

Figure 7 shows our first methane beam apparatus which permitted detection of the saturated absorption in a methane beam. In figure 8 the improved beam system is shown. It features a high pumping capacity (2000 l/s) at the methane source, an ability to cool the source to liquid nitrogen temperatures, a beam with a cross section of 1.5 mm by 80 mm, and a 1 m long interrogation path for probing laser beams.



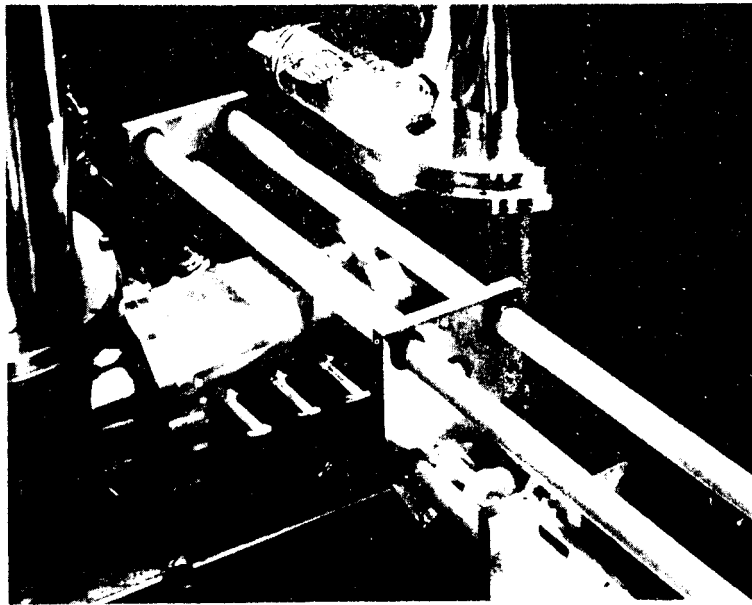


Figure 7. Original methane beam/helium-neon laser system. The beam travels inside of the vacuum enclosure at right angles to the laser light.

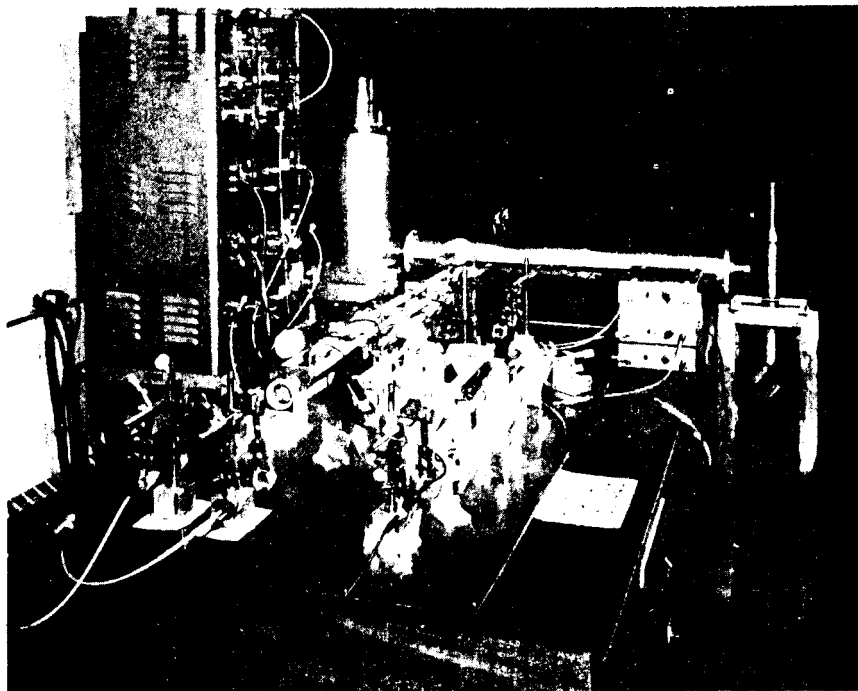


Figure 8. Present, improved methane beam system.

