HYPERFINE TRANSITIONS IN RUBIDIUM-87 VAPOR

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SUMMARY

Microwave resonance in buffer vapors of rubidium-87 and cesium-133 have been studied by optical techniques. Many of the factors which affect the center frequency and the width of the resonance have been investigated, particularly the dependence of these parameters on the kind, density, and temperature of the buffer gas. In addition, an attempt is being made to de= termine the magnitude of time=dependent shifts resulting from evolution of gas from the glass walls.

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At the last Frequency Control Symposium we described an optical means for observing hyperfine resonances in cesium, and reported the results of experiments on the shifts of frequency associated with various experimental parameters. Since then, similar experiments have been done with rubidium-87. The method used has been described elsewhere.[1] One of the principal conclusions from the further experimentation is that, as predicted earlier by Dicke and Carver, rubidium appears to be more satisfactory for gas cell frequency standards than cesium. Signal-to-noise ratios obtained with rubidium are considerably higher than have been found with cesium. Although the rubidium-87 frequency is 25 percent lower, the minimum linewidth found is about a factor of 2 less, giving a higher Q.

An adequate physical understanding of the processes which cause the linewidths and observed frequency shifts is not yet available. Such an understanding will be necessary to make maximum use of the gas cell method; nevertheless, some data are available which can be used to evaluate the usefulness of the gas cell as a frequency standard, even in the absence of a complete understanding. First consideration will be given to effects which affect the resonance frequency.

It has been found that the frequency of a gas cell depends upon the amount and kind of buffer gas. Also, a sealed cell has its frequency somewhat dependent upon the temperature. Measurements of these effects have already been reported for cesium.[2] With rubidium the effects are qualitatively the same, and are illustrated in Fig. 1.

The formula is expressed in terms of measured parameters and would look somewhat simpler if one used the fact that for a sealed cell the pressure is proportional to the temperature. α_p is the rate of change of frequency with buffer gas pressure, with the temperature held constant at 300°K. α_T is the rate of change of frequency with buffer gas temperature with the volume and density held constant. There is no reason to believe that this formula is valid except near room temperature. Actually, the detection sensitivity is poor for large departures from room temperature anyway. The units of pressure are cm of Hg, and the pressure range which makes the detection most convenient is from about 1 cm to 10 cm.

Some of the coefficients are positive and some are negative. Also, α_T is not proportional to α_p. In the cases of mixtures that have been tried, the resultant coefficients are just the weighted means of the coefficients of the constituents. Coefficients of two mixtures are indicated to show that it is possible. Note that if one is willing to regulate the temperature to 0.1°C, it would be easy to use the 88 percent argon mixture and reproducibly create cells to a precision of 1 part in 10^{10}. Alternately, if a temperature insensitive mixture is used, the thermal control problem is simpler, but more effort is required to produce samples agreeing to 1 part in 10^{10}.

Mixtures can be made which will make both coefficients small. For instance, about:

30% Kr	or	35% Kr	
40% A		25% A	
30% He		40% N_2.	

However, it is questionable whether high signal-to-noise ratios can be obtained with such mixtures.

If we assume that the shift for an average impurity is 1 part in 10^7 per mm-Hg, maintenance of the frequency to 1 in 10^{11} requires keeping the impurity pressure down to 10^{-4} mm of Hg. This is approximately the residual pressure in vacuum tubes. Employing modern vacuum technology it should be possible to do better than this for several years of operation. Literature on ultra-high vacuum technology indicates that for very clean systems the principal contaminant is helium which diffuses through the glass wall. This leak is expected to cause an aging effect on the frequency of less than 1 part in 10^{11} per year at 40°C. For helium buffer gas the aging would be a serious nuisance, unless the diffusion through the glass were stopped by some means. We have little data on the rates of disappearance of other gases but they are expected to be much smaller.

