ATOMIC FREQUENCY STANDARDS AND CLOCKS

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THERE HAS recently been a great deal of interest in the high stability and accuracy obtainable with atomic frequency standards and clocks. Scientific interest has been stimulated by the recent ether drag experiments[1] using ammonia beam masers and by the possibility of using an atomic clock in a satellite to check on the gravitational frequency shift predicted by the general theory of relativity.[2] The need for high-precision frequency and time measurements in satellite and missile tracking, in space and terrestrial communication systems, and in navigation has provided impetus for rapid developments in the field.

Three types of atomic standards appear today to be the most promising for the immediate future. They are the cesium beam, the ammonia maser, and the alkali vapor frequency standard. The first two types are discussed at length in other conference papers and their general characteristics will be assumed known. However, a description of one kind of alkali vapor frequency standard* will be given in order to facilitate comparison with the cesium beam and ammonia maser instruments.

RUBIDIUM VAPOR STANDARDS

Rubidium vapor frequency standards are now being constructed at a number of laboratories. They are based on the hyperfine transition at 6835 Mc/s in the ground state of Rb^{87}. A schematic

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*Alkali vapor frequency standards using optical pumping and Doppler reduction techniques were suggested by R. H. Dicke. The earliest experimental work on them was done with rubidium by T. R. Carver, and the first optical detection of the transitions was achieved by Arditi and Carver with sodium.

diagram of the apparatus used at the National Bureau of Standards and the Naval Research Laboratory[3] to observe this transition is shown in Fig. 1. Light from a rubidium discharge was passed through a Rb^{85} filter bulb and then to the sample. The intensity of the light absorbed and spontaneously reemitted by the Rb^{87} in the sample was observed as a function of the applied microwave frequency.
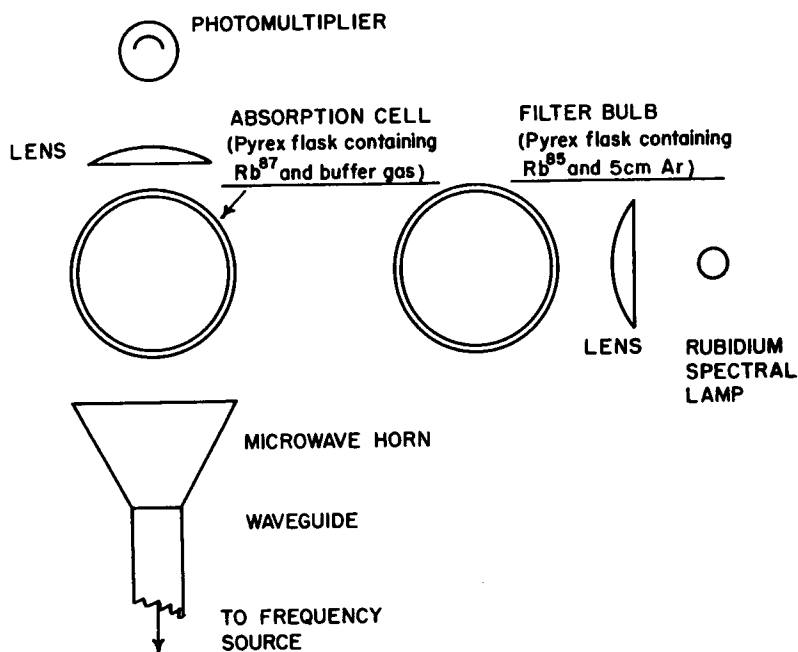
PHOTOMULTIPLIER

LENS

ABSORPTION CELL
(Pyrex flask containing
Rb^{87} and buffer gas)

FILTER BULB
(Pyrex flask containing
Rb^{85} and 5cm Ar)

LENS

RUBIDIUM
SPECTRAL
LAMP

MICROWAVE HORN

WAVEGUIDE

TO FREQUENCY
SOURCE

Fig. 1. Schematic diagram of apparatus for optical detection of the ground state hyperfine transition in Rb^{87} vapor