## Quartz Crystals:

Over the past several years, indirect evidence pointed to the possibility that quartz crystal resonators might not be the limiting component in the short-term stability of crystal oscillators [26, 27]. Following suggestions originally made by D. Halford, we studied experimentally the noise behavior of quartz crystals in a passive circuit as shown in figure 9. Two very similar crystal resonators form two branches of a phase comparison bridge. A variable inductance and resistance in one of the branches assure complete matching. The phase noise of the driving source is cancelled to high order. The measured phase fluctuations can be interpreted as originating from frequency fluctuations of the two crystal resonators. We thus obtain directly the spectral density of frequency fluctuations of the crystals and can convert this into time domain frequency stability which can also be measured directly (see fig. 9). More detail can be found in [28].

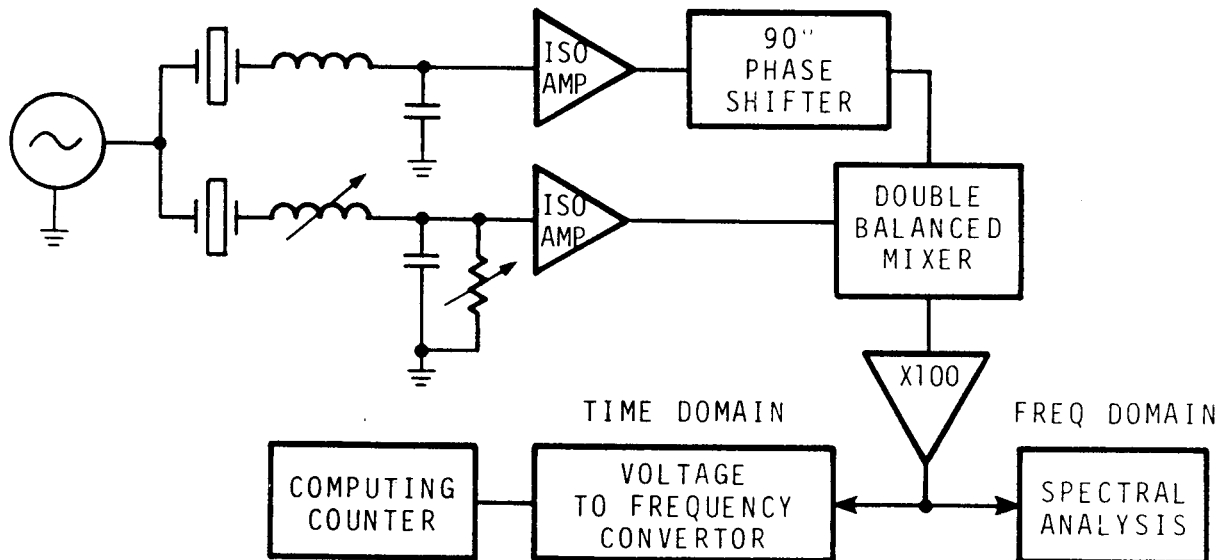


Figure 9. Block diagram of the passive crystal resonator measurement system.

Figure 10 depicts the frequency stability of very good 5 MHz quartz crystals ( $Q \approx 2 \times 10^6$ ) as measured in the passive circuit. The points are direct time domain measurements; the dashed curve is calculated from the frequency noise measurement. The performance of a crystal oscillator using these very same crystals is also shown. It can be seen that the frequency stability of the passive crystals is much superior to the stability of the crystal oscillator in the millisecond region and slightly better for averaging times larger than 1 s. It must be noted that the oscillator of figure 10 is an excellent one by the present state-of-the-art of crystal oscillators. Very recently, improved oscillator stability in the millisecond region was reported [29] supporting our conclusion that crystal oscillators can still be improved significantly in short-term stability.

