SPECTRAL LINES AS FREQUENCY STANDARDS

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Introduction

As part of its program for the development of precision in primary standards in the microwave region of the spectrum, the National Bureau of Standards (NBS) has been engaged in a broad program investigating the possibilities of atomic frequency and time standards.[1,2] By atomic is meant any atomic system in the general sense. The ultimate goal of the program, considered most optimistically, would be the introduction of atomic standards for frequency and time in the physical sciences if this is feasible. This can also be considered as part of a broader program for the introduction of atomic standards for all of the units of physical science.[3]

Although the problems involved in the design of accurate and reliable frequency and time standards are somewhat different, these two topics will be dealt with together since a clock can be used as a frequency standard while a frequency standard by itself will not read time. A survey of the status of some of the NBS projects relating to the frequency and time program will be given.

Astronomical Time

The present standard of time is the mean solar day. It is of interest to review the accuracy of this unit in order to judge the requirements to be placed on a new unit if it is to be useful as a primary standard. The earth is continually slowing down because of the forces of tidal friction, mostly in shallow seas.[4] In addition, fluctuations in the period of rotation take place from time to time for unknown reasons. This subject has recently been reviewed by Dirk Brouwer[5] who reinterprets the problem as treated by de Sitter[4] and gives more accurate data and results, including work of Spencer Jones and G. M. Clemence. Brouwer concludes that the length of the day is increasing at the rate of 0.00164 second per century with fluctuations in the rate of rotation of the earth occurring at irregular intervals. Principal discontinuities in the derivative of the earth's rotational velocity occurred in the years 1856, 1871, 1877, 1887(?), 1890, 1902, 1911, 1925, and 1936. In 1870, the earth's rate of rotation was fast by about 1.6 seconds per year, compared with the average rotation at that epoch, or one part in 20 million. In 1900, it was slow by 0.6 second per year, or one part in 50 million. Brouwer states:[5] "The deviations from the average rate during the past two hundred years are known to astronomers and can be allowed for in obtaining frequency standards from astronomical time. The record of the earth's rotation during the last century shows that the rate can be predicted for one year ahead with an uncertainty rarely exceeding 0.3 second, or one part in a hundred million." The corrections to the rate of the rotation are obtained by astronomers by using the moon

and planets essentially as precision clocks. The motions of the moon and planets are observed to deviate in the same manner and the same time from that expected according to theory. These deviations indicate that the earth's rotation is not constant during the interval of observation of these motions and furnish a means for measuring the changes in the earth.[44-49]

The day also has a seasonal periodicity as shown in FIGURE 1, taken from N. Stoyko.[6] These measurements have been verified by Finch and others.[7] Munk and Miller[8] review the theories advanced to explain these variations and
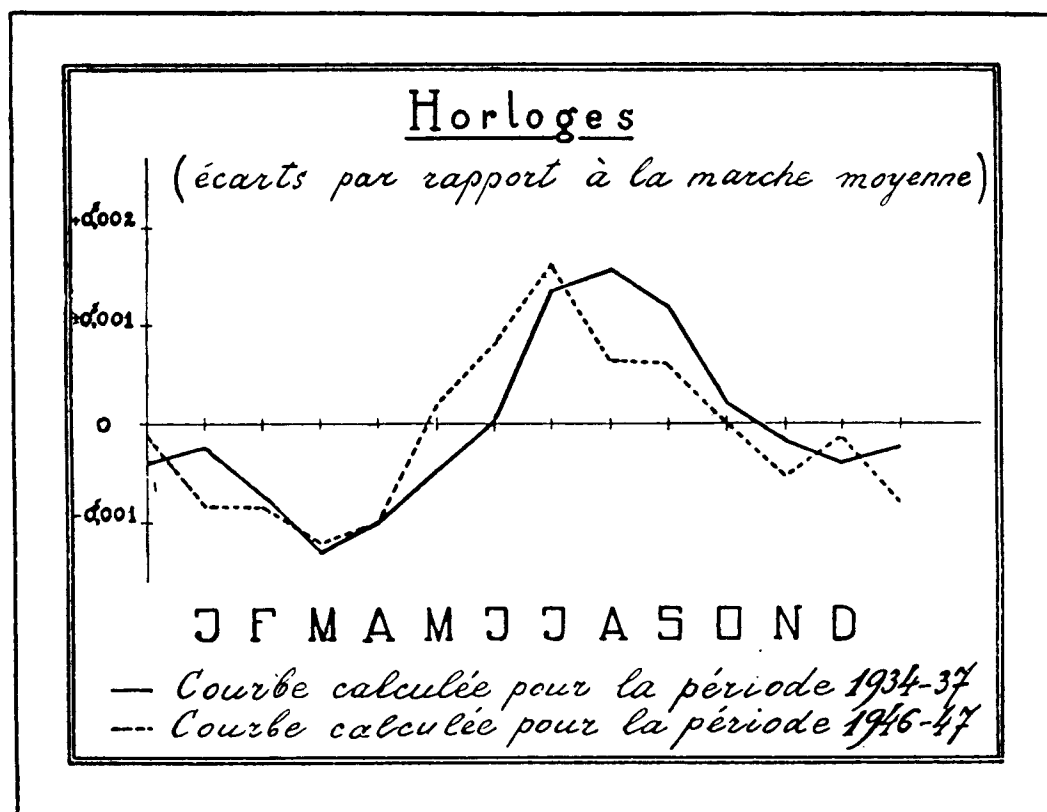

FIGURE 1. The seasonal fluctuation in the length of the day after N. Stoyko.[6]

themselves calculate the effect of seasonal fluctuations in world-wide systems of atmospheric circulation which interact with the earth, the total angular momentum of earth and air being essentially constant. They obtain agreement with the observations both as to magnitude and phase. The maximum change in length of the day is about 1 part in 60 million.[44-49]

It is clear from the above data that a new standard of frequency and time should be capable of a precision of the order of 1 part in 100 million or better. In order to obtain greater accuracy for long-range time problems, astronomers have already adopted a resolution to use the mean sidereal year rather than the day as the unit of time. This resolution was taken at the international conference on the fundamental constants of astronomy held in Paris in 1950. Brouwer[5] also states that modern practice permits annual corrections to the

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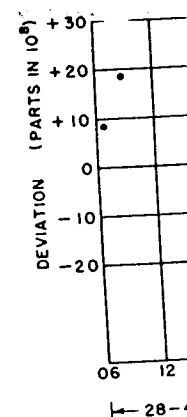be determine
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time based on the rotation of the earth to 0.1 second so that the length of a century can be obtained to one part in 20 billion. In considering such a long time interval, it is worth noting that the fractional accuracy of an atomic clock limited only by random errors will also increase with the length of time interval considered.

The long time taken to make astronomical measurements and corrections raises serious problems in some of the most precise applications of astronomical time to frequency standards. Instrumental errors for one night's observations in the determination of star transits and the effect of atmospheric refraction set limits of about 2 to 3 milliseconds in the accuracy with which the day can be determined, unless averages are taken over intervals of many days. These procedures make time observations inaccessible at frequent and arbitrary intervals.
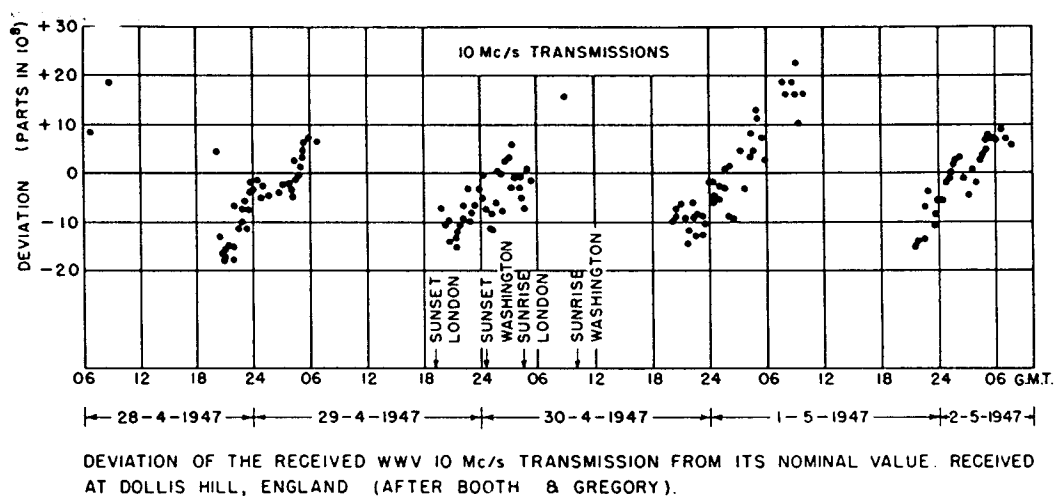

DEVIATION OF THE RECEIVED WWV 10 Mc/s TRANSMISSION FROM ITS NOMINAL VALUE. RECEIVED AT DOLLIS HILL, ENGLAND (AFTER BOOTH & GREGORY).

FIGURE 2. Deviation of WWV transmission frequency as received in England from measurements of Booth and Gregory.[9]

Another factor in the use of the present frequency and time standard which limits their accessibility is the existence of radio transmission errors. The standard frequency broadcasts, which provide world-wide coverage for frequency and time services, rely on ionospheric propagation for long distance transmissions. The motion of the ionosphere produces Doppler shifts in the frequency of the received signals. This varies cyclically as the diurnal variation of ionosphere changes takes place. FIGURE 2, after Booth and Gregory[9] is typical. This figure shows the frequency of Station WWV as received in England. Deviations of 1 part in 4 million are recorded and greater deviations may occur during ionospheric storms. These data also indicate that an invariant frequency and time standard, constantly accessible anywhere on the earth (or off of it) would be valuable.

Microwave Spectrum Lines

The limitations of macroscopic frequency standards immediately suggests the use of periodic phenomena in the microscopic domain to avoid the effects

of environmental changes on frequency constancy. The use of the period of vibration of a spectrum line as a time standard is analogous to the use of the wavelength as a length standard. To make an "atomic ruler," it was necessary to develop an interferometer, as Michelson did, to count the number of times the standard was contained in an unknown length being measured. Similarly, it is necessary to count the number of times the period of a time standard goes into an unknown time being measured. An atomic system by itself thus gives a time standard, but is not a clock until the counting equipment can be added. This greatly complicates the problems associated with accuracy and long running time. It is clear that spectrum lines at frequencies low enough to count the oscillations are a prerequisite to making a clock. In addition, sharp lines of as great an intensity as possible are necessary if great accuracy is to be achieved. If the intensity is such that measurements can be made only to one line width, because the signal-to-noise ratio is near unity, an accuracy of $2\Delta f/f_0 = 1/Q$, where $2\Delta f$ is the line width, f_0 is the transition frequency, and Q is the usual quality factor, is all that will be possible. To measure to 0.001 of a line width or better, as is commonly done for quartz-crystal resonators, requires high signal-to-noise ratios. In searching for spectrum lines which offer promise as frequency and time standards, all of the above factors must be kept in mind. In TABLE 1, various spectrum lines are compared on the basis of Q alone and also on the basis of Q with conventional resonant systems. Many other types of lines are ruled out because of low Q, dependence on external fields, or other factors. In TABLE 1, some of the data are compiled from sources given in later references, as follows: oxygen frequencies and line widths, references 16 and 17; thallium frequencies, from P. Kusch; cesium, reference 42; quadrupole data from reference 23; some ammonia data is taken from reference 14, particularly Bleaney and Penrose and Carter and Smith; and the ammonia 3,3 frequency is an average from NBS measurements of L. J. Rueger and others.

In order to compare the measurement accuracies attainable with different spectrum lines, many pertinent factors such as power jitter, standing waves, long-term stability of components and others must be considered for each technique being compared. This can only be done precisely by experimental methods. Certain basic limitations to accuracy can be calculated, however. Townes[10] has given the theory of many of these in a recent paper, and in particular has considered the ultimate accuracies as limited by noise alone. This accuracy for the 3,3 line of ammonia, in an equipment having a noise bandwidth, B, of one cycle per second, comes out to be about 1 part in 2×10^{14} when considering the noise power, $2kTB$, due to thermal radiation transmitted down the waveguide absorption cell and received in the detecting system. It is of interest to carry out such calculations for various spectrum lines and techniques in order to determine pertinent design parameters. The noise-limited resolving power will be derived for the case of molecular gas absorption as typical of this type of consideration. In addition, the actual noise figure of a spectrometer will be taken into account in order to give an idea of the noise limitations encountered in practice.

There is a minimum detectable frequency shift away from the center of a spectrum line, or other fiducial point, which can be measured as limited by noise. If this minimum shift is δf, then $f_0/\delta f$ is defined as the noise-limited resolving power, R.P. The R.P. will vary according to whether the center of the line is taken as reference point or some other point, such as that at maximum slope of the absorption curve. For these points we define $(R.P.)_0$ and $(R.P.)_{max.}$, respectively. $(R.P.)_{max.}$ will obviously be much higher than $(R.P.)_0$. Because of the finite absorption of the empty waveguide, the maximum possible R.P. will not be the same as $(R.P.)_{max.}$, where the slope of the absorption curve is a maximum. For ammonia, this difference will not be great and will be neglected in the interests of simplicity. The point of maximum slope of the absorption curve will occur at a frequency where $\alpha_f = \frac{3}{4}\alpha_0$. α_0 is the maximum coefficient of the gas at resonance and α_f the coefficient at the frequency in question.

As the frequency of a signal sent through an absorption cell is varied and this range of variation is decreased, the change in output signal will decrease until it is equal to the noise level, which then sets a basic limit on maximum accuracy. This change in output power level in a waveguide cell for a frequency change δf must be calculated in order to find this limit.

Let
$$\alpha = \alpha_c + \alpha_f \tag{1}$$

$$\alpha' = \alpha_c + \alpha_{f'} \tag{2}$$

where the initial and shifted frequencies are f and f', respectively. The total power absorption coefficients are α and α', while α_c, α_f and $\alpha_{f'}$ are for the wall losses in the waveguide, and the gas at frequencies f and f', respectively. These gas coefficients are the waveguide values, not the plane wave values.

The change in power at the detector when changing frequency from f to f' will be

$$\Delta P = P_i e^{-\alpha' L} - P_i e^{-\alpha L} \tag{3}$$

where P_i is the input power and length of the cell is L.

By using equations 1 and 2 and defining

$$\Delta \alpha_f = \alpha_f - \alpha_{f'} \tag{4}$$

where $\Delta \alpha_f \cdot L < 1$, there results

$$\Delta P = P_i e^{-(\alpha_c + \alpha_f) L} \cdot (\Delta \alpha_f \cdot L) \tag{5}$$

From the theory of collision broadening,[11] the change in α_f for a change in frequency from f to f' can be calculated. For waveguide, the α for waves travelling in an unbounded space filled with the absorbing gas should be multiplied by a factor λ_g/λ_1, where λ_g is the guide wavelength and λ_1 the intrinsic wavelength in the gas. Since this factor is varying slowly with frequency as compared to the effect of the absorption line, it will be treated as constant and included in the definition of α_0.