Fig. 2 shows an idealized picture of the hyperfine components of either of the two main rubidium resonance lines at 7800 and 7947 Å. Excited state hyperfine splittings are neglected. The Rb^{85} absorption lines in the filter, broadened and perhaps somewhat shifted by the presence of another gas, absorb one of the two Rb^{87} hyperfine components more strongly than the other. The light reaching the sample thus consists mainly of component (b). Atoms in the $F = 1$ sublevel are therefore preferentially raised to the excited states. Since the atoms return by
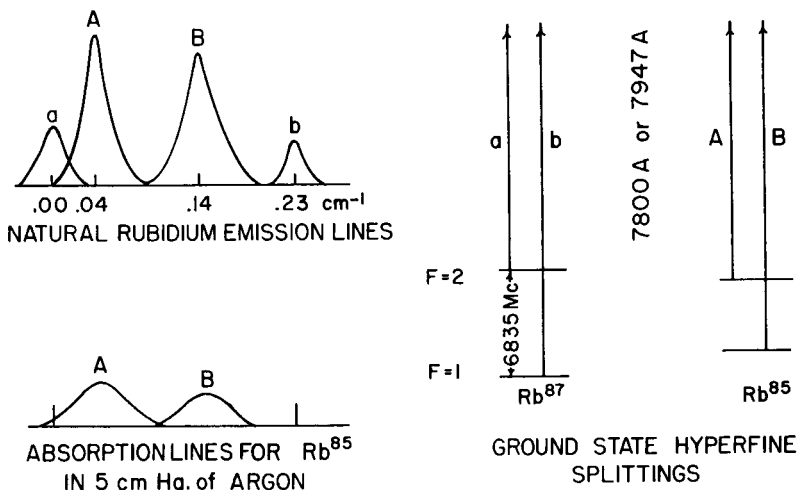
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Fig. 2. Hyperfine structure of rubidium resonance lines

spontaneous emission to both ground state sublevels with simi-
lar rates, an excess population in the $F = 2$ sublevel is pro-
duced. As a result, the optical absorption coefficient for the
sample decreases, since fewer atoms are in the state where they
can absorb component (b).

The equilibrium level populations will depend on the rate of
photon absorption and the rate at which atoms can return from
the $F = 2$ to the $F = 1$ sublevel by relaxation or by some other
means. When hyperfine transitions are induced, the population
of the $F = 1$ sublevel increases. More light is absorbed and re-
radiated by the sample, and the photomultiplier output increases.

For a compact unit it is desirable to place the sample in a
cylindrical TE_{011} mode cavity with holes cut in the ends and to
detect changes at resonance in the amount of light absorbed by
the sample. The amount of transmitted light can be measured by
a silicon solar cell. Satisfactory rubidium light sources have
been run on three watts of power and transistorized versions
running on one watt or less should be possible. The remainder
of the apparatus, exclusive of temperature control, can be run on
about one-half watt. This would include, for instance, a quartz
crystal oscillator at 4.996 Mc/s, a frequency multiplier to 59.95
Mc/s, a crystal harmonic generator giving the final factor 114

multiplication, and a phase modulation and servo system to lock the oscillator to the center of the hyperfine resonance line.

PRACTICAL LIMITATIONS OF PRESENT ATOMIC STANDARDS

A discussion of the precisions obtainable with passive atomic standards requires some assumptions about how well one can pick out the center of the resonance line. I will assume that with care and a reasonable signal-to-noise ratio one can pick out the center of a symmetrical line to 1 part in 10^3, and that any higher stability requires experimental demonstration.

For the cesium beam frequency standard, the primary limitation comes from transit time broadening of the resonance line. Essen and Parry[4] in England have succeeded in achieving an accuracy of 1 part in 10^{10} using a Ramsey cavity separation of about 50 cm and a line width of 350 cycles. For the National Company Atomichron, a commercially available version of the cesium beam standard, the separation between Ramsey cavities is about 100 cm and the linewidth is 120 cycles. The signal-to-noise ratio is about 100 for a one second time constant. This should permit a stability of 1 part in 10^{11}, but there are some additional difficulties with phase shifts between the cavities, frequency synthesis, stability of the flywheel oscillator, etc. Currently available models have shown an accuracy[5] of about 2 parts in 10^{10} but have frequently given day-to-day stabilities over periods of weeks or more of several parts in 10^{11}.