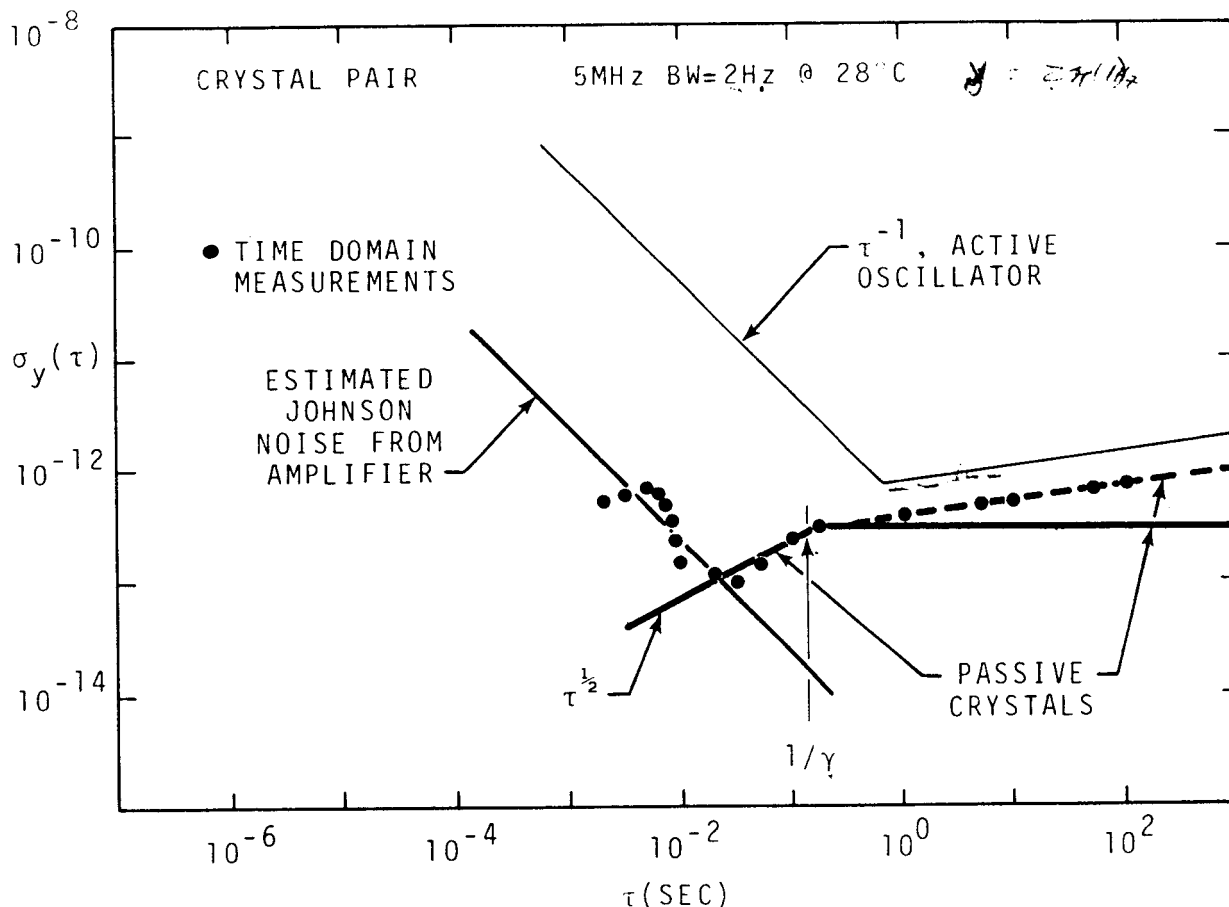


Figure 10. Time domain stabilities of 5 MHz crystal oscillators and passive crystal resonators.

Conclusions:

The present accuracy of cesium standards is close to  $1 \times 10^{-13}$ . Our studies indicate that accuracies of better than  $1 \times 10^{-13}$  appear possible by use of a combination of different techniques, as well as by some design refinements. Shot-noise-limited short-term stabilities of close to  $1 \times 10^{-13}$  also appear possible. This would tax the ability of present crystal oscillators; however, our studies indicate that crystal oscillators with adequate short-term stability should be realizable.

In the not too distant future, both active and passive hydrogen devices should rival cesium standards in accuracy as well as long-term stability (weeks, years). Stabilized lasers are very stable devices in short term and their long-term stability and accuracy looks promising and is under study. The methane stabilized helium-neon laser looks particularly attractive.

The development of high precision frequency synthesis into the infrared is an essential aspect if stabilized lasers are to be considered as true time and frequency standards. Although realized in principle [30], a good deal of refinement remains to be done. One such refinement is the development of frequency sources of high spectral purity in the RF region in order to yield narrow lines after synthesis into the infrared. Our studies indicate that quartz crystal oscillators with the needed spectral purity could be developed.

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