TABLE 1

Q and Bandwidth of Frequency and Time Standards*

Macroscopic standards

Description of Standard	Total width of resonance curve at half-intensity	$Q = f_0/2\Delta f$	Remarks
RLC series circuit	$2\Delta f = R/2\pi L$	10^2-10^3	Radio circuits, R = Resistance, L = Inductance
Tuning fork	Depends on design	$\sim 10^4$	Good fork in vacuum
Pendulum	Depends on design	10^4-10^5	Good gravity pendulum
Cavity resonator	Depends on mode, shape, and conductivity of cavity	10^4-10^5	Ordinary microwave cavities
Piezoelectric resonator	$2\Delta f = R/2\pi L$	$10^4-6 \times 10^6$	Depends on equivalent R and L of particular mode and crystal cut
Rotating or revolving earth giving the day or the year	These are nonresonant systems	—	—

Spectroscopic standards

Cause of line broadening	Total width of resonance curve at half-intensity	$Q = f_0/2\Delta f$	Remarks
Atomic beams			
Natural line width; zero-point vibrations, thermally induced transitions	Negligible line width	—	Governed by formulas for spontaneous and induced emission and absorption
Collisions; Doppler; saturation	Negligible	—	Excitation at right angles to beam avoids Doppler effect; unexcited atoms continuously supplied by oven
Finite absorption time in transition region of length L	$2\Delta f \cong \dfrac{1.3}{L}\left[\dfrac{2kT}{m}\right]^{1/2}$	$\sim 2.7 \times 10^7$ $\sim 4.5 \times 10^7$	Cesium $- f_0 = 9192.632$ Mc. Thallium$^{205} - f_0 = 21{,}368$ Mc. Thallium$^{203} - f_0 = 21{,}168$ Mc. Ramsey method of excitation for path length of 50 cm.
	Multiply by 0.64 for Ramsey method of excitation		
Nuclear electric quadrupoles in crystals			
Interactions between the nuclear quadrupole and surrounding electrons and nuclei in the crystal lattice	$2\Delta f = 1/\pi T_2$ $\dfrac{1}{T_2} \cong \dfrac{1}{T_2'} + \dfrac{1}{2T_1}$	$\sim 2 \times 10^1$ $\sim 1.3 \times 10^4$ $\sim 8.3 \times 10^3$	T_1 = relaxation time which limits lifetime of the states. T_2' is a measure of the broadening caused by magnetic dipole interactions. No simple, generally valid, microscopic formulas can be given. $\left.\begin{array}{l}\text{SnI}_4\ 204.5\ \text{Mc.}\\ \text{ICN}\ 382.4\ \text{Mc.}\\ \text{I}_2\quad 331.3\ \text{Mc.}\end{array}\right\}$ At $293°$K. $\overline{\begin{array}{l}I_2\\ f_1 = 331.3\ \text{Mc.}\\ f_2 = 644.4\ \text{Mc.}\end{array}}$

(Continued)

TABLE 1 (*Continued*)

Spectroscopic standards

Interactions between the nuclear quadrupole and surrounding electrons and nuclei in the crystal lattice | $2\Delta f = 1/\pi T_2$; $T_1 = $ relaxation; T_2 is a measure of the broadening caused by magnetic dipole interactions. No simple, generally valid, microscopic formulas can be given ; $\frac{1}{T_2} \cong \frac{1}{T_2'} + \frac{1}{2T_1}$ | ~8.3 × 10³ | 331.5 Mc. I₂

(Continued)

TABLE 1 (Continued)

Spectroscopic standards

Cause of line broadening	Total width of resonance curve at half-intensity	$Q = f_0/2\Delta f$	Remarks
Molecular absorption in gases			
Natural line breadth due to spontaneous emission induced by zero-point vibrations of the electromagnetic field	$2\Delta f = \frac{32\pi^3}{3hc^3} \cdot f_0^3 \cdot |\mu_{ij}|^2$	~10^{18}	For NH₃ inversion (3,3) line 23.870.127 Mc.
Induced emission and absorption in equilibrium with thermal radiation	$2\Delta f = \frac{64\pi^3}{3h^2c^3} \cdot f_0^2 \cdot |\mu_{ij}|^2 \cdot kT$	~10^{16}	For NH₃ at room temperature (300°K.)
Interruption of inversion by a transition to another rotational level	Sum of line widths due to spontaneous emission and absorption between rotational levels	~10^{10}	For NH₃ (3,3) line (300°K.)
Collisions of molecules with walls of absorption cell	$2\Delta f = \frac{A}{\pi V} |RT/(2\pi M)|^{1/2}$	~8 × 10⁵; ~6 × 10⁶	NH₃ at 300°K.; O₂ at 90°K. } For long K-band waveguide cell
Doppler effect	$2\Delta f = \frac{2f_0}{c} \left[\frac{2RT}{M} \log_e 2 \right]^{1/2}$	3.3 × 10⁵; 8.3 × 10⁵; 1.26 × 10⁶	For NH₃ at 300°K.; For O₂ at 90°K.; For O₂ at 39°K.
Self-broadening due to molecular collisions	$2\Delta f = 1/\pi t = Nv \cdot \sigma/\pi = Nv b_e^2 = (2\Delta f)_1 \cdot p_{mm}$, for low pressures; t = lifetime of the state. No simple, generally valid, microscopic formula can be given	4.4 × 10⁵; 3.5 × 10⁷	*Oxygen* K₊ = 760,436Mc. K₋ = 759,164.2Mc.; K₊ = 961,120Mc. K₋ = 958,324.9Mc.; For NH₃ at 1 micron pressure; For O₂ at 1 micron pressure; NH₃(3,3) → (2Δf)₁ = 54Mc./mm.; O₂(K₋ = 9) → (2Δf)₁ = 1.7Mc./mm.
Saturation	$2\Delta f \cong (2\Delta f)_0 \left[1 + \frac{4\pi\mu^2 \cdot P}{3ch^2 \cdot (\Delta f)_0^2} \right]^{1/2}$	~3.1 × 10⁵; ~2.5 × 10⁷	NH₃:1 micron, P ≅ 8.3 microwatts/cm.²; O₂:1 micron, P ≅ 53 microwatts/cm.²; $\frac{2\Delta f = (2\Delta f)_0 \cdot 2^{1/2}}{\mu(NH_3) \cong 80\mu(O_2)}$

*f₀ = Resonance frequency
T = Absolute temperature
M = Molecular weight
N = No. molecules/unit volume
b_e = Effective diameter for collisions
P = Power per unit cross section

μᵢⱼ = Dipole matrix element
k = Boltzmann's constant
A = Cross-sectional area of absorption cell
v = Molecular velocity
p_{mm} = Pressure in mm. Hg
(2Δf)₀ = Line width for zero input power

h = Planck's constant
R = Gas constant
V = Volume of absorption cell
σ = Effective cross-section for collisions
(2Δf)₁ = Line width at 1 mm. Hg
c = Velocity of light
m = Mass of atom

To a good approximation, then

$$\Delta\alpha_f = \alpha_f - \alpha_{f'} = \alpha_0 \left[\frac{1}{1 + 4Q^2 \left(\frac{f - f_0}{f_0}\right)^2} - \frac{1}{1 + 4Q^2 \left(\frac{f' - f_0}{f_0}\right)^2} \right] \quad (6)$$

The resolving power will be different according to whether the initial, or stabilization, frequency is taken as f_0, where the slope of the resonance curve is zero, or at the point of maximum slope. The latter point, of course, gives greater sensitivity but introduces additional difficulties in procedures and equipment. For these two cases, we first set $f = f_0$ and then

$$\left(\frac{f - f_0}{f_0}\right)^2 = \frac{1}{12Q^2} \quad (7)$$

$$\alpha_{f\,\text{max. slope}} = \tfrac{3}{4}\alpha_0 \quad (8)$$

into equation 6 and finally substitute for $\Delta\alpha_f$ in equation 5. At resonance,

$$\Delta P \cong P_i e^{-(\alpha_c + \alpha_0)L} \cdot L\alpha_0 \cdot 4Q^2 \left(\frac{f' - f_0}{f_0}\right)^2 \quad (9)$$

At maximum slope,

$$\Delta P \cong P_i e^{-(\alpha_c + \frac{3}{4}\alpha_0)L} L\alpha_0 \tfrac{3}{4}\sqrt{3}\, Q \left(\frac{f' - f}{f_0}\right) \quad (10)$$

In these equations, $(f' - f_0)$ and $(f' - f)$ will both be referred to as δf. Putting the minimun value for ΔP which can be detected into each of these equations will give the minimum δf which can be measured. Here again two cases must be differentiated, namely, the $(\Delta P)_{\text{min.}}$ for a linear or superheterodyne detector, and for a square-law or crystal video (at low power) detector. From the review article of W. Gordy,[12] the minimum detectable powers as limited by noise are given as:

(linear case) $(\Delta P)_{\text{min.}} = [16FkTBP_i e^{-(\alpha_c + \alpha_f)L}]^{1/2} \quad (11)$

where F is the overall noise figure of the spectrometer, and B is the noise bandwidth; and

(square-law case) $(\Delta P)_{\text{min.}} = (4kTB)^{1/2}/M \quad (12)$

where M is the figure of merit of the crystal detector.

With these values for $(\Delta P)_{\text{min.}}$ we can solve equations 9 and 10 for δf and minimize the result by finding the optimum value for the cell length, L. On differentiating with respect to L and setting equal to zero, the general results are obtained:

(linear) $L_{\text{opt.}} = 2/(\alpha_c + \alpha_f). \quad (13)$

(square-law) $L_{\text{opt.}} = 1/(\alpha_c + \alpha_f). \quad (14)$

This gives four equations when it is remembered that $\alpha_f = \alpha_0$ and $\tfrac{3}{4}\alpha_0$ for

resonance and maximum slope conditions, respectively. Likewise, equations 9 and 10, combined with 11 and 12, give a total of four separate results.

These results can now be simplified and put into a convenient notation if the optimum cell lengths are put into equations 9 and 10 and new coefficients are introduced.

Using the values of the minimum detectable absorption coefficients as given by Townes[13] and Gordy,[12] we introduce

(linear)
$$\alpha_{g\min.} = 2\alpha_c e\left[\frac{FkTB}{P_i}\right]^{1/2} . \tag{15}$$

(square-law)
$$\alpha_{g\min.} = \frac{\alpha_c e(4kTB)^{1/2}}{P_i M} \tag{16}$$

The reciprocals of the $\alpha_{g\min.}$ may be considered as measures of the signal sensitivities of the systems.

When equations 9 through 16 are combined, the final results can be expressed by two equations, since, in this notation, the linear and square-law cases are the same. Finally, therefore, we have the resolving power, R.P. $= f_0/\delta f$. At resonance, for both linear and square-law detectors,

$$(\text{R.P.})_0 \cong 2Q\left[\frac{\alpha_0\alpha_c}{\alpha_{g\min.}(\alpha_0 + \alpha_c)}\right]^{1/2} \tag{17}$$

At maximum slope, for both linear and square-law detectors,

$$(\text{R.P.})_{\max.} \cong \sqrt{3}\ Q\left[\frac{\alpha_f\alpha_c}{\alpha_{g\min.}(\alpha_f + \alpha_c)}\right] \tag{18}$$

where $\alpha_f = \frac{3}{4}\alpha_0$. Here, the proper values for $\alpha_{g\min.}$ appropriate to the linear and square-law cases must be used as well as the correct values of the four different optimum cell lengths.

Inspection of equations 17 and 18 shows that the resolving power increases with the Q and intensity of the spectrum line, and the signal sensitivity of the system, as expected. The $\alpha_{g\min.}$ are smaller the greater the input power and lower the waveguide wall losses, α_c . The input power cannot be increased indefinitely without considering saturation. In addition, the above analysis will change if the detector changes its law with power input. For example, a crystal detector will only be square-law for low input powers. The best noise figures will give the highest resolving power. For very intense spectrum lines, the resolving power approaches a constant value unless the input power is simultaneously increased.

It is clear that accuracy will be greater for a superheterodyne detector, since $\alpha_{g\min.}$ can be smaller than for a crystal video detector in most methods of operation. In addition, resolving power is much improved by operating at the point of maximum slope of the spectrum line. A system operating in this manner might have trouble defining the center frequency of the line, f_0 , and might also be sensitive to pressure changes of the gas, which change the line

width. Obviously, one can use both points of maximum slope and take the mean frequency. Additional complications in equipment result, however, and great care must be exercised if the benefits to be derived from this method of operation actually are to be achieved in practice. An FM method of measurement to be described later has been developed which uses both points of maximum slope to obtain maximum sensitivity.

Some numerical results will indicate the limitation in accuracy set by noise alone. From the papers of Gordy, Townes, and others, some representative values for the parameters in equations 17 and 18 may be taken. For K-band operation in the one-centimeter wavelength region, and for gases like ammonia, we have roughly, $\alpha_c = 10^{-3}$ cm.$^{-1}$, $\alpha_C = 10^{-3}$ cm.$^{-1}$, and $Q = 10^5$. Optimum cell lengths are given in TABLE 2.

If we take $\alpha_{g\,min.} = 10^{-8}$ cm.$^{-1}$ for the square-law case and $\alpha_{g\,min.} = 1.5 \times 10^{-11}$ cm.$^{-1}$ for the linear case, we get approximate values for the resolving power (TABLE 3).

TABLE 2
OPTIMUM LENGTHS

	Linear detector	Square-law detector
At resonance	10 meters	5 meters
At max. slope	11.4 meters	5.7 meters

TABLE 3
RESOLVING POWERS

	Linear detector	Square-law detector
At resonance	1.2×10^9	4.5×10^7
At max. slope	5.0×10^{12}	7.4×10^9

The value 1.5×10^{-11} cm.$^{-1}$ for $\alpha_{g\,min.}$ in the case of the superheterodyne detector corresponds to a noise figure of 2, an input power of 1 milliwatt and a noise bandwidth of 1 cycle per second. These are idealized figures.

Similarly, low bandwidths would not help the case of an ordinary crystal video detector because the crystal noise goes up inversely with the video frequency. The video frequency will decrease as sweep speed decreases in the attempt to use lower and lower bandwidths. The use of some kind of signal modulation to raise the video frequency would alleviate this difficulty.

The results given above indicate that noise alone will prevent achievement of the accuracy goal of 1 part in 100 million, discussed earlier, if a crystal video detector is used at the resonance point. Although two NBS atomic clocks using ammonia have been built, using crystal video detectors in the interest of simplicity, and resolving powers approaching those listed in the table obtained, enough progress has now been made to warrant the change-over to superheterodyne detection being incorporated in Model 3. Operation at the points of maximum slope has already been incorporated. Even for the case of a superheterodyne detector, it appears in practice that this mode of operation will be necessary.

Townes[10] has given the theory of noise limitations when including the effect of power saturation. In this case, an optimum input power exists, and Q, in equations 17 and 18, should be replaced by

$$Q = Q_0 \left[1 + \frac{4\pi |\mu|^2 P}{3ch^2(\Delta f)_0^2} \right]^{-1/2} \tag{19}$$

where Q_0 and $2(\Delta f)_0$ are the Q and line width, respectively, at zero input power, P is the incident power per unit cross-section,[14] and c and h are the velocity of light and Planck's constant, respectively. Molecules of different orientations saturate at different values of radiation intensity. For this reason, averages over the various magnetic quantum numbers M must be taken. Equation 19 is an approximation[10] and, in addition, $|\mu|^2$ is used to approximate $|\mu_M|^2$, the square of the dipole matrix element $|\mu_z|^2$ for a particular value of M. Similarly, we have

$$\alpha = \frac{\alpha_{00}}{1 + 4Q_0^2 \left(\dfrac{f - f_0}{f_0}\right)^2 + \dfrac{4\pi |\mu|^2 P}{3ch^2(\Delta f)_0^2}} \tag{20}$$

where α_{00} is the absorption coefficient at resonance for zero input power. If the power is increased indefinitely and it is assumed that the detector does not overload, the R.P. will go to zero because α and Q both go to zero. This more than compensates for the improvement in $\alpha_{g\,min.}$ because of the increased power. If the power is made proportional to the square of the gas pressure and therefore proportional to $(\Delta f)_0^2$, the terms in equations 19 and 20 responsible for the saturation will remain constant. It is of interest to consider the results when this condition holds and the power is kept at the approximately optimum value where

$$\frac{4\pi |\mu|^2 P}{3ch^2(\Delta f)_0^2} = 1 \tag{21}$$

If we vary the power and gas pressure together while keeping equation 21 fulfilled, the Q will still vary according to equation 19. However, α will remain constant at resonance or at the fiducial point used for stabilization if we always readjust the fiducial frequency, for example, so that we remain at the point of maximum slope. In this case, equations 17 and 18 show that:

(at resonance) $\qquad\qquad (\text{R.P.})_0 \sim \dfrac{Q}{(\alpha_{g\,min.})^{1/2}} \tag{22}$

(at maximum slope) $\qquad (\text{R.P.})_{\text{max.}} \sim \dfrac{Q}{\alpha_{g\,min.}} \tag{23}$

The dependence of $\alpha_{g\,min.}$ on power is obtained from equations 15 and 16. Combining this with equations 22 and 23 and remembering that Q is proportional to $1/(\Delta f)_0$ with power proportional to $(\Delta f)_0^2$ the result in TABLE 4 is obtained. In the case of a linear detector at maximum slope, this result shows

that the reduction of Q by increasing gas pressure is offset by the increased input power which can then be used.