A new model of the Atomichron which will be available soon is expected to have an accuracy of about 5 parts in 10^{11} and a day-to-day stability of 1 part in 10^{11} or better. In this model, the R.F. structure has been carefully designed to minimize phase shift errors. Direct multiplication from an oscillator at about 5.4 Mc/s is used to avoid frequency synthesis difficulties, with synthesis of an even frequency output being done in a separate frequency translation unit. Longer beam tubes with cavity separations of 3 to 4 meters should be in operation soon at the National Physical Laboratory in England and at the Boulder Laboratories of the National Bureau of Standards. Linewidths of 50 cycles have already been achieved with the NPL apparatus. It

thus appears that accuracies of a few parts in 10^{11} may be attainable within the next year or so. Several proposals for still longer apparatuses have also been made. The accuracy of comparison with astronomical time[6] is at present limited mainly by the accuracy of the astronomical measurements, and this will apparently continue to be the case for some time.

For alkali vapor frequency standards, the linewidth limitations are not as well understood as for the cesium beam. In order to reduce disorientation by collision with the wall, one has to have a suitable buffer gas in the sample or a non-disorienting coating on the wall. If the phase of the R.F. field is uniform over the sample or if the diffusion rate of alkali atoms through the buffer gas is low, the Doppler breadth will also be greatly reduced.[7] With a completely non-disorienting buffer gas or wall coating, one could hope to obtain arbitrarily narrow lines. Dehmolt actually showed that one could obtain relaxation times of the order of a few tenths of a second with an inert buffer gas[8] and seconds with an eicosane wall coating. Since then linewidths as low as three cycles have been obtained for Zeeman transitions in rubidium vapor.

Despite the demonstrated long relaxation times, it has not been possible to obtain correspondingly narrow hyperfine transition lines. For cesium, the narrowest lines obtained have been 40 cycles with neon or helium as buffer gas and 120 cycles with argon. For rubidium-87, linewidths of 20 cycles at 6835 Mc/s were obtained with neon or helium as the buffer gas. In all cases the observed relaxation times were much longer than would correspond to the observed linewidths. For rubidium in neon, the linewidth remained about 20 cycles over a range of pressures from 1 cm Hg to 20 cm Hg.

In addition to broadening the lines, collisions with the buffer gas or wall coating can also cause shifts in the frequency at which the transition occurs.[9] The shifts are of the order of 1 part in 10^6 per cm Hg of buffer gas, with the lighter gases giving positive shifts and the heavier gases negative ones. One can make mixtures of buffer gases which will give very small pressure shifts. However, since sealed samples are used in the frequency standards, the important quantity is the change in fre-

quency with temperature. The shifts in frequency with temperature for sealed glass samples were found to be of different signs for different buffer gases, but not proportional to the corresponding pressure changes. Samples containing rubidium-87 in a 50-50 mixture of argon and neon at a pressure of 1 cm Hg were found to have a temperature coefficient of only 3 parts in 10^{11} per degree Centigrade near room temperature, and still smaller temperature coefficients should be possible.

The frequency shifts can be thought of as being caused by successive small relative phase changes in the coefficients of the $M_F = 0$ parts of the alkali wave function. In each collision, the relative phase is changed somewhat by the integrated effect of the shift in hyperfine splitting as a function of distance between the atoms. The shift in angular frequency is given by the average phase shift per collision times the number of collisions per second. The average phase shift per collision is found to be of the order of 10^{-3} radians.