In order to check on aging, three samples were filled in September, 1958, with a 50 percent A-50 percent Ne mixture at a pressure of 1 cm of Hg. No unusual precautions were taken in the filling process. The temperature coefficient was 3 parts in 10^{11} per degree C. The frequencies obtained with these samples were checked a number of times at NRL within two months after filling and have been checked again in May, 1959, at NBS. One of the three samples was heated for extended periods between measurements at NRL. The unheated samples have maintained the same relative frequencies over this 8-month period to within the measuring accuracy of 1 or 2 parts in 10^{10}. The heated sample survived three days at 100°C, three days at 150°C, and one week at 200°C without significant change. However, an additional two weeks at 200°C caused an increase in resonance frequency of about 4 parts in 10^{10}. This sample then retained the new frequency relative to the other two samples for the remaining 6 months of the measurements. The recent measurements at NBS gave results, in terms of Atomichron No. 110, which were apparently 4 parts in 10^{10} higher than the NRL values obtained 6 to 8 months earlier in terms of this standard. The comparison was made via WWV on a single day and the difference is possibly not significant.

The values for f_0, obtained by extrapolating to zero buffer gas pressure are, on the A.1 time scale:[3]   Rubidium 6,834,682,608 ±7 sec^{-1}
Cesium   9,192,631,735 ±10 sec^{-1}

Changing the light intensity and microwave power by a factor of 4, produced no noticeable change in frequency. The extrapolated frequency obtained with the cesium gas cells disagrees with that obtained from the Atomichron by 30 or 40 cps. We have no explanation of the discrepancy but certainly agree that the beam result is the one which is characteristic of a free atom. Impurities in the cells could cause the shift; however, it seems unlikely that the amount of impurity can be so reproducible. This is particularly true in view of the fact that some of the cells were prepared on a very good vacuum system.

Although the amount of microwave power at the sample and the conditions in the lamp apparently do not affect the frequency, they do, in general, affect the linewidth and detection sensitivity. Also, the linewidth and sensitivity can be affected by the amount and kind of buffer gas. Our measurements have indicated that neon and helium buffer gases permit greater sensitivity than other gases, with both cesium and rubidium. For low fixed-light intensity and microwave power level, neon and helium give stronger signals, longer relaxation times, and narrower resonance lines than the other gases. Argon and heavier gases increase the width somewhat in rubidium, and by a factor of 3 or more in cesium.

In rubidium and cesium, with most buffer gases, the line width is surprisingly independent of buffer gas pressure over a fairly large range of pressures. The minimum linewidth obtained was about twenty cycles per second for rubidium in neon and helium at pressures of 1 to 10 cm. For the Zeeman resonances of the same cells, in the earth's field, the observed linewidths were only slightly smaller in this pressure range. At low light and microwave power levels the linewidths do not depend on these parameters. No broadening due to alkali-alkali collisions was observed up to $40^{\circ}C$ for rubidium and $30^{\circ}C$ for cesium. Experiments on the recovery of the samples from saturation of the hyperfine resonance showed that the spin-lattice relaxation time T_1 was considerably longer than the spin-spin relaxation time T_2 indicated by the linewidth of the Zeeman resonance. The source of the pressure-independent line width is not known. However, it seems likely that an explanation of it would also lead to an explanation of the difference between the cesium cell hyperfine frequency and the cesium beam value.

Most of the data on rubidium were taken at NRL using a frequency source supplied by the Radio Techniques Branch. A synthesized 2278 mc frequency was tripled in a crystal harmonic generator and applied to the cell by a horn. The applied frequency was known in terms of cesium to one or two parts in 10^{10}.

Apparatus now set up at NBS uses a direct multiplication system. A quartz crystal oscillator at .999223 mc with a rather high crystal drive is multiplied to 360 mc. Crystal diodes are used for harmonic generation and tubes are used as amplifiers. A varactor is used to generate the 19th harmonic at 6835 mc. The output is applied to the cell by a horn.