The above considerations may be applied to the problem of choosing the best gas for a primary frequency and time standard. Two unique gases exist, ammonia and oxygen.[15, 10] In comparing these, it is apparent that the high Q and low saturation of the oxygen lines tend to offset the effect of their lower intensities. From equation 21, it is seen that a power flux of about $(80)^2 = 6400$ times greater than that for ammonia could be used for equal line widths because of the 80 times smaller value of μ. Because μ is small, the collision broadening in oxygen is likewise very small compared to ammonia, the collision Q being of the order of one million at a pressure of 30 microns.[16] For this reason, combined with the low saturation, it should be possible to reach the Doppler width with oxygen. Since oxygen can also be cooled to a very low temperature, we may estimate a Doppler Q of roughly 10^6 as possible. Oxygen will still have a vapor pressure of about 1 micron at a temperature of 39°K. and 1759 microns at its melting point of 54.39°K. The frequency of the oxygen lines will be about 2.5 times that of the ammonia lines and the Q of 10^6 about

TABLE 4

	Linear detector	Square-law detector
$(R.P.)_0$	Proportional to $Q^{1/2}$	Independent of Q
$(R.P.)_{max.}$	Independent of Q	Proportional to Q^{-1}

ten times that normally used with ammonia. This gives a line width roughly four times smaller and an input power of $6400/16 = 400$ times larger than for ammonia. Many of the oxygen lines are fairly intense, the strong $K_+ = 9$ line having an absorption coefficient of about $1/20$ that of the $3,3$ line of ammonia.[17] This factor will be assumed without correcting for differences at low temperatures.

If the above rough figures are put into equations 15 and 18 and if α is taken as small, compared to α_c, the $(R.P.)_{max.}$ for oxygen will be as great as for ammonia, even if the noise figure for the equipment is 100 times larger than that obtainable with ammonia at the lower working frequency. This allows considerable leeway, since experience has shown that the low Q of ammonia is more troublesome than its value of $(R.P.)_{max.}$ because of nonfundamental errors other than those due to noise. A working Q of 10^6 would help greatly in reliably reaching clock accuracies of 1 part in 10^8 or better, as long as increasing Q does not entail too low a value of $(R.P.)_{max.}$.

The above assumes that the necessary power will be available at the oxygen frequency. At low pressures, the power required does not seem too high, particularly when considering modifications of available klystron multiplier tubes. The high Q would also make it easier to use a resonant cavity absorption cell instead of waveguide without as much difficulty from frequency sensitivity as encountered with lower Q's.

Since the oxygen line will be split by the earth's magnetic field, it will be

necessary to shield the absorption cell. Available data on mu-metal indicates that the splitting of the order of a megacycle could be reduced satisfactorily even without employing special field-nulling methods. The broadening due to the splitting would not then prevent high Q. During magnetic storms, the earth's field might vary from 1 to 20 milligausses. The usual variations, even during storms, are very slow and of the order of 1 milligauss. The variations in the residual fields after shielding by factors of perhaps 1000 would then be negligible. Because of its high Q and large Zeeman effect, oxygen suggests itself for use in special applications in a magnetometer to measure weak fields through the Zeeman splitting, rather than preventing the splitting, as in the present application.

Nuclear Electric Quadrupole Absorption

The use of pure nuclear quadrupole spectra for frequency and time standards merits consideration because they have several attractive features. The spectra should bridge the frequency gap between the low frequencies, where quartz crystal oscillators are applicable, and the microwave region. This makes possible the use of ordinary tubes where relatively good noise figures could be obtained and the advantages of amplifiers, modulators, etc., would be available. Crystalline solids would be used as standards instead of gases or beams, making the equipment simpler, smaller and more rugged. The problems of aging and mechanical vibrations or shock encountered with quartz crystals should be obviated. Large samples can be rather conveniently used to help improve intensity.

The temperature dependence of quadrupole lines would require temperature control of the sample. Temperature coefficients for chlorine compounds of about one part in 10,000 per degree have been measured.[18] Livingston's work, however, shows that the frequency for chlorine compounds becomes relatively temperature-insensitive at very low temperatures.[19] In addition, there is some possibility that crystals can be found with lower temperature coefficients. One such possibility would concern crystals with low temperature coefficients of expansion, because available data indicate that the temperature shift in compounds such as iodine is largely due to changes in the asymmetry parameter with temperature changes.[20] Temperature shifts can also be caused by torsional oscillations of the molecules in the crystal lattice.[21] In either case, crystals with rigid lattices would probably give lower temperature shifts of the quadrupole lines.

The noise-limited resolving power, R.P., can be estimated for quadrupole lines from formulas given by Pound for the signal-to-noise ratios.[22] In attempting to do this for iodine at 332 Mc., the amplitude signal-to-noise ratio comes out to be very high for relaxation times, T_1, of from 0.003 to 1 second, and other parameters corresponding to a laboratory set-up at NBS using a large sample in a high-Q resonant cavity. The relaxation time is actually unknown, but the work of Dehmelt and Kruger[18, 23] on halogen compounds indicates that an upper limit would probably be below the one-second limit given above as an extreme case.[24] Even for one second, the ratio obtained seems high in view of

difficulties met with at NBS, and elsewhere, in attempting to detect these lines with a Zeeman-modulated, resonant cavity, absorption cell but without using regenerative detectors. However, we have not had much difficulty detecting iodine with a super-regenerator at room temperatures and with fairly good signal-to-noise ratios. Although the advantages of regenerative detectors are clear, the fact that only such systems have been used in quadrupole work indicates the small value of the absorption coefficients obtainable. More work remains to be done to obtain better information on these problems. In the meantime, experimentally measured quadrupole lines show Q's from 3,000 to 20,000 for iodine compounds.[23] Frequencies have been reported by Livingston for chlorine compounds, in one case giving six significant figures[25] and in another case five figures,[19] but with an absolute accuracy claimed of 1 part in 10,000. These results indicate that the line frequencies could be read to considerably better than 1 part in 10,000. It is difficult to judge at this time as to the ultimate potential frequency precision indicated experimentally with quadrupole lines so far discovered. At higher frequencies and with refined frequency measuring techniques, however, these results may become somewhat more promising.

Atomic Oscillators and Microwave Frequency Dividers[26]

There are many different ways of using absorption lines to make frequency standards and clocks.[1, 2] Some of these methods apply to different kinds of spectroscopic standards such as quadrupole lines, gases and atomic beams. Others do not, as will be self-evident. We will consider first what may be termed an atomic oscillator, that is, an oscillator controlled in frequency directly by the absorption line rather than indirectly thru a servo-mechanism. It would be desirable to design such an oscillator exactly analogous to precision, stabilized, quartz-crystal oscillators. The most commonly used circuit for this, where the highest precision is necessary, is the Meacham, wheatstone bridge oscillator shown in FIGURE 3.[27] Changes in both gain and phase in the feed-back loop of this self-excited oscillator are prevented from shifting the oscillation frequency much if gain is sufficiently high. This stabilization is accomplished by the balancing action of the bridge and the amplitude-limiting action of the tungsten lamp and may also be considered as the result of a form of negative feed-back. The analogous circuit using a gas absorption line for control, a six-arm waveguide wheatstone bridge[28] and thermal unit to replace the lamp is also shown in FIGURE 3 where the position of the thermal unit (barretter or thermistor) depends on the sign of its temperature coefficient of resistivity. A magic tee could also be used. The six-arm bridge allows impedance matching through adjustment of the ratio arms. In this circuit, the bridge becomes a transmission filter, allowing feed-back and oscillation only at the resonance frequency of the gas.

The input impedance of a shorted waveguide filled with absorbing gas is, in the usual notation, following Smith, et al.,[29]

$$Z = Z_0 \tan h[\alpha_0 + j\beta + \gamma/2]L \qquad (24)$$

where

$$\gamma = \gamma_0 \Delta\omega_0 / [\Delta\omega_0 + j(\omega - \omega_0)] \qquad (25)$$

The complex gas attenuation constant, γ, is taken from the theory of collision broadening and has a real part equal to the absorption coefficient already given. The absorption and dispersion are equal at the angular frequencies, ω, where $\omega - \omega_0 = \Delta\omega_0$, and $\Delta\omega_0$ is half the angular line breadth. In using the gas filled cell in an oscillator, both the absorption and dispersion are involved so that this method illustrates one application where dispersion is employed, compared to most methods, which use the absorption only. If the cell is used to terminate one side arm of a magic tee as shown in FIGURE 3, the insertion loss of the tee can be calculated. If the ammonia cell were completely absorbing, giving a maximum unbalance of the tee, a minimum insertion loss
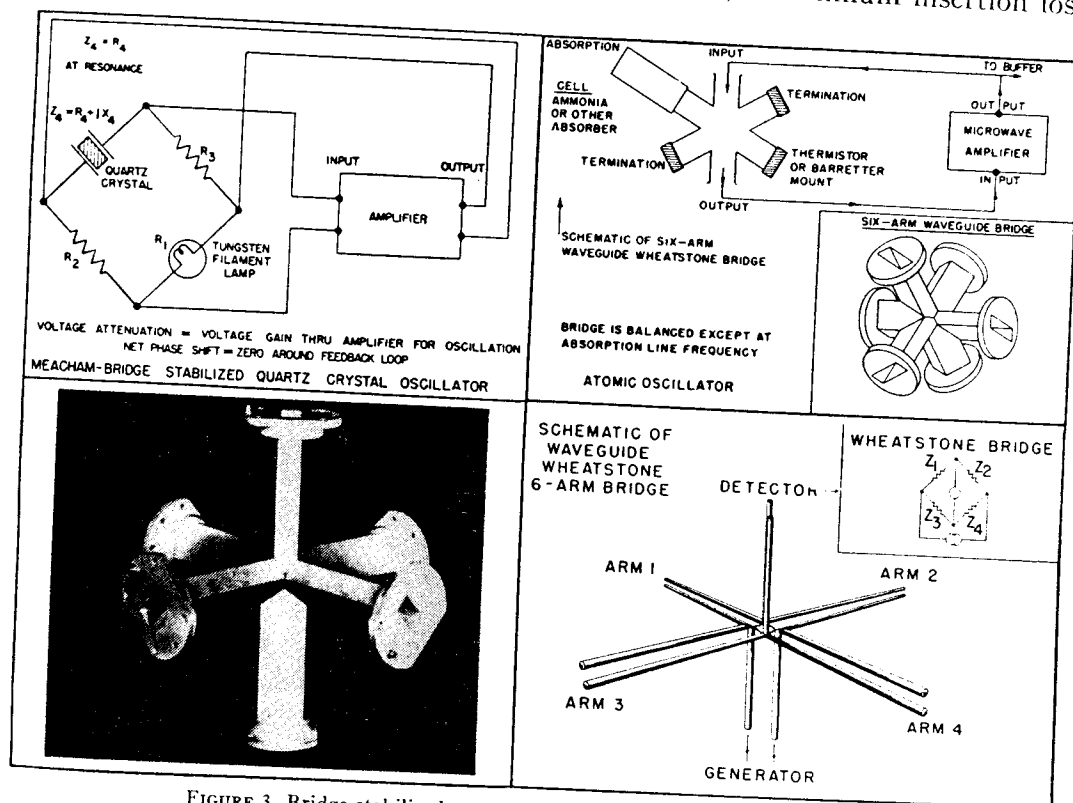
FIGURE 3. Bridge-stabilized quartz oscillator and microwave analogue.

of six db. would result for matched generator and load. This loss would have to be made up by the amplifier in the feed-back loop of the oscillator. For oscillation, the net phase shift around the feed-back loop must be zero and the total loss equal to the gain. In general, the insertion loss would be found from[30]

$$P_4 = P_0 \left| \frac{Y_1 - Y_2}{(1 + Y_1)(1 + Y_2)} \right|^2 \tag{26}$$

where P_4 is the output power from arm 4 of the tee into a matched load, P_0 is the available power from a matched generator on arm 3, and Y_1 and Y_2 are the normalized admittances terminating the side arms 1 and 2. This equation shows, of course, that the tee is balanced with equal side arms, but will transmit if one arm is filled with absorbing gas at the absorption frequency, since

the admittance or impedance will be different for the two arms and will go through resonance, as shown by equations 24 and 25 for the gas-filled arm. Smith et al.[29] have shown that the impedance of the cell looks like a series-resonant cavity, having constants depending on the waveguide, in series with a parallel-resonant cavity depending on the gas only. This indicates, as expected, that the cell can be replaced by a resonant cavity, in an equivalent circuit to test the oscillator, at frequencies where amplifiers are available. This has been done at frequencies in the 3,000 and 6,000 Mc. ranges. In the meantime, a klystron amplifier has been developed by Varian Associates for this application at the 23,870 Mc. frequency of the ammonia 3,3 line. A gain of about 13 db. has been achieved. The tube is shown in FIGURE 4 and is quite small
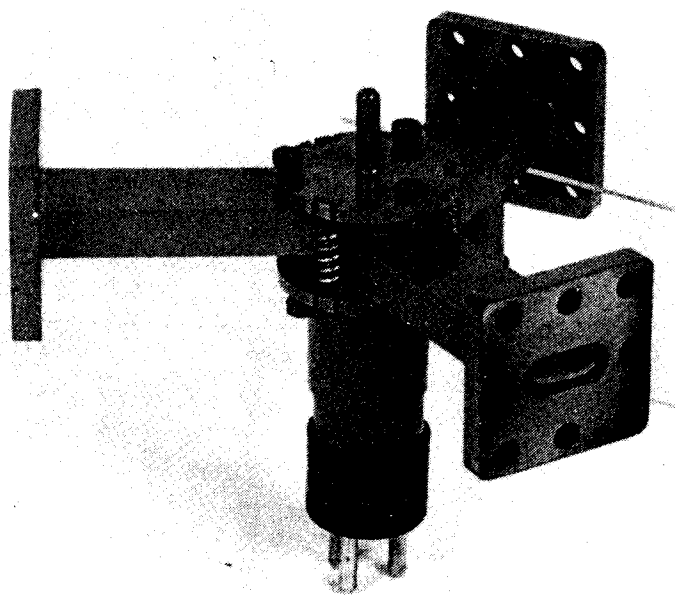
FIGURE 4. Klystron amplifier (Varian Associates X-15) developed for oscillator at 23,870 Mc.

and relatively easy to use. Production of additional tubes has been delayed with the result that further progress will also be delayed. High gain obtained by using several tubes in series will increase the stabilization obtained.

A suggested method for avoiding the need for K-band amplifiers combined with a means for frequency division has been given in a previous publication.[26] It has been shown that microwave frequency division for use in clocks or to provide additional frequencies from an oscillator such as that discussed here can be carried out with a precision of at least one part in 10^{10}. Accordingly, the combination of an atomic oscillator and frequency divider should have an accuracy determined by the oscillator alone. Reference may be made to the above paper[26] concerning further details of the frequency divider circuits. It may be noted here, however, that further details on klystron multiplier tubes which can be modified for use in a divider for 23,870 Mc. are given in the next section.

