A Microwave Frequency Standard Employing Optically Pumped Sodium Vapor*

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Summary—An instrument in which a simple microwave triode oscillator is stabilized by reference to a natural atomic resonance—the field-independent hyperfine resonance of sodium—is described. Light from a sodium lamp is transmitted through an absorption cell containing sodium vapor and argon, which is placed in a resonant cavity. This light produces population differences between the two quantum levels which are involved in the desired atomic resonance and provides a means of detecting resonance. The cavity is excited by an external microwave triode oscillator which is frequency modulated to a small degree at 60 cycles. When the exciting oscillator frequency coincides with the center of the atomic resonance line, the signal observed by a photocell will be a modulation of the transmitted light at 120 cycles and higher even-order harmonics. Any deviation from line center will introduce a 60-cycle component whose phase and magnitude may be detected to produce an error signal to retune the oscillator in the usual servo loop manner. Theory predicts that an accuracy of possibly one part in 10^{10} can be achieved by systems using sodium and suitable local oscillators. It is evident also that such systems can be engineered into quite small packages, making possible many new applications of microwave oscillators stabilized to high order.

AN instrument is described in which a simple microwave triode oscillator is stabilized by reference to a natural atomic resonance—the field-independent hyperfine resonance of sodium. Although atomic resonances of this type have been used to produce primary frequency standards of hitherto unobtainable stability, it is not our intent to demonstrate such an instrument here. What we have built is a compact and simple microwave oscillator which has a short-time stability of about 1 part in 10^7 and a long-time stability of 1 part in 10^8 or better. As such, it represents an improvement by at least a factor of ten over previous simple microwave oscillators in this frequency range. The limiting factors determining the stability of this system have been shown to reside in the oscillator, rather than in the stabilization effected by the atomic resonance. It is clear that greater stability could be achieved by use of a microwave source with greater inherent stability, for example, a quartz crystal with multipliers. Such a system would not have the simplicity and compactness of the system presented here.

The over-all system is portrayed in the block schematic diagram of Fig. 1. Light from a sodium lamp is transmitted through an absorption cell containing sodium vapor and argon which is placed in a resonant cavity. The light produces population differences between the two quantum levels which are involved in the desired atomic resonance, and also provides a means of indication of the existence of resonance.[1] The cavity is excited by an external microwave triode oscillator which is frequency modulated to a small degree at 60 cycles. When the exciting oscillator frequency coincides with the center of the atomic resonance line, the signal observed by the photocell will be a modulation of the transmitted light at 120 cycles and higher even-order harmonics. Any deviation from line center will introduce a 60-cycle component whose phase and magnitude may be detected to produce an error signal to retune the oscillator in the usual servo loop manner.

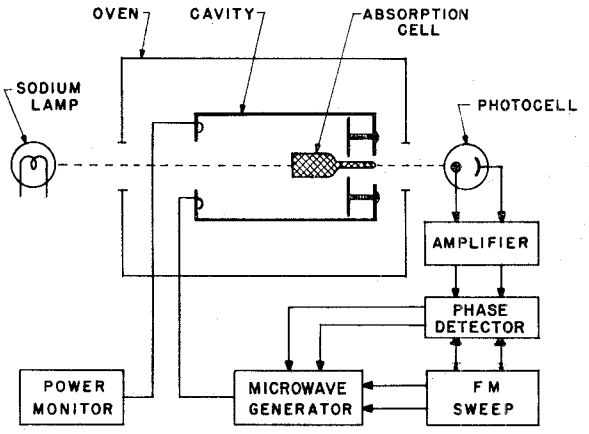

Fig. 1—Sodium vapor stable oscillator, block diagram.

General

A number of methods by which irradiation of a vapor by light can be used to provide population differences (optical pumping) has been described in the literature.[2–4] The one employed here is probably one of the simplest and depends on the fact that the presence of a relatively high pressure of buffer gas tends to disorient the spins of the atoms when they are in the optically excited states (but not in the ground states which are used in the radio-frequency resonance). This may be taken to imply that, although the various sublevels of the ground

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[1] W. Bell and A. Bloom, "Optically detected field-independent transition in sodium vapor," *Phys. Rev.*, vol. 109, pp. 219–220; 1958.
[2] A Kastler, "Optical methods of atomic orientation and of magnetic resonance," *J. Opt. Soc. Amer.*, vol. 47, pp. 460–465; 1957.
[3] H. G. Dehmelt, "Slow spin relaxation of optically polarized sodium atoms," *Phys. Rev.*, vol. 105, pp. 1487–1489; March, 1957.
[4] J. P. Wittke, "Molecular amplification and generation of microwaves," PROC. IRE, vol. 45, pp. 291–316; March, 1957.

state may have different probabilities for absorbing light, they all have the same probability for receiving atoms which are reradiating light and returning from the excited states.[5] Steady-state conditions obviously require that the number of atoms leaving a given ground state sublevel must equal the number returning per unit time, so that one can make the following statement. If a_i is the number of atoms in the ith sublevel and P_i is the number of times per second each atom in the ith sublevel absorbs light, then the steady state condition is $a_i \propto 1/P_i$. In addition, we have the requirement that the total population is a constant. It is convenient to select this constant equal to one. This condition, plus the one given above, determines uniquely the populations of the various sublevels of the ground state provided only that we also know the rate, P_i, at which atoms absorb light in the various sublevels.