Figure 2 shows a resonance line obtained with one of the 50 percent A-50 percent Ne cells at 1 cm pressure. The time constant of the detector was .02 seconds and the signal-to-noise ratio about 60. The horizontal frequency scale is 3 parts in 10^9 per cm. The time of the sweep was about 15 seconds. The two ends of the trace were not the same because of a slow drift in the background light intensity. On the sides of the resonance, frequency noise in the oscillator or multipliers as big as 1 part in 10^{10} would have caused a noise about equal to the noise off resonance. The line was considerably broadened by excessive light and microwave power to about 40 cycles per second. Signals have been obtained using a Rb^{85} lamp instead of the Rb^{85} filter which we normally use, but the signal-to-noise ratio was lower.

A direct frequency multiplication such as we are using is desirable for a simple and accurate standard. When even output frequencies such as 1 or 5 mc are needed, a frequency translator must be used. In this respect the gas cell frequency shifts are a help in that it is possible to choose the pressure and temperature so that the resultant frequency is easy to translate. Figure 3 shows a block diagram of one proposed scheme. It contains an oscillator at 1 mc and a means of generating 9/386 of the oscillator frequency. A simple system can then be used to phase-lock the oscillator to the rubidium standard.

The main reason for the interest at NBS in gas cell frequency standards has been the desire to use them in an accurate check of the gravitational frequency shift, predicted by the General Theory of Relativity. The use of an atomic clock in an earth satellite for this experiment has already been discussed by S. F. Singer and others. Suitable prototype rubidium vajor clocks for this purpose are now being developed at NBS with the aid of Varian Associates. The work is being done under the sponsorship of the National Aeronautics and Space administration. The power required for a prototype will be about 10 watts. The weight of the apparatus without the power supply will be less than 20 pounds. The stability of one clock with respect to another clock of the same type is expected to be 1 part in 10^{11} over a period of at least one month.