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Ammonia or Oxygen Control of Quartz Clock

In contra-distinction to the atomic oscillator-frequency divider type of clock, it is possible to make a clock controlled by the spectroscopic standard through a servo-mechanism. Any spectroscopic standard can be used, so the method also applies to the atomic beam technique to be described later. In order to count the oscillations of the standard, the frequency is lowered by one means or another to a suitable value for driving a counter such as a synchronous motor clock. It is preferable that no more than the minimum of one servo-mechanism be included in the equipment in order to obtain maximum accuracy and simplicity. In addition, the oscillator to be controlled should be inherently as stable as possible without control, in order to obtain the maximum flexibility and simplicity in design and maximum continuous frequency accuracy. Frequent servo-correction of an unstable, controlled oscillator requires a wider band servo loop than would be the case for a stable oscillator. The larger noise band-width which this entails reduces the ultimate accuracy attainable for a single correction and thus the maximum continuous frequency accuracy. If one servo is used to control the main oscillator and additional servos are used for controlling lower frequency equipment in order to run the clock, the desirable minimum of one servo will be exceeded.

FIGURE 5 shows the basic design principle of all the types of NBS servo-clocks.[31] Here, only one servo is used and the controlled oscillator is the most stable and reliable type obtainable, namely a quartz crystal oscillator, which in turn drives the motor clock. Two ammonia controlled clocks of this type have been built and operated for developmental purposes.[32] A third unit to be described briefly later is planned for greater accuracy, reliability and running time. It is also planned to investigate the use of oxygen in more detail, since an oxygen absorption cell could be used with the present servo clock methods.

The detailed block diagram of the Model 2 ammonia clock is shown in FIGURE 6. Here it is clear that the frequency gap between the controlled quartz oscillator and the absorption line is bridged by means of a frequency multiplier chain. Irregular frequency variations or frequency jitter in the controlled oscillator, which occur at a faster rate than the frequency of correction by the servo, would obviously result in limitations on accuracy. The use of the quartz crystal oscillator greatly reduces such difficulties, as compared to the control of a klystron oscillator. Irregular frequency variations have been found to occur when reflex klystrons are used to sweep the oscillator frequency slowly over the spectrum line in order to achieve low noise bandwidths. FIGURE 7 was taken by L. J. Rueger in a special set-up at NBS and illustrates the lack of frequency jitter when quartz-crystal drive is used. The figure was obtained by sweeping the multiplier chain over the ammonia 3,3 line in a period of about 15 minutes. The markers are about 26 kc. apart. The periodic zig-zag on the curve is due to the gear-teeth in a gear reducer used to drive a rotating condenser which in turn swept the chain in frequency. Unfortunately, a cleaner recording, illustrating the lack of frequency jitter with a quartz oscillator, is not now available. The results may be observed, nevertheless; a slow variation in output level is barely evident in the interval of fifteen min-
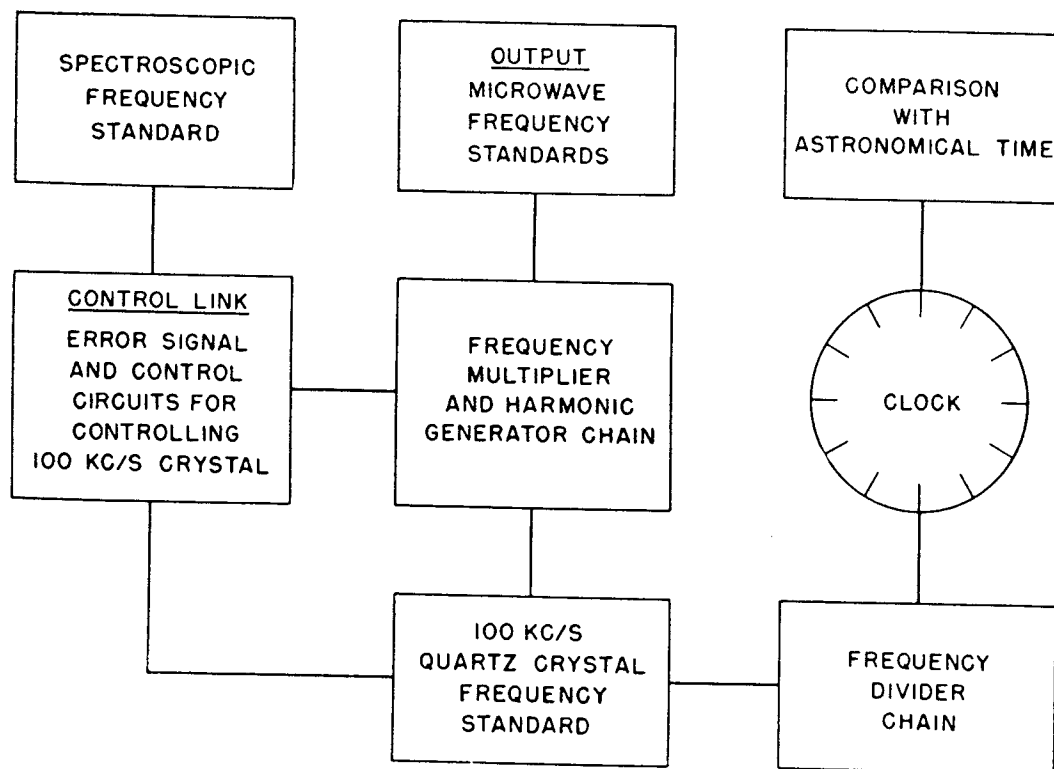
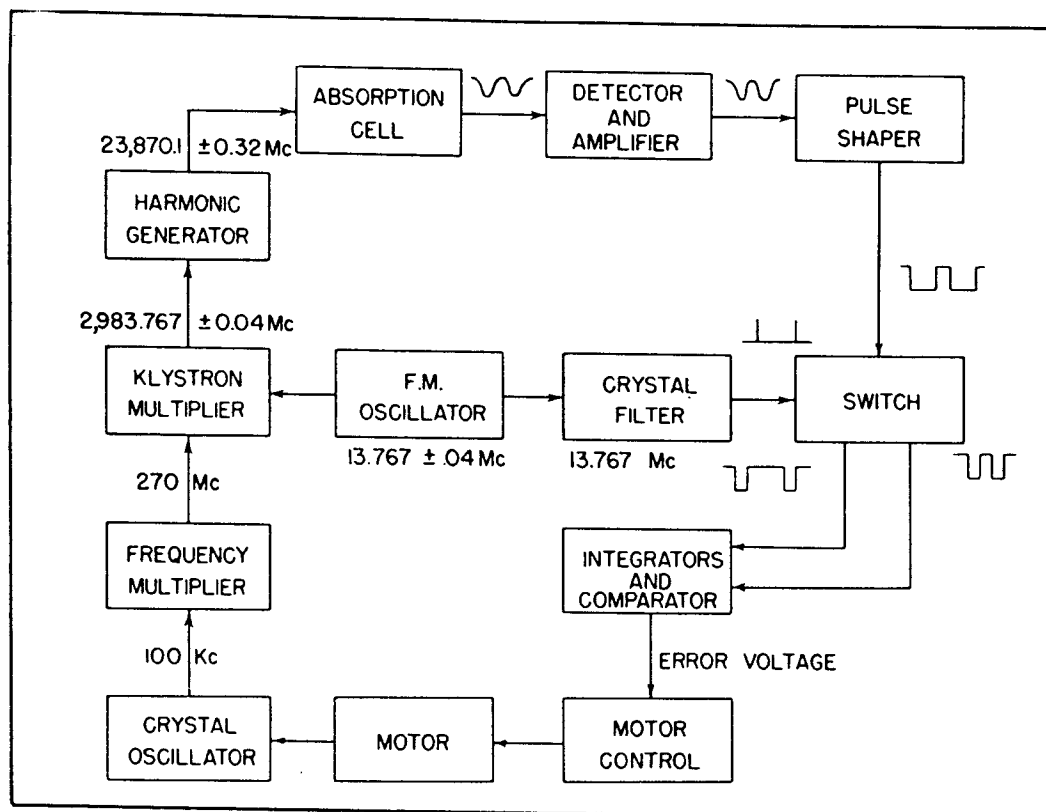FIGURE 5. Basic schematic of NBS spectroscopic servo clocks.

FIGURE 6. Schematic of NBS Model 2 ammonia clock.

utes. Automatic gain or power normalization methods might be necessary if longer intervals were used in this particular set-up, which used most of the Model 2 circuits in making the recording.

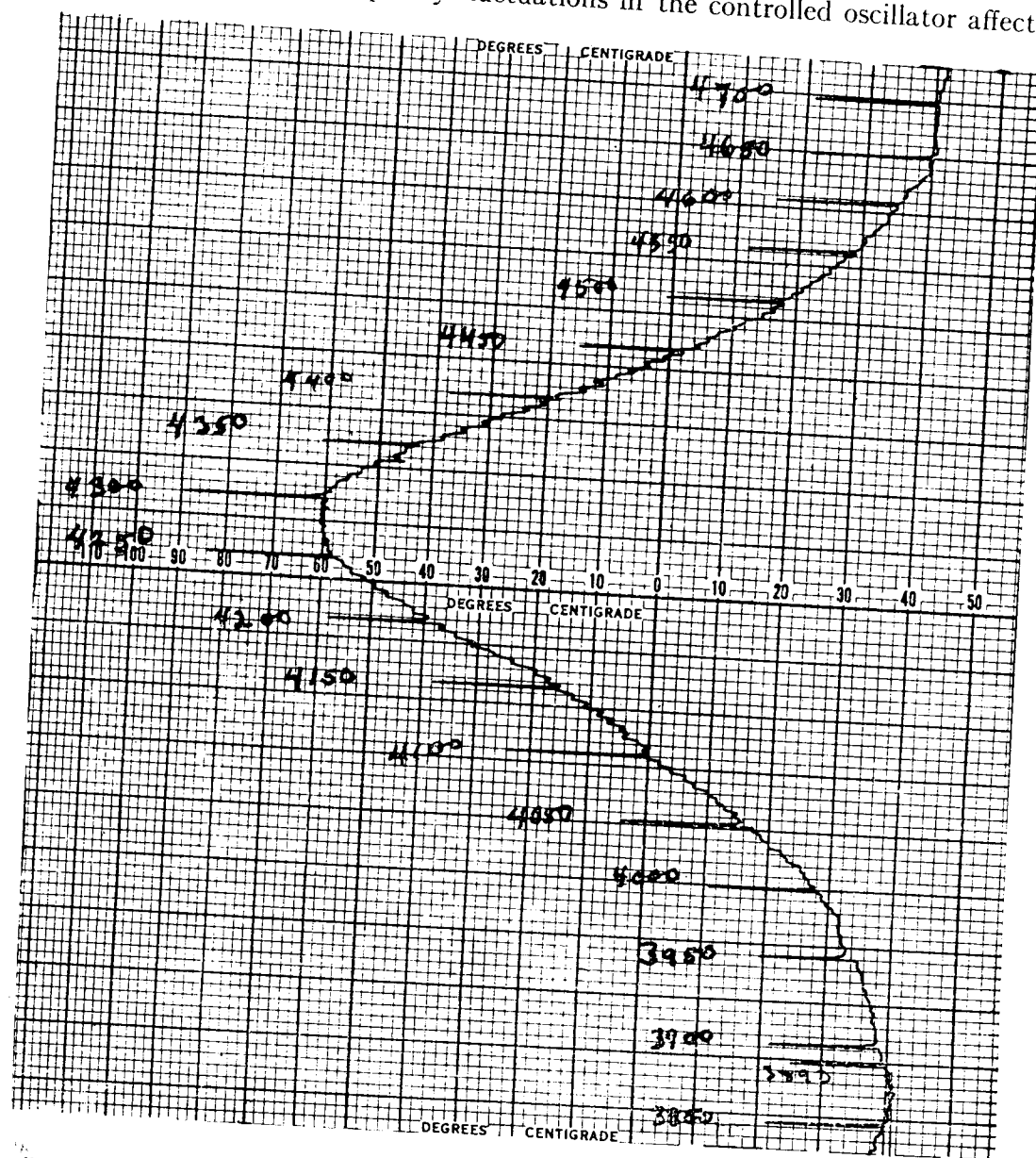The occurrence of frequency fluctuations in the controlled oscillator affects

FIGURE 7. Ammonia 3,3 line recorded with a very slow sweep illustrating stability of quartz crystal drive.

the minimum rate of servo-correction which must be used to obtain a given over-all accuracy. This raises the question as to the magnitude of the short-time fluctuations in a quartz-crystal oscillator. FIGURE 8 (after Booth and Laver) shows the variations in some of the best quartz oscillators maintained at the Post Office Radio Branch in England over both a period of hours and minutes.[33] The fluctuations amount to 10 to 20 parts in 10^{10} over a period of hours and of the order of 1 or 2 parts in 10^{10} from minute to minute. If a

frequency accuracy of one part in 100 million is desired for a servo-controlled atomic clock, infrequent servo corrections will be needed for a precision quartz oscillator. On the other hand, if an accuracy of one part in 10^{10} is desired and otherwise possible, as potentially indicated for atomic beam techniques, the servo must correct the quartz clock at short time intervals even for the best quartz oscillators, and the noise-limited resolving power for such short-time corrections must be great enough to allow attainment of the desired accuracy. The progress recently made in high-Q, low-drift quartz crystals will help reduce this difficulty, but the above data indicate the order of magnitude of the effects.
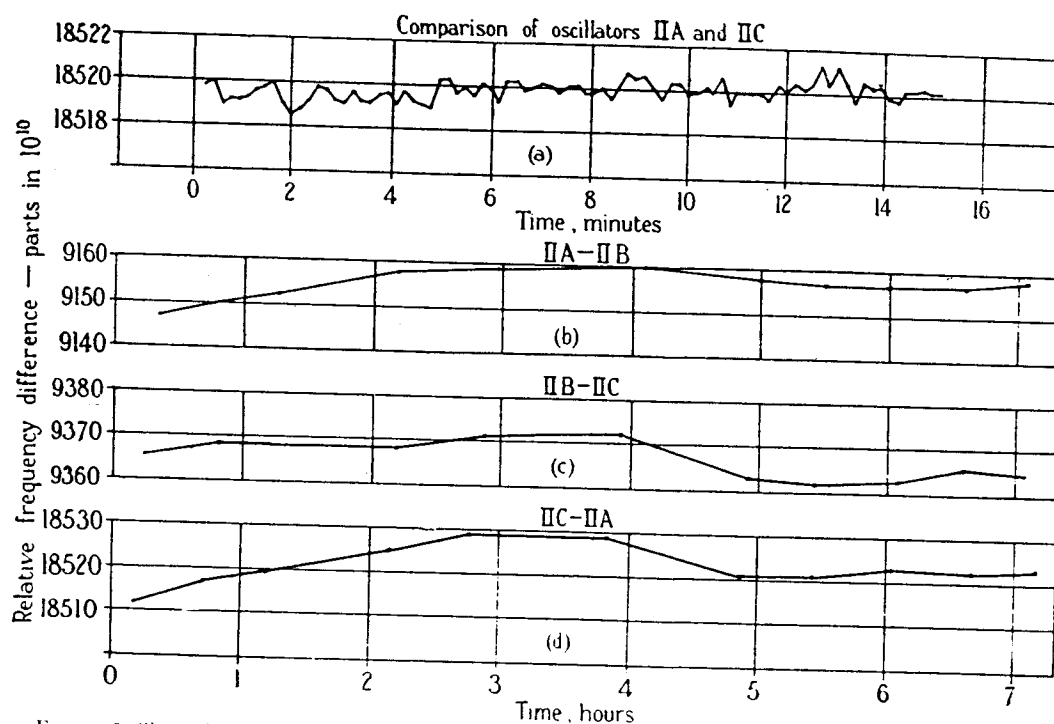
FIGURE 8. Short time fluctuations of precision quartz oscillators of British Post Office, after Booth and Laver[33].