The observed linewidths are at present believed to be due to statistical fluctuations in the phase shifts. If σ is the standard deviation of the phase shift per collision and Σ is the standard deviation in the total accumulated phase shift for an observation time T, then $\Sigma \sim \sigma \cdot (nT)^{\frac{1}{2}}$ where n is the number of collisions per second. The observed linewidth will then be given by $\Delta \nu \sim \Sigma / T \sim \sigma \cdot (n/T)^{\frac{1}{2}}$, or $\Delta \nu \propto \sigma \cdot (p/T)^{\frac{1}{2}}$. In the linewidth measurements mentioned above, the observation time T was evidentally limited in some cases by diffusion of the alkali atoms into regions of quite different relative R.F. phase, and T was thus proportional to P. This would then give a roughly pressure-independent linewidth, as observed for rubidium in neon. At high enough pressures one would, of course, expect T to be proportional to $1/P$ because of relaxation, and the linewidth would then be proportional to P.

Although one can obtain lines as narrow as 20 cycles with gas cells, it is probably desirable to allow this width to be about doubled by light intensity and R.F. power broadening in order to improve the signal-to-noise ratio. Under these conditions, a signal-to-noise ratio of about 100 has been obtained for a labo-

ratory apparatus with a time constant of .01 sec. For reasonable periods of time, one would thus expect a stability of somewhat better than 1 part in 10^{11}. However, the possibility of changes in the buffer gas pressure or composition must also be considered. The leakage of 10^{-4} mm Hg of nitrogen into the sample, for instance, would cause a frequency shift of 1 part in 10^{11}. Such a large change in buffer gas composition for a carefully prepared sample seems unlikely except over quite long periods of time. The accuracy with which one can determine the unperturbed transition frequency with a gas cell device will, of course, be limited by the need to extrapolate to zero buffer gas pressure. For this reason, alkali vapor devices will probably be much more useful as secondary standards than as primary standards.

For the ammonia maser frequency standard,[10],[11] the limitations are of quite a different nature than for the other types of frequency standards. The existence of unsymmetrical quadrupole structure in the 3,3 transition has been a problem until recently. However, this difficulty has now been overcome by Bonanomi and by Shimoda by using either nitrogen-15 ammonia or the 3,2 transition in natural ammonia.

The largest remaining difficulty is pulling of the transition frequency when the resonant cavity is detuned. The frequency shift is given by

$$\left(\frac{\Delta \nu}{\nu}\right) \sim \left(\frac{Q_C}{Q_L}\right) \cdot \left(\frac{\Delta \nu_C}{\nu_C}\right) ,$$

where $\Delta \nu_C$ is the error in cavity setting, Q_C is the quality factor of the cavity, and Q_L is the ratio of the frequency to the width of the resonance line when the device is used as a spectrometer. Under normal conditions $(Q_C/Q_L) \sim 10^{-3}$, and thus the requirements on cavity tuning are quite severe even with careful thermostating and the use of low expansion coefficient materials for the cavity. One method which has been used to meet this problem is to couple two cavities together in such a way that the cavity response is quite flat on top.

Recently Bonanomi has achieved a resettability of 3 parts in 10^{11} by using a magnetic modulation method. If a magnetic field of a few gauss is applied when the cavity is detuned, Q_L will be

decreased by broadening of the resonance line and the frequency shift will increase. The cavity is swept slowly through resonance and the emitted frequency is measured with respect to another standard alternately with and without the magnetic field present. When the successive measurements agree, the cavity is on resonance and the pulling is very small.

If the effect of cavity pulling is taken care of, the next problem becomes one of Doppler shifts due to running waves in the cavity.[12] For a parallel beam of molecules and conditions well away from saturation, the stimulated emission will take place mainly in the rear part of the cavity and there will be a net flow of power toward the front of the cavity. The Doppler shift will be negative, since the frequency seen by the moving molecules tends to agree with the molecular resonance frequency. For operation near saturation or for a diverging beam, the shift can be of either sign. Mockler has built a double beam maser at Boulder with beams coming in from each end in order to reduce the Doppler shift. He finds that shifts as high as 2 parts in 10^9 can occur when one beam is turned off.[11] However, this experiment needs to be repeated with nitrogen-15 ammonia and with the two beams symmetrized as much as possible. If this is done at several laboratories, with magnetic modulation to adjust the cavity tuning, it seems quite possible that an accuracy of several parts in 10^{11} can be achieved.