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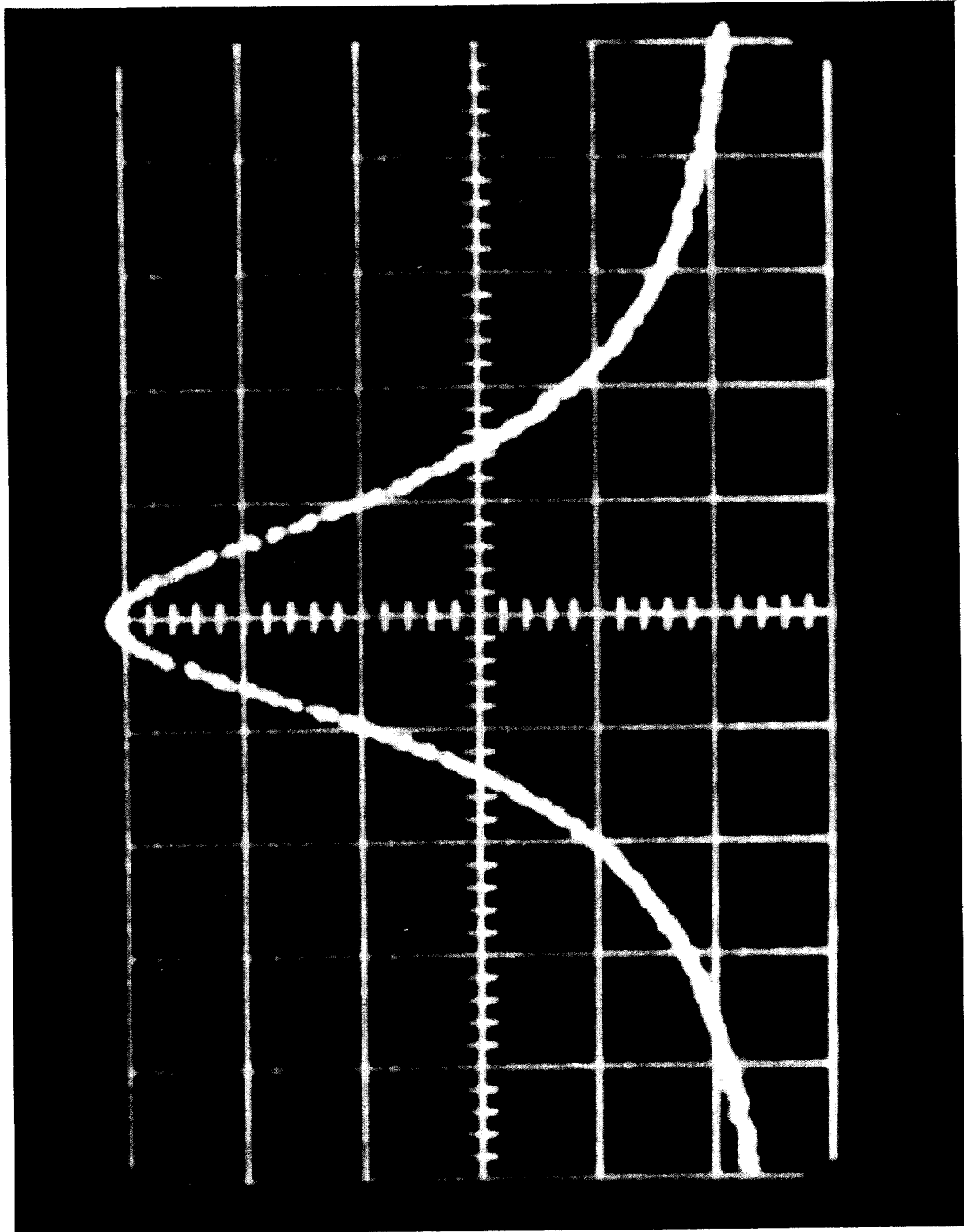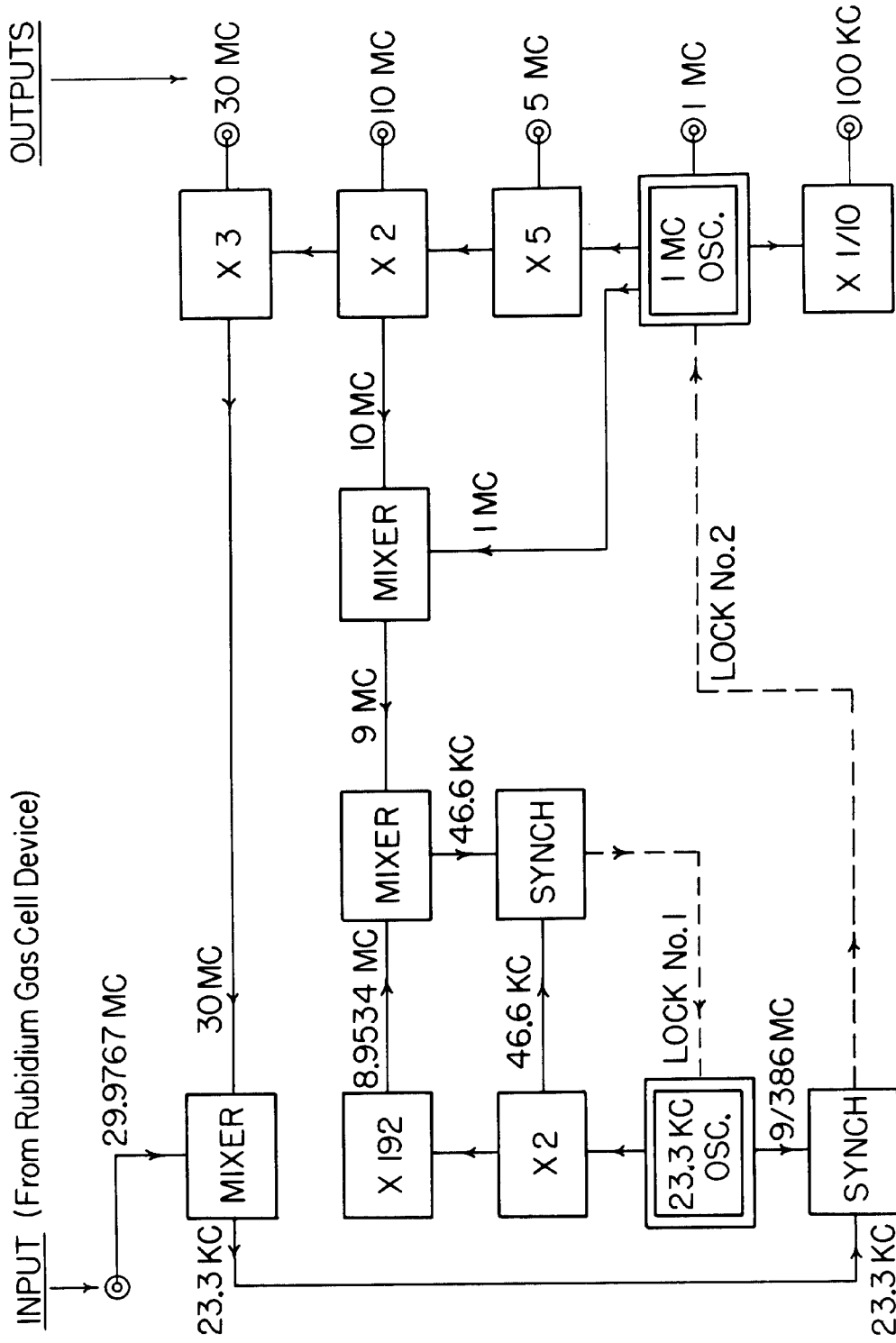
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References