If the servo corrects the quartz clock N times in such a way that only random errors are left, as in the case of errors caused by noise, the averaged fractional time error will be reduced by a factor $N^{1/2}$, as compared to a single correction.[10] If the noise bandwidth is reduced by a factor N, the random fractional time error will also be reduced by a factor $N^{1/2}$. It is clear that in correcting the quartz clock the noise bandwidth must be larger when frequent corrections to one part in 10^{10} are attempted, than for the case of infrequent corrections to one part in 10^8.

In considering the advantages or disadvantages of frequent versus infrequent corrections, we must consider the needs for both low average time errors over long intervals and high instantaneous-frequency constancy on a continuous basis. Will there be any advantage in frequent corrections even if only infrequent corrections are necessary to control the quartz crystal variations for

a particular desired accuracy? The running time of a clock during an interval of N corrections will be N times the period between corrections or N times the reciprocal of the minimum noise bandwidth. For a fixed running time, the random fractional time error will be independent of the rate of correction if the minimum noise bandwidth is always used, which implies essentially continuous control. However, highest instantaneous frequency accuracy requires, in addition, the lowest possible noise bandwidth by using the longest practicable period between corrections and making this period equal to the reciprocal noise bandwidth. The quartz-crystal drive stability greatly aids in this.

A correction in which the servo operates rapidly to determine the correction, using a large bandwidth, but under conditions such that these corrections are infrequently made, will result in random errors larger than minimum. Suppose, for example, that an ammonia clock uses such a stable quartz oscillator that servo corrections are made once a week in an interval of one second. During the period between corrections, the servo is disconnected, and the quartz clock runs uncontrolled at an almost constant rate but with a random value of rate determined by the servo. In this way, maintenance of the complex clock circuits can be carried out, and reliability and long, uninterrupted running times increased. Theoretically, the random time error in this method of operation will be larger over a period of one week than if frequent corrections had been made, i.e., every second, utilizing the same noise bandwidth as was utilized in the weekly corrections but computing the average of the random errors. If essentially continuous control by the servo were possible, utilizing some averaging or integrating process, such that an effective noise bandwidth of the reciprocal of one week were used, the random errors would be reduced. Even though higher accuracy might be possible with essentially continuous control of the quartz clock by the ammonia, it might be necessary to make corrections infrequently to allow for maintenance work and to insure that the clock can run for very long, uninterrupted intervals by reducing to a minimum the amount of equipment required to run continuously. If a whole bank of clocks is used, as in quartz clock practice, continuous control might be more practicable, while ensuring continuous time-keeping records. In any case, it is clear that quartz-crystal drive will result in several different kinds of advantages as compared to a less stable type of drive.

The frequency multiplier chain shown in FIGURE 6 uses ordinary techniques at the lower frequencies and a Sperry 2K47 klystron multiplier tube followed by a Sperry 2K35 klystron amplifier at the 2983.8 Mc. level. The multiplier chain is very frequency-sensitive because of the large number of tuned circuits operating in cascade. It is therefore difficult to swing the signal across the spectrum line with constant input to the absorption cell. This is necessary if an apparent shifting of the absorption line is to be avoided. The tuning of the multiplier chain becomes critical and is difficult to maintain at the proper, constant operating point. In addition, microphonics, hum and other factors cause the output of the chain to be modulated. All of these factors result in what may be termed slow power variations and faster power jitter, which adversely affect the accuracy obtainable. FIGURE 9 shows the output of the

multiplier chain of the Model 2 clock when operated at r.f. power levels which are either too small or too large, illustrating the change in the average level with frequency due to mistuning and also the modulation. FIGURE 10 shows
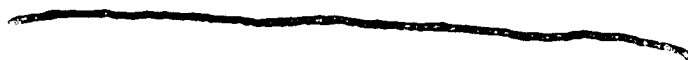
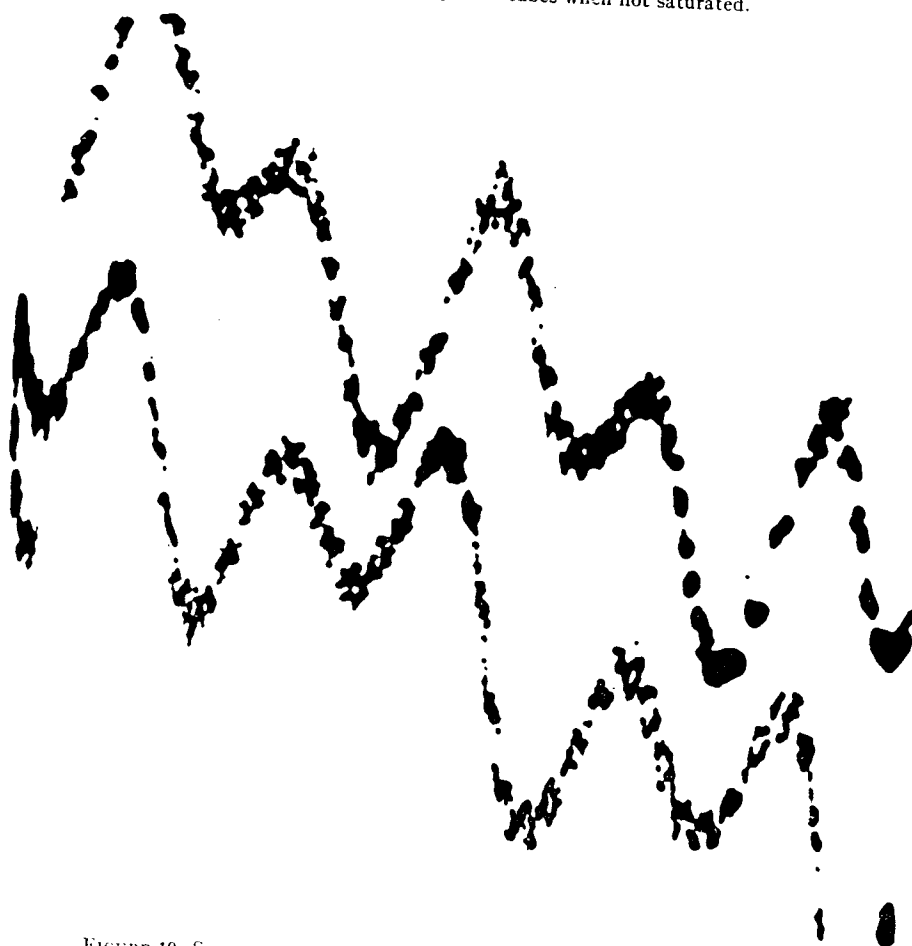FIGURE 9. Output of klystron tubes when not saturated.

FIGURE 10. Same output as for FIGURE 9 with r.f. level adjusted for saturation.

the same output when the r.f. level is adjusted so that the klystron tubes are saturated. The output is now flat and the modulation removed. This mode of operation of the chain employs the klystrons as microwave limiters and is a simple, noncritical and powerful method of improving the performance of such chains in these applications.

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The Model 2 ammonia clock shown in FIGURE 6 can now be discussed in somewhat more detail. The waveforms shown on FIGURE 6 are illustrated more clearly in FIGURE 11. The detailed circuit diagrams and discussion will be published elsewhere.[34]
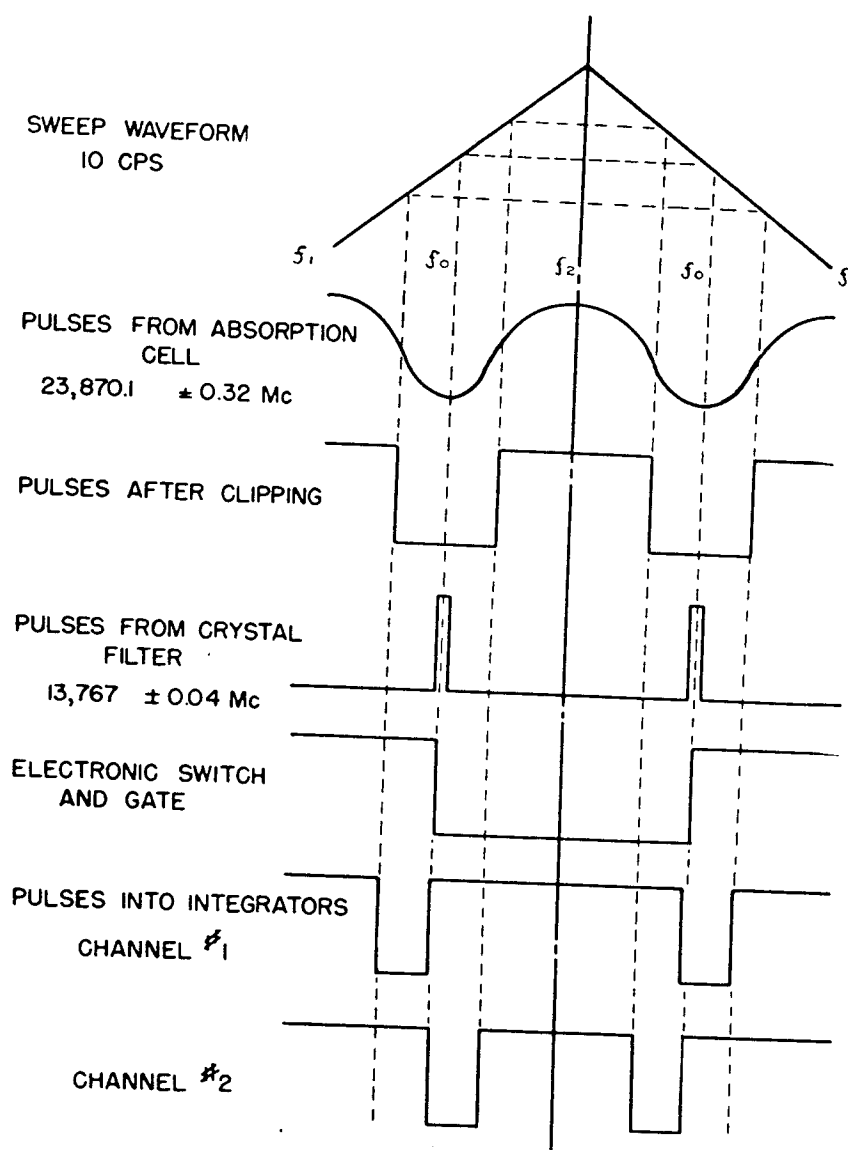
SWEEP WAVEFORM
10 CPS

S_1 S_0 S_2 S_0 S_1

PULSES FROM ABSORPTION
CELL
23,870.1 ± 0.32 Mc

PULSES AFTER CLIPPING

PULSES FROM CRYSTAL
FILTER
13,767 ± 0.04 Mc

ELECTRONIC SWITCH
AND GATE

PULSES INTO INTEGRATORS
CHANNEL #1

CHANNEL #2

FIGURE 11. Waveforms of signals in Model 2 ammonia clock.

The primary signal source for the ammonia atomic clock is a quartz crystal, bridge-stabilized oscillator having a short-time stability of about two parts in 10^8 per day. The frequency of the oscillator may be shifted several parts in 10^6 above and below 100 kc. by an external capacitor connected to the quartz crystal bridge circuit. A frequency divider provides a 1000-cycle source directly controlled by the crystal oscillator for operating the synchronous clock.

In order to reach the particular frequency of the 3,3 line of ammonia and to

provide a sweeping signal across the line, a second signal source is incorporated in the system. This source is a modified, electron-coupled oscillator operating at a center frequency of 13.8 Mc. and is frequency modulated ±0.04 Mc. by a rotating capacitor. The plates of the capacitor are cut for a linear frequency variation with time in both directions of the sweep. The sweep frequency is 10 cps. allowing 20 sweeps per second across the ammonia line.

Conventional vacuum tube multiplier circuits are used to multiply the 100 kc. source to 270 Mc. A klystron multiplier tube is used in the range of 270 to 2970 Mc. The driving power required at 270 Mc. is approximately 1 watt.
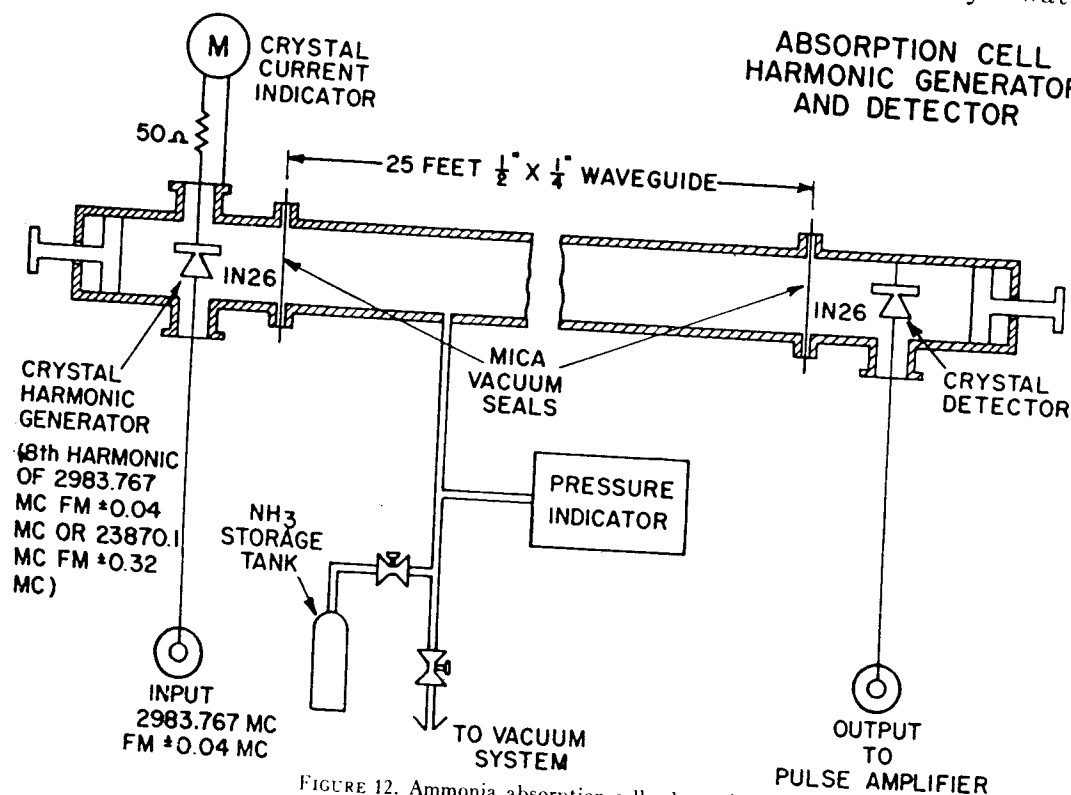
FIGURE 12. Ammonia absorption cell schematic.

The 2970-Mc. signal is combined with the frequency-modulated 13.8 Mc. source in a synchrodyne circuit at the klystron multiplier. The output cavity of the klystron is tuned to the sum frequency of 2983.8-Mc. frequency modulated ±0.04 Mc. A 2K47 klystron is used in the multiplier synchrodyne stage and is followed by a 2K35 klystron amplifier from which an output power of 500 mw. is obtained at 2983.8 Mc. The eighth harmonic of the sum frequency, or 23,870-Mc. frequency modulated ±0.32 Mc., is generated in a germanium crystal mounted in the waveguide of the absorption cell, as shown in FIGURE 12.

The cell consists of 25 feet of $\frac{1}{4}$ in. by $\frac{1}{2}$ in. waveguide with mica window vacuum seals at each end, and is shown in FIGURE 13. The frequency-modulated signal is introduced at one end of the cell, and a germanium crystal at the other end detects the loss of power due to absorption by the ammonia gas.