A maser, of course, has the advantage that a quartz crystal oscillator is not needed as a flywheel, and that the very short term stability can thus be quite high. Minute-to-minute stabilities of one or two parts in 10^{12} have been demonstrated for two masers beating against each other. For this reason, Mockler has used an ammonia maser to check on the spectral purity of quartz crystal oscillator and frequency multiplier chain outputs. It has also been suggested that it may be necessary to use masers to drive long cesium tubes in order to obtain sufficient spectral purity for use with very narrow resonance lines.

APPLICATIONS

The types of atomic frequency standards which have been discussed actually complement each other quite well. While the

long cesium beam tube appears to be the best hope at present for high accuracy, the ammonia maser is the most suitable for short term stability and the alkali vapor devices are promising as portable secondary standards. Which type of standard is most desirable will depend on the intended application.

Recently there has been a demand for rugged frequency standards for use in missile guidance systems. Here the desired stability is of the order of 1 part in 10^{10} for about .1 second averaging times, but the frequency needs to be uniform only for the several minutes of launching. Two types of short cesium beam tubes are being built for this purpose, as well as rubidium vapor devices and ruggedized ammonia masers. For the passive devices, the philosophy has been to broaden the resonance lines in order to improve the signal-to-noise ratio and to permit the use of a short time constant in the feedback loop.

For the gravitational frequency shift experiment, there has not yet been a choice of which type of device will be used. Precision, reliability, and power requirements will be the most crucial factors here, although weight and size will also be important. It has been suggested recently that the experiment could also be done by using a quartz crystal oscillator in a satellite with a highly elliptical orbit.[13] A transponder with provision for frequency subtraction in the satellite could be used to cancel out the first order Doppler shift. The expected shifts should then show up as periodic differences between the frequencies near apogee and near perigee. Accuracies of a few percent in checking the gravitational frequency shift seem possible in the near future with several of the proposed methods.

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DISCUSSION

S. HOPFER: It has been mentioned in the paper read that the effect of the existing traveling waves in the maser cavity upon the frequency of oscillation may be sharply reduced by employing a balanced dual beam of molecules traversing the cavity in opposite directions.

Experience shows, however, that a balanced beam is difficult to realize and results in a rather bulky system. It seems possible to reduce greatly the Doppler effect by employing a dual cavity system coupled not at a single point as shown by Bonanomi but over the entire length of the cavity. Practically, this is accomplished by a series of small irises along the cavity. In this way, the real part of the Poynting vector at any point in the cavity should be primarily normal to the axis of the cylindrical cavity and should thus produce no Doppler effect if the molecular velocity vector is primarily axial.

F. H. REDER: Higher accuracy of an atomic beam frequency standard may preferably be obtained through using slower atoms rather than a longer beam.

Extension of the beam length makes it increasingly difficult to maintain phase coherence in the two Ramsey cavities, magnetic field homogeneity in the C region, and the proper high vacuum along the beam axis. Also the beam intensity seen by the detector will decrease somewhat with length.

Consequently, the use of slower atoms in beam tubes of the present length seems to be a solution worth investigation. It would certainly be more convenient to use in practical applications. It should be noted that the National Company has obtained a 60 cps bandwidth by selection of slow atoms in a 6-foot beam tube. (Normal bandwidth: 120 cps.)

J. BONANOMI: This comment is on the resetability of atomic clocks over long periods of time as achieved in practice. DePrins and I have determined since March 1957 through August 1959 an atomic time scale, which we called TA_1, by means of ammonia masers. Markowitz at the Naval Observatory defined another time scale, A_1 based on the frequency of one Atomichron. We have compared these two time scales by comparing the reception times of WWV time signals in terms of TA_1 and A_1: this was done regularly twice a month. During the stated period of 2.5 years, the difference $TA_1 - A_1$ remained constant within $a \pm 2.5$ msec, indicating that any systematic drift of NH_3-frequency at our laboratory from Cs-frequency at Washington has been smaller than 10^{-10} during this period.