(1) P. L. Bender, E. C. Beaty, and A. R. Chi, Phys. Rev. Lett. 1, No. 9, (1958).

(2) E. C. Beaty, P. L. Bender, and A. R. Chi, Phys. Rev. 112, 450 (1958).

(3) W. M. Markowitz, Proc. 13th Annual Frequency Control Symposium, May, 1959.

Buffer gas	$10^{10} \times \alpha_P \left[\dfrac{1}{cm\text{-}Hg}\right]$	$10^{10} \times \alpha_T \left[\dfrac{1}{(cm\text{-}Hg)(°K)}\right]$
hydrogen	+9,700 ±200	+15
deuterium	+9,800 ±200	+15
helium	+10,500 ±200	+15
neon	+5,700 ±100	+ 3.8
nitrogen	+7,600 ±150	+ 9
argon	- 750 ±15	- 4.4
krypton	-8,500 ±200	- 7
11.7% Ne-88.3% Ar	+ 18 ±5	- 3.5
50% Ne-50% Ar	-2,500 ±50	- 0.3

$$f = f_o \left\{ 1 + \alpha_P \left[\frac{300\ P}{T}\right] + \alpha_T (T - 300) \left[\frac{300\ P}{T}\right] \right\}$$

Fig. 1. Dependence of the hyperfine resonance frequency in rubidium-87 vapor on the pressure, P(cm-Hg) and temperature, T(°K). The formula is expected to be valid only near room temperature, 300°K.

OUTPUTS ⟶

INPUT (From Rubidium Gas Cell Device)

29.9767 MC

30 MC

23.3 KC

MIXER

8.9534 MC

X 192

46.6 KC

X 2

23.3 KC
OSC.

LOCK No.1

9/386 MC

SYNCH

23.3 KC

MIXER

9 MC

46.6 KC

SYNCH

LOCK No.2

MIXER

1 MC

10 MC

X 3 — ◎ 30 MC

X 2 — ◎ 10 MC

X 5 — ◎ 5 MC

1 MC OSC. — ◎ 1 MC

X 1/10 — ◎ 100 KC

Frequency Translator for Rubidium Gas Cells

29.9767 MC input is from multiplier chain locking
4.99614 MC oscillator to Rb^{87} cell containing about
7mm-Hg of 47% Ar-53% Ne mixture.

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