The discriminator described below has been greatly improved over that used in the Model 1 clock and has been developed by B. F. Husten and E. D. Heberling.[35]   A large variety of discriminators could be used;[1, 2] a promising new method is discussed later.[36]

A pulse is developed at the detector having the appearance of a resonance curve and fed to a pulse amplifier.   The pulse amplitude at the amplifier out-
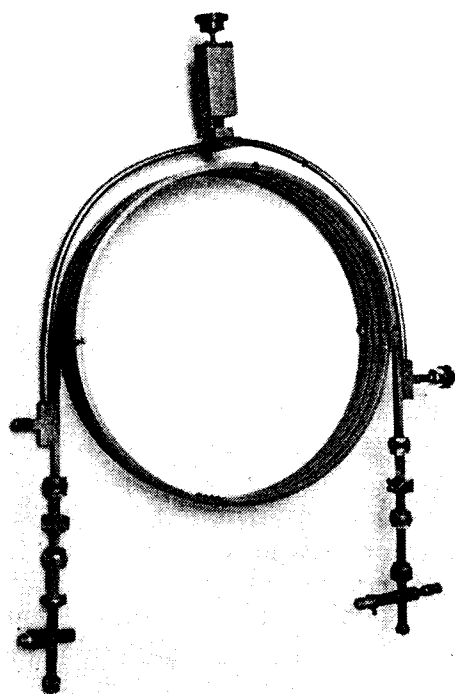

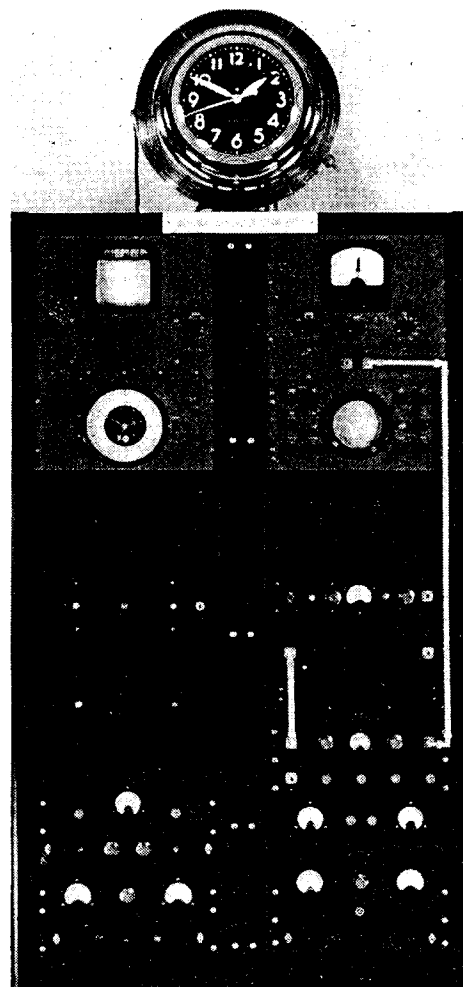FIGURE 13. Photograph of one type of ammonia absorption cell.

FIGURE 14. First NBS ammonia clock.

put is approximately 20 volts.   A direct-current voltage approximately equal to the peak-to-peak amplitude is developed by rectification of the pulse.   A portion of the voltage is obtained in a resistance divider and is used to determine the conducting point of a diode gate thereby passing only that part of the pulse which is equal to or greater than the direct-current voltage applied to the diode.   The divider therefore determines the clipping point and is set for about the half-intensity point on the sides of the ammonia pulse.   The peaks of the pulse are clipped in the following circuit, leaving only a small section of the sides of the original pulse.   After further amplification and amplitude

limiting, a rectangular wave is produced, having a constant amplitude and a length about equal to the half width of the ammonia pulse.

A quartz crystal is used as a passive filter and is ground to the frequency of the 13.8-Mc. FM source corresponding to the center frequency of the ammonia line or 13.767 Mc. Sharp pulses are produced as the FM signal sweeps across the crystal frequency. The exact frequency to which the clock maintains control is dependent upon the quartz crystal filter. The stability requirement of the crystal is reduced from the ultimate stability of the clock by the ratio of 2983 to the crystal filter frequency.

An electronic switch having one input and two output channels is triggered by the crystal filter pulses at a time when the frequency at the ammonia cell is passing through the region of maximum absorption. Therefore the switch is actuated at times during the progression of the rectangular wave derived from the ammonia line, as shown in FIGURE 11. The first part of the rectangular wave is diverted into channel 1 and the remaining part into channel 2.

A separate amplitude limiter and equalizer for each channel is provided so that the only variable shall be pulse length. An integrator for each channel converts changes in pulse length to changes in DC voltages. The circuit arrangement provides for opposing DC voltages of equal magnitude resulting in a net DC error voltage of zero when pulses of equal length are applied to the channel inputs. This condition corresponds to the quartz crystal frequency of 100 kc. when no correction is required.

The DC error voltage is applied to an amplifier and servo motor system having a sensitivity of about ±0.05 volts DC for rotation reversal. A speed reducing gear train completes the servo loop in coupling to the variable capacitor in the 100-kc. quartz-oscillator bridge circuit.

In the event of a frequency deviation at the 100 kc. source, a corresponding shift occurs in the center frequency of the sweeping signal applied to the absorption cell. The position of the ammonia pulse is therefore displaced from its original position and the electronic switch no longer divides the rectangular wave derived from the ammonia line equally for each integrator channel. A DC error voltage is developed, the polarity of which is dependent on the direction of drift at 100 kc. and which has a magnitude in the vicinity of zero error voltage, dependent on the amount of drift.

Time delays in the circuitry causing the ammonia pulse to appear late are minimized by sweeping in both directions, the effect of the delays averaging out in the integrating circuits.

Variations in the ammonia pulse width as may be encountered with gas pressure variations are reduced by cancellation. Pulse width variations which are symmetrical about the center frequency of the ammonia line produce equal changes in DC voltages at the output of the integrators resulting in a net change in error voltage of zero. In practice, the clock has been found to be quite insensitive to pressure broadening changes.

The outputs of channels 1 and 2 are integrated by long-time-constant filter circuits which effectively control the over-all noise bandwidth. This filtering helps greatly in reducing the effect of transients which might get into the cir-

cuits through interference or from the power line. The use of quartz crystal drive, of course, makes it possible to use long filter-time-constants with ease, since the servo does not have to correct such a stable oscillator at a rapid rate. In these circuits, large bandwidths are used where convenient, at certain stages, without making the over-all noise bandwidth large. Specifically, sweep rates of ten per second make it possible to sweep across the spectrum line before power level changes at the input to the absorption cell can take place. The discriminator output for individual sweeps does not, however, control the quartz oscillator but only the average of a large number of sweeps.

FIGURES 14 and 15 show respectively the Model 1 ammonia clock and its performance in controlling a very poor, unstabilized and non-temperature controlled crystal oscillator. Front and rear views of the Model 2 clock are
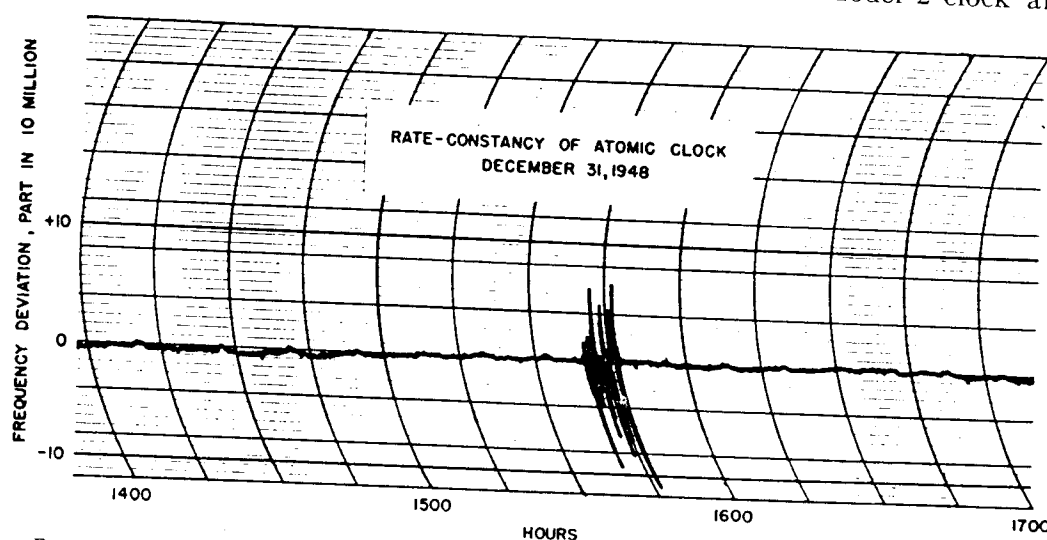

FIGURE 15. Recording showing control of quartz clock by ammonia and deviations when the servo control is removed for the Model 1 clock.

shown in FIGURES 16 and 17. For this clock, FIGURE 18 shows the frequency constancy in the face of forced perturbations of the quartz crystal. The frequency remains constant to about one part in 100 million in spite of perturbations of roughly ten times this amount. The figure shows the frequency variations with the servo loop open and then the control effected by closing the loop. The clock, therefore, has a sensitivity of about 1 in 10^8, that is, the servo will detect and correct for frequency drifts of about ± 1 part in 10^8. The long-time stability is, however, about ± 2 parts in 10^8, as shown by FIGURES 19 and 20. Here, a run lasting about eight days and nights was made and the clock frequency continuously recorded by beating against a standard frequency obtained from the NBS primary frequency standards. In the future, much longer runs will be necessary to obtain definitive information on the long-term constancy of such clocks. The timekeeping accuracy will also be determined in addition to the frequency constancy by integrating the time error over long intervals. If only random errors are present, the averaged time should be very accurate.

Referring to FIGURES 19 and 20, certain features of the recording require explanation. In regions between points marked A and A', the servo control was removed from the 100-kc. crystal oscillator, which then run uncontrolled.
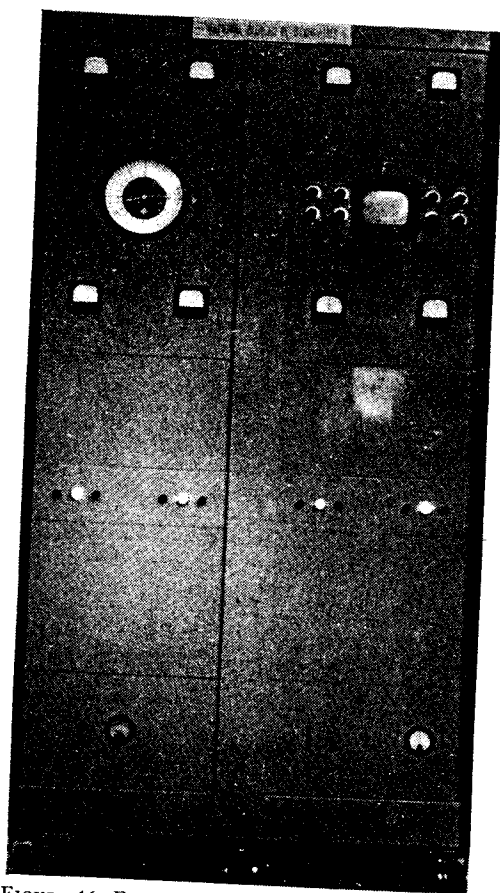
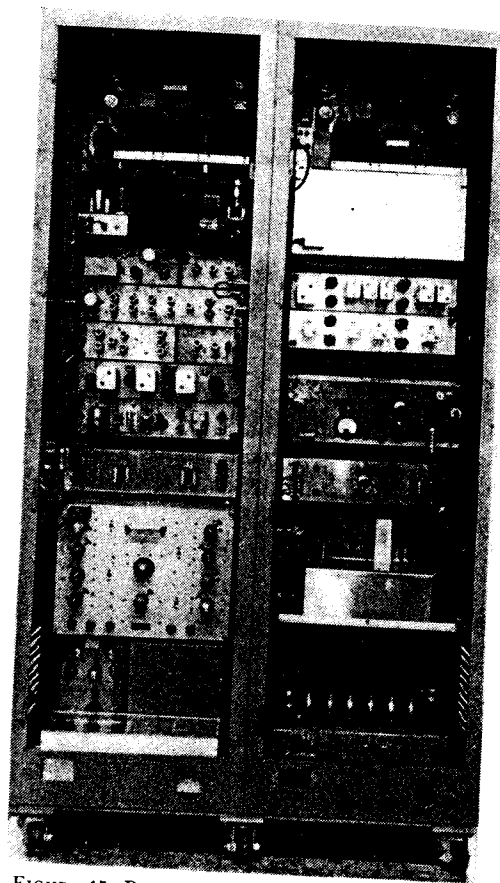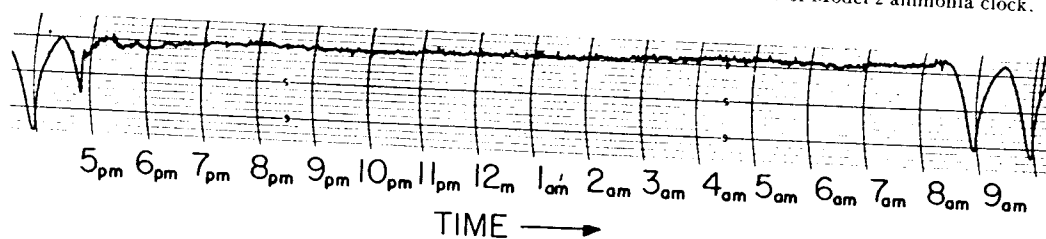

FIGURE 16. Front view of Model 2 ammonia clock.

FIGURE 17. Rear view of Model 2 ammonia clock.

CONTROL OF FORCED PERTURBATIONS. I 1/2 DIVISIONS EQUAL I PART IN 10^8

FIGURE 18. Control of forced perturbations of quartz crystal within ± 1 part in 10^8 by Model 2 clock.

During this time, the absorption cell was repumped and adjusted. This procedure indicates a method for allowing maintenance and servicing while allowing accurate and uninterrupted timekeeping. The recording shows a more constant frequency while uncontrolled than controlled because the short-time constancy of the quartz crystal oscillator is affected by its connection to the servo. With all these connections, the oscillator will drift fairly rapidly if not

controlled by the ammonia. Similarly, if all the connections are removed, there will be the usual cumulative, long-time drift of the quartz crystal but the frequency will be quite constant over short time intervals. Recordings showed the present oscillator frequency to be much more constant when controlled than when uncontrolled, under the condition that all the servo circuits are connected to it as in the normal operation of the clock. The points marked B in FIGURES 19 and 20 are transients affecting the recording system but not the clock. This can be seen by examining the deviations in the clock frequency at the points marked C which occurred during unattended operating periods. The long-time constant involved at points C as compared to the transients at B indicates the recorder only was affected. At the point D, the frequency multiplier chain was tuned up while the servo control was off. This affected the recording system only since the output at 270 Mc. was used to beat against the standard frequency reference.

In the Model 3 ammonia clock being developed, the design calls for completely different tubes, circuits and cells in order to provide further constancy, capacity for resetting to a standard value, and long-time running of perhaps a year or more. The multiplier chain will use the Sperry SMC-11 and SMK-40 klystrons shown in FIGURES 21 and 22. The SMK-40 has an output at the 3,3 line frequency of ammonia and has been successfully operated with an output of one watt. This will allow excess power to drive an absorption cell of large cross-section and a large reserve for padding so as to control undesired reflections in the system. The tube can be run at a low level to increase its life.

A minimum of two separate quartz oscillators using the best crystals available will be controlled by the ammonia at intervals of several days, depending on the drift rate of the crystals. Between corrections, the oscillators will run uncontrolled and will be continuously compared against each other. In this way, maintenance work can be carried out, and breakdowns elsewhere than in the oscillators can be prevented from interrupting the running of the clocks. Before the automatic control is applied, all circuits can be checked for proper operation, further enhancing reliability.

Preliminary work has been carried out on a new discriminator method which shows great promise for the Model 3 clock. The method is illustrated by FIGURE 23. The frequency multiplier chain is slowly frequency-modulated about the center frequency of the absorption line, and the line is used as a demodulator converting the FM to AM as shown. If the quartz-crystal frequency is centered, the output signal has equal peaks, while for off-center operation the output is asymmetric with the direction of off-centering indicated by the asymmetry. The FM sweep width is adjusted for optimum sensitivity such that the turning points are at the frequencies where the rate of change of signal is a maximum. If peak readings of the output signal are used to indicate the correct center frequency, the method will be inherently independent of pressure broadening, and the magnitude, rate and linearity of the frequency modulation. Equal frequency deviations each side of center are assumed.

A relatively crude spectrograph has been built to test the possibilities of

FREQUENCY STABILITY RECORDING OF THE AMMONIA ATOMIC CLOCK
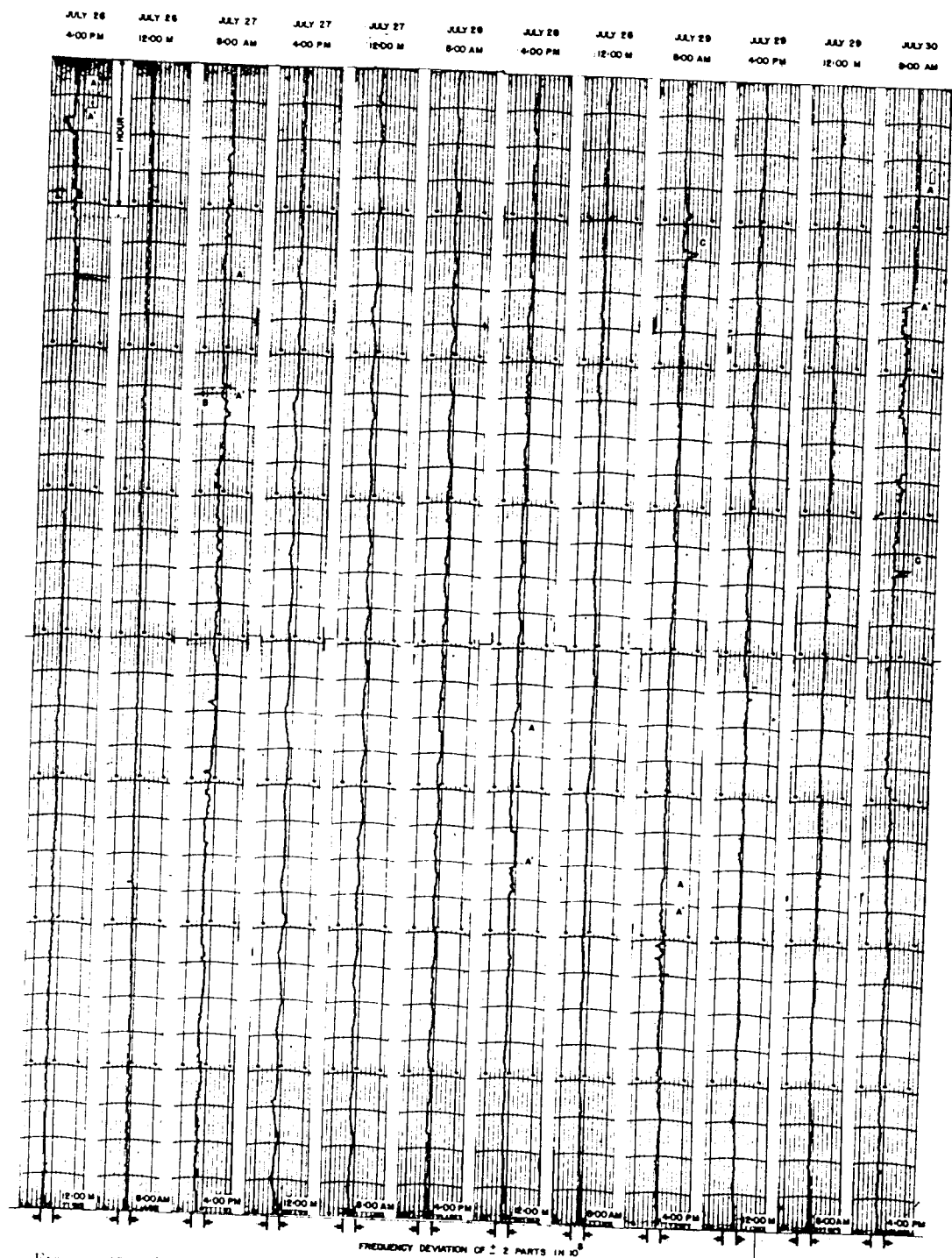
FIGURES 19 and 20. Frequency constancy of Model 2 clock showing stability of ±2 parts in 10^8 for test running about eight days.
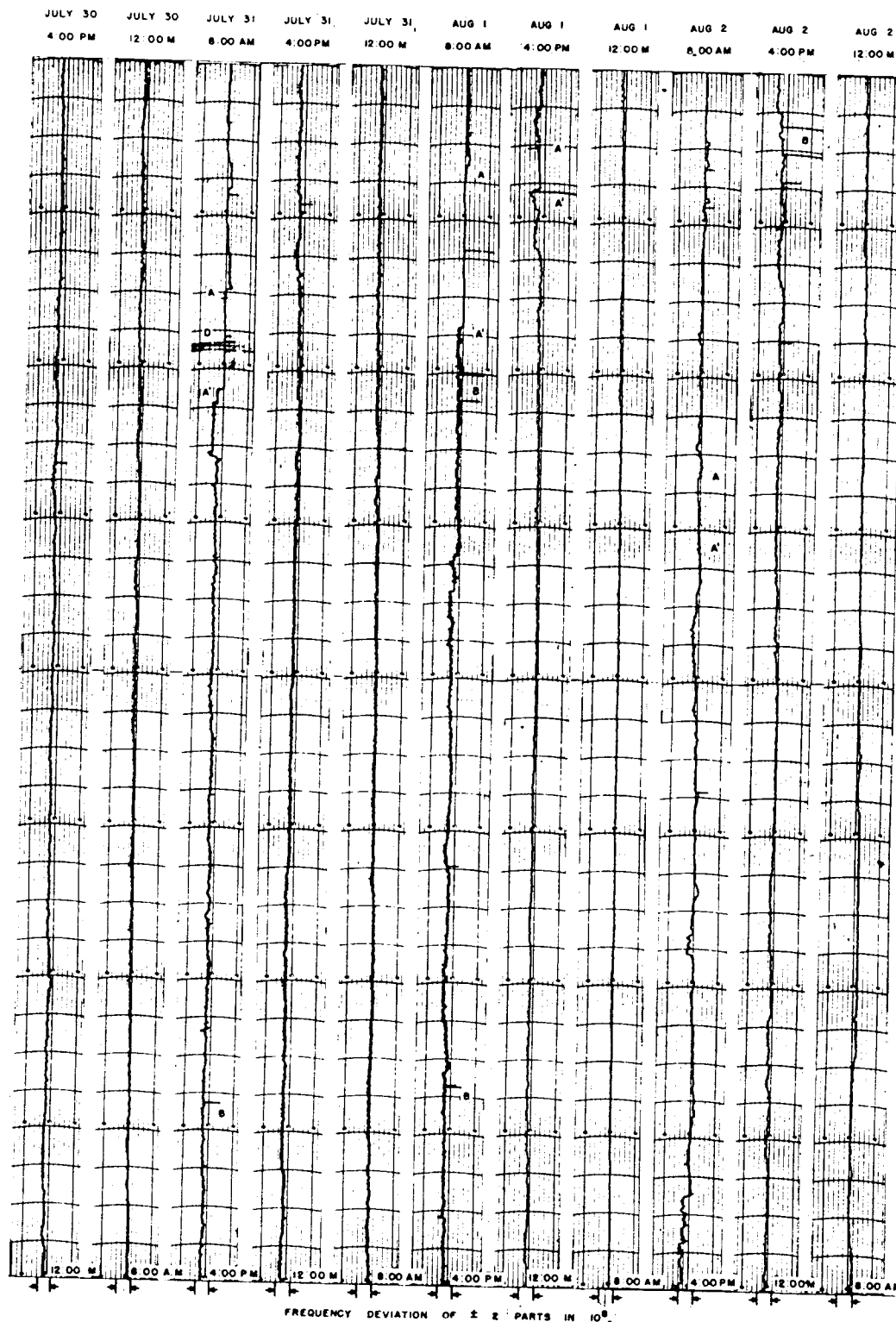
FREQUENCY STABILITY RECORDING OF THE AMMONIA (ATOMIC CLOCK)

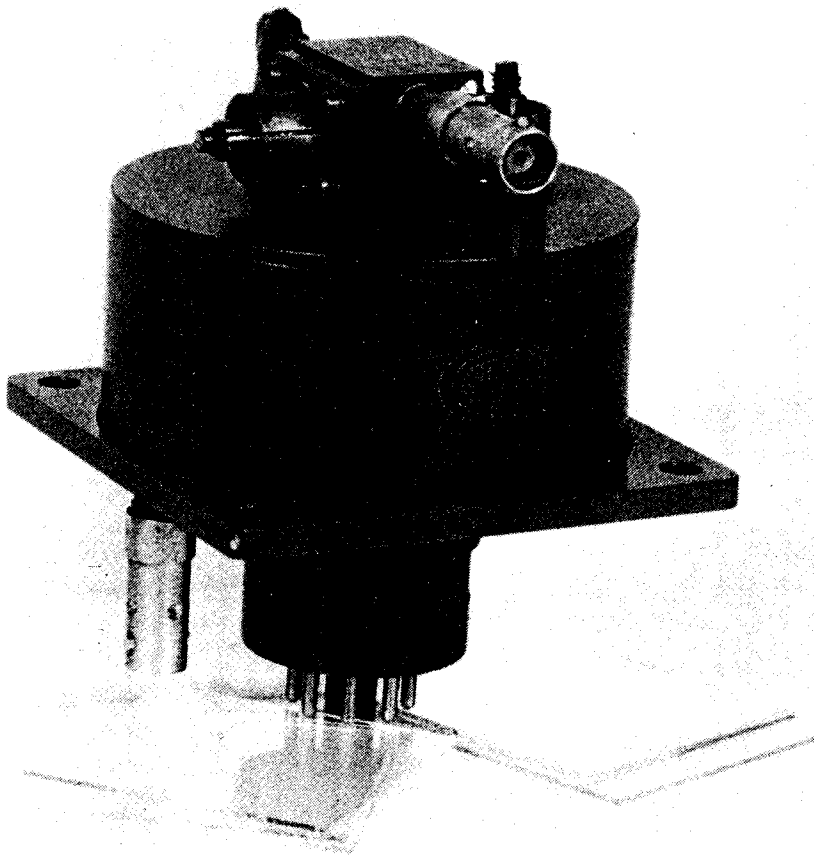FREQUENCY DEVIATION OF ± 2 PARTS IN 10^8.

FIGURE 21. Sperry SMC-11G klystron multiplier for use with ammonia 3,3 line.

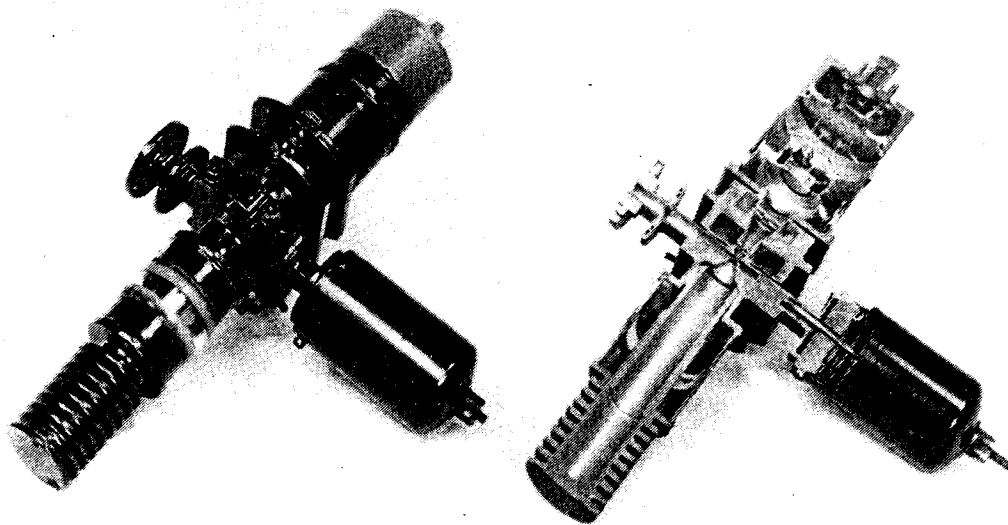FIGURE 22. Sperry SMK-40 klystron multiplier driven by the SMC-11G with one watt output at 23,870 Mc.

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the essential scheme given above. Microphonics, shielding difficulties and other factors, due to the simple setup used, limited the accuracy obtained in the test. Quartz crystal drive was used with a broad-banded multiplier chain to allow for the FM. The final multiplier stages used the Amperex AX-9903, the 2C39-A, SMC-11 and SMK-40 tubes in that order. Saturation of the output tubes was obtained without use of additional amplifiers between multiplier stages so that the klystrons operated as microwave limiters. A superheterodyne detector was used; and an improved unit, using a local oscillator signal derived from the input signal, and sweeping with it so that a constant i.f. frequency is obtained, is now under development. Frequency setting sensitivities of one part in 10 million were easily obtained and the internal consistency of the settings was good, using only an oscilloscope as an indicator.
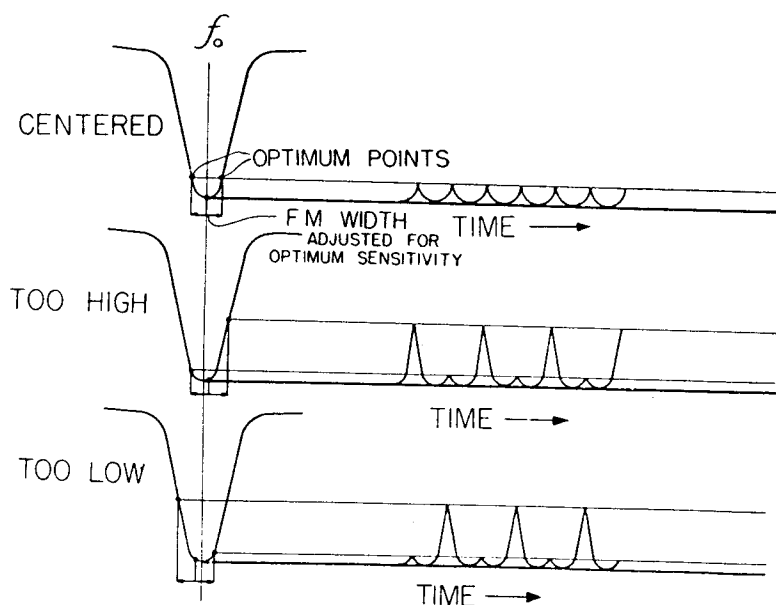

FIGURE 23. FM discriminator method for use in clock or spectrograph.

In particular, the ability to set precisely to the center frequency was demonstrated. This is a necessary prerequisite for a primary frequency standard or clock as compared to making a clock which will run at a constant rate but in which it is difficult to adjust this rate to the primary standard value without external reference standards. Proper instrumentation of the indicator will greatly improve the sensitivity obtainable. In the Model 2 clock, difficulty in setting to the center frequency of the ammonia line has been experienced.

Atomic Beam Control of Quartz Clock

The adaptation of the previously described servo method to an atomic beam equipment is shown in FIGURE 24. The beam equipment is of the Rabi magnetic resonance type[37] and is shown in FIGURE 25. The advantages of atomic beams result from their virtual elimination of collision and Doppler broadening. Power saturation is also avoided since a fresh stream of unexcited atoms constantly emerges from the oven. Unprecedented Q's should be possible using

cesium or thallium atoms. In this first unit, cesium has been used. Both cesium and thallium were suggested by P. Kusch.[38]

The Q obtained by this method is determined by the time the atoms spend in the transition or excitation region shown between points 4 and 6 in FIGURE
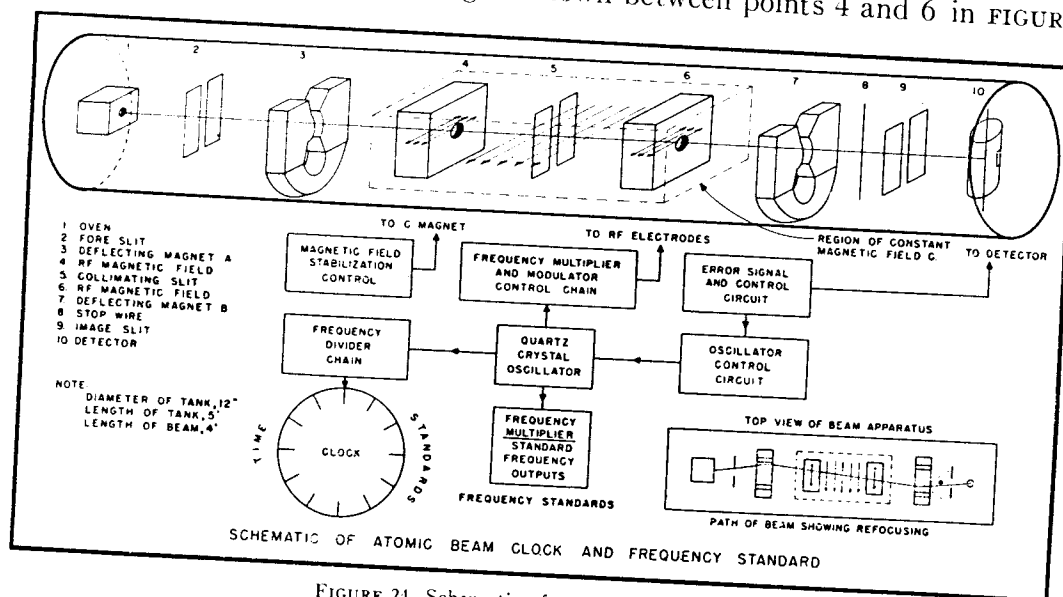
FIGURE 24. Schematic of atomic beam clock.

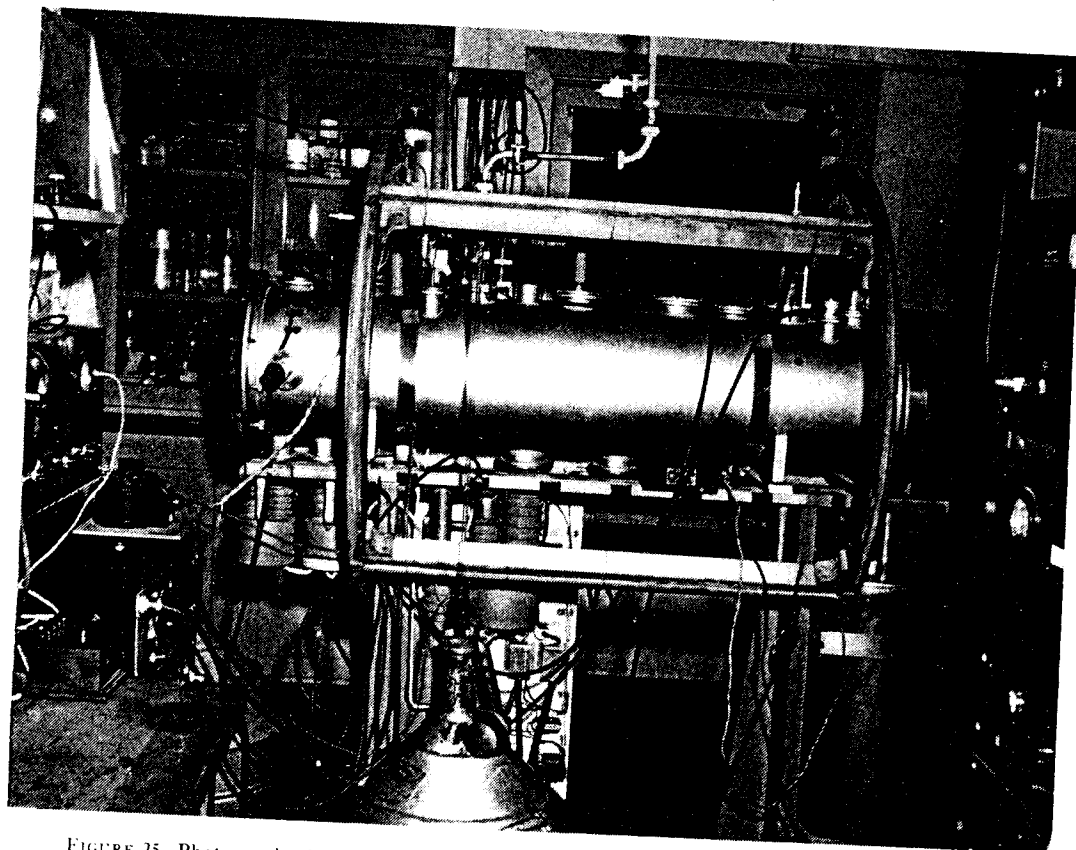FIGURE 25. Photograph of cesium atomic beam apparatus exclusive of the electronic equipment.

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24. The sharpness of resonance is determined by the uncertainty principle, $\Delta E \cdot \Delta t \sim h$, where ΔE is the width of the energy level and Δt is the lifetime of the state, that is, the excitation time, and h is Planck's constant. In terms of the transition path length, L, and the thermal velocity of the beam,

$$Q = f/\Delta f = Lf/\text{velocity} \tag{27}$$

which indicates that high frequencies and long path lengths should be used to obtain high Q. It is partly for this reason that heavy atoms, such as cesium or thallium, are planned for use in this clock, since they have frequencies high up in the microwave range. The theoretical values of Q for many atoms have been verified experimentally at lower frequencies and lower Q's in work at Columbia University as well as for cesium at NBS. It is difficult to obtain the desired excitation of the beam over a long path length such as 50 cm. at the cesium and thallium wavelengths of approximately 3 and 1.5 cm. The separated oscillating field method developed by N. F. Ramsey[39] can be applied to this problem and is planned for the NBS unit. P. Kusch has carried out preliminary experiments to verify the operation of this method and has obtained the expected Q values using potassium at about 500 Mc.[40] For a 50-cm. path length, the Q's of cesium and thallium should be about 30 and 90 million, respectively. In the Ramsey method, output-current curves from the detector can be obtained which look like either absorption or dispersion curves according to the adjustments in the excitation circuits.[39] Here the beam is excited only at the beginning and the end of the transition path length, making excitation easier. The dispersion type output offers another promising method for setting to the center of the transition, since the output signal has a maximum rate of change at the resonance frequency.

The transition used in cesium depends on the splitting of the ground state by the interaction of the magnetic moment of the nucleus with the extranuclear electrons. This splitting amounts to 9192.632 Mc. The atoms are space quantized and are focused on the detector by the inhomogeneous fields of magnets at points 3 and 7 shown in FIGURE 24. The exciting field in the region of homogeneous magnetic field C causes transitions from one orientation to another, thus changing the focusing action and detector beam current.

The technique of beam methods requires that magnetic field C be finite rather than zero, or space-quantization of the atoms is destroyed and the beam is defocused.[41] The transition frequency for zero magnetic field is the ground state splitting of the atom, and this, of course, will be altered by the Zeeman effect. It is therefore necessary to reduce the effect of this external field on the frequency to negligible proportions.

This can be done, as seen from an inspection of the following formula for the transition frequency:[38]

$$f = \Delta\nu + (g_J - g_I)^2 \frac{\mu_0^2 H^2}{2h^2\Delta\nu} \tag{28}$$

This is the frequency of the σ-line of cesium for a change of total angular momentum quantum number F from $(F = 4) \rightarrow (F = 3)$. F is the sum of the
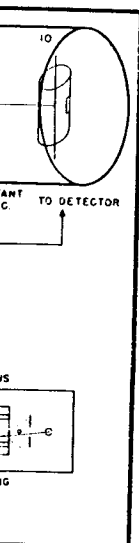
nuclear plus the electron angular momenta. In this transition, the magnetic quantum number change $\Delta m = 0$. Numerically, the frequency is given by $f = (9192.632 + 0.000427H^2) \times 10^6$ sec.$^{-1}$. The symbols in the above equation are as follows: f = frequency; h = Planck's constant; H = magnetic field strength; $\Delta\nu$ = transition frequency at zero field (ground state splitting); μ_0 = Bohr magneton; $I = 7/2$ for cesium, nuclear angular momentum in units of $h/2\pi$; $J = 1/2$, electronic angular momentum in units of $h/2\pi$; $g_J/g_I = -5018$, $g_J = 2$; and $\Delta\nu = 9192.632 \pm 0.002$ Mc. In the above equation, H must be small and $\Delta\nu$ large, indicating that atoms such as cesium or thallium with large values of $\Delta\nu$ should be used to reduce the Zeeman effect. For cesium, the magnetic field must be kept constant within only ten per cent for fields of 0.1 gauss for a frequency constancy of one part in 10^{10}. The transition $(4,0 \rightarrow 3,0)$ given above is almost completely field independent; other field-sensitive transitions will give lines completely resolved from this line at fields of 0.1 gauss.

The beam equipment shown in FIGURE 25 has been operated with a preliminary transition path length of about one cm. obtained by passing the cesium beam through the exciting x-band waveguide. The excitation was obtained by means of a quartz crystal oscillator and frequency multiplier chain terminated in Sperry 2K47 and 2K46 klystron multiplier stages. This method gave the necessary stability and made precision measurements possible. The expected Q of the order of 350,000 was obtained. The Ramsey method of excitation was not used in order to make the first excitation of this transition relatively easy to find. The field-insensitive σ-line given above was measured giving the value 9192.632 ± 0.002 Mc.[42] A recording showing two traces of this line is shown in FIGURE 26. The symmetry and large signal-to-noise ratio indicate the possibility of frequency measurements to a small fraction of a line width. By measuring with the steeply sloping sides of the line, it has been found in preliminary work that measuring sensitivities of better than one part in 20–50 million can be obtained.[42] This work is now in progress with indications that further improvements of perhaps an order of magnitude may be possible. If a path length of 50 cm. can be used, however, increasing the Q over 50 times (the Ramsey method further sharpens the output curves), these results give experimental evidence indicating that an accuracy of one part in 10^9 or better can be achieved. Still higher values may be indicated for thallium.*

Conclusion

If the accuracies indicated in the previous section can be achieved over long time intervals, it may be possible to compare atomic time against the mean sidereal year. The day does not seem constant enough for a significant comparison of this type in a reasonable length of time. A long-running, precision atomic clock could also be used for time-keeping between measurements of the mean sidereal year, just as the quartz clock is used between measurements of the day through observation of successive star transits. The application

* Since this paper was written, the cesium line has been excited with a path length of 50 cm. by the Ramsey method. A Q of 30 million has been obtained with intensity and symmetry similar to that shown in FIGURE 26. A point-by-point plot of the line and recordings have both been made. These results indicate an accuracy of one part in 10 billion may be achieved. Further measurements are in progress.

of atomic clocks and microwave spectrographs to measurement of fluctuations in the length of the day appears a promising possibility. The application to a new unit for time and frequency also seems encouraging. A related possibility appears if millimeter wavelength interferometers can be built with high precision, since these could be illuminated by radiation derived from a clock and the standard of time and of length, perhaps based on a single spec-
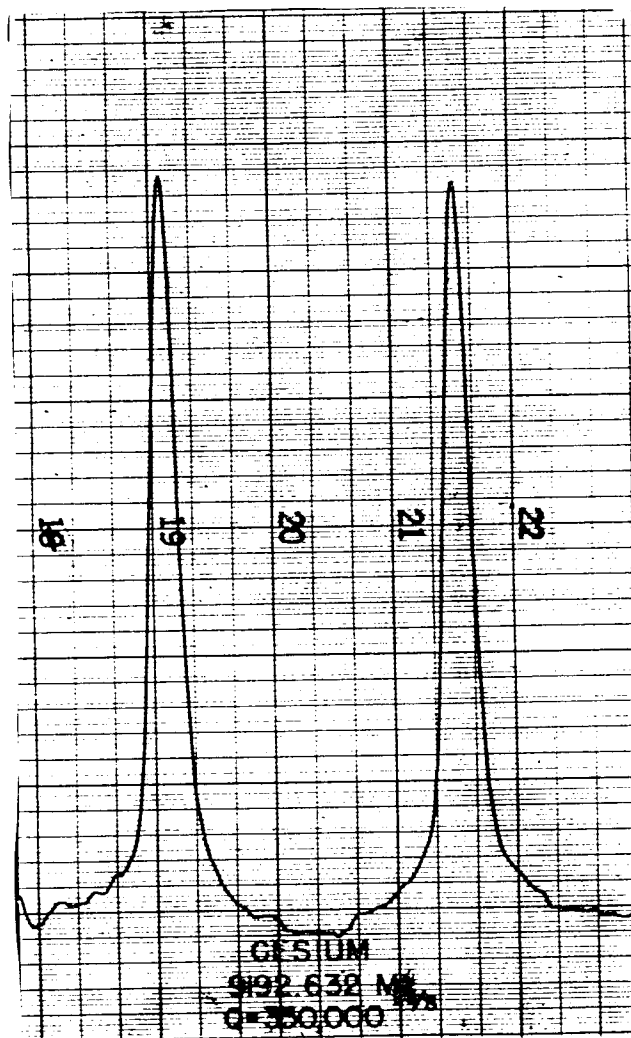
FIGURE 26. Recording of field-insensitive cesium line; two traces of the same line are shown.

trum line.[43] Progress in millimeter radiation sources and detectors should help this application.

In the references which have been listed it will be noted that an attempt has been made to provide, at least in part, a working bibliography. Several of the papers cited also contain extensive bibliographies.

Acknowledgment

The aspects of the NBS program covered above include most of the projects relating to atomic frequency and time standards and include contributions

from many groups. Some projects on pressure shifts, nuclear quadrupole absorption, the deuterated ammonias, spectral tables and checks on the rotation of the earth by microwave spectroscopy methods are not covered here. In addition to the more detailed references in the text, it is a pleasure to list the following contributors to the program. Professors C. H. Townes and P. Kusch have been consultants on the gas and quadrupole absorption techniques, and on the beam techniques, respectively. B. F. Husten and E. D. Heberling have worked on the first two ammonia clocks. L. J. Rueger has assisted on the test of the Model 2 clock and is working on the development of the Model 3 clock. J. E. Sherwood and R. H. McCracken have worked on the cesium clock, and Sherwood is particularly responsible for the design and construction of the beam part of the apparatus itself, with the assistance of Professor Kusch. G. J. Griffin has assisted in work on atomic oscillators and quadrupole absorption, and P. Wacker has assisted with the theory of quadrupole absorption. To all of these co-workers, the author wishes to express his appreciation.

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Reference 48 shows that the annual fluctuation in the rate of rotation of the earth has diminished by 40 per cent in amplitude in 1950–1951 as compared to that of 1934–1949. In reference 44, Mintz and Munk use new meterorological data to calculate the seasonal fluctuation in the rotation of the earth. The zonal winds are now found to account for only one-third of the reported fluctuation, with a large uncertainty in this result because of inadequate data in tropical and southern latitudes. In reference 47, a new interpretation of the irregular fluctuations in the earth's rotation is suggested.