Most of the light which is emitted by a sodium lamp and which is absorbed by cold sodium atoms lies within the so-called D lines of sodium, the familiar yellow light of a sodium lamp. The difference between the D_1 and D_2 lines is of no importance here. However, what is important is the fact that each of these lines is in turn split into two closely spaced but completely separate hyperfine components. Fig. 2 shows part of the energy level diagram for the sodium atom, not drawn to scale. The difference between the two lines in a pair of hyperfine components is that one of them is capable of exciting atoms out of the $F=1$ state while the other is capable of exciting them out of the $F=2$ state. These frequencies are quite well separated as to function, since the naturally occurring width of an optical spectrum line of these wavelengths is of the order of several hundred megacycles due to Doppler broadening. This is considerably less than the 1772-mc separation of the energy levels. If the populations of all eight magnetic sublevels shown in Fig. 2 were equal, then it is clear that light capable of exciting atoms out of the $F=2$ level would be attenuated 5/3 as fast as the light exciting atoms out of the $F=1$ level, since there are 5 atoms in $F=2$ for every three atoms in $F=1$.

We are now in a position to describe the light absorption on a quantitative basis. Let us choose x as a normalized distance through the absorption cell; in practice, x is a function of sodium vapor density. Let us assume that we can neglect other effects besides the optical pumping which might contribute to the final population distribution. Then the intensity of the light as a function of distance through the absorption cell can be described by the following system of equations:

$$\begin{aligned} 3a_1 + 5a_2 &= 1 \\ dI_1/dx &= -3a_1I_1 \\ dI_2/dx &= -5a_2I_2 \\ a_1I_1 &= a_2I_2 \end{aligned}$$

[5] W. Bell and A. Bloom, "Optical detection of magnetic resonance in alkali metal vapor," *Phys. Rev.*, vol. 107, pp. 1559–1565; 1957.

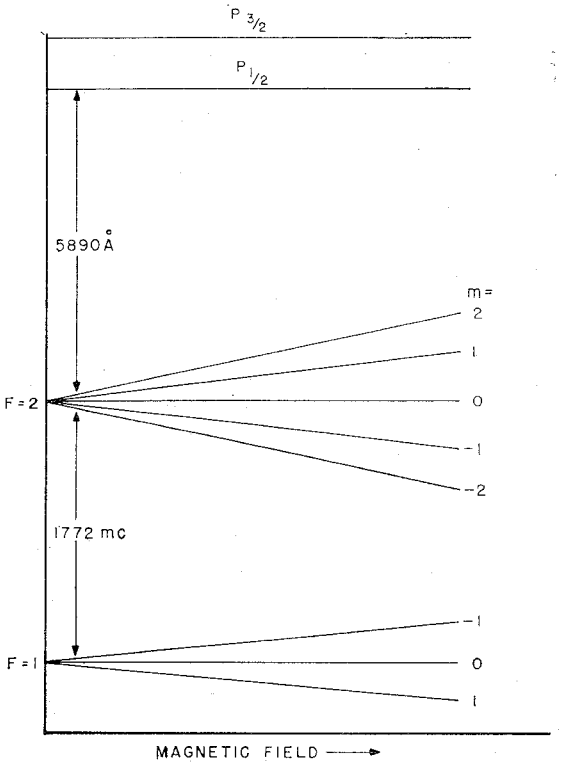

Fig. 2—Sodium atom energy levels diagram.

where we have implicitly substituted I_i for P_i. This is permissible because of their proportionality, which in turn holds because the transition probability per unit time for atoms in all sublevels are equal (assuming unpolarized light). This system of equations has an exact solution as follows, assuming $I_1=I_2=1$ at $x=0$,

$$I_1 = \frac{1}{5}\left(1 + \sqrt{1 + 15e^{-x}}\right)$$

$$I_2 = \frac{1}{3}\left(-1 + \sqrt{1 + 15e^{-x}}\right).$$

These curves have been plotted in Fig. 3 together with the sum I_1+I_2, which is the light intensity as seen by the photocell when the light path through the absorption cell is x. On the other hand, if by some external agent we constrain the populations of all of the eight magnetic sublevels to be equal, then the light intensity would be given by $e^{-3x/8}+e^{-5x/8}$, which is indicated by the dashed line in Fig. 3. The actual reduction in light intensity by equalizing a particular pair of levels, such as the two $M=0$ levels, is probably about one fourth of that indicated. Thus, for example, at $x=3$ one might expect the change in light intensity due to the zero-zero resonance to be something of the order of 4 per cent of the total remaining light intensity. With known light sources, a reduction of intensity equal to that reached at $x=3$ is probably the greatest that can be expected before other effects such as thermal relaxation become important in determining population differences.

In the actual system, the radio frequency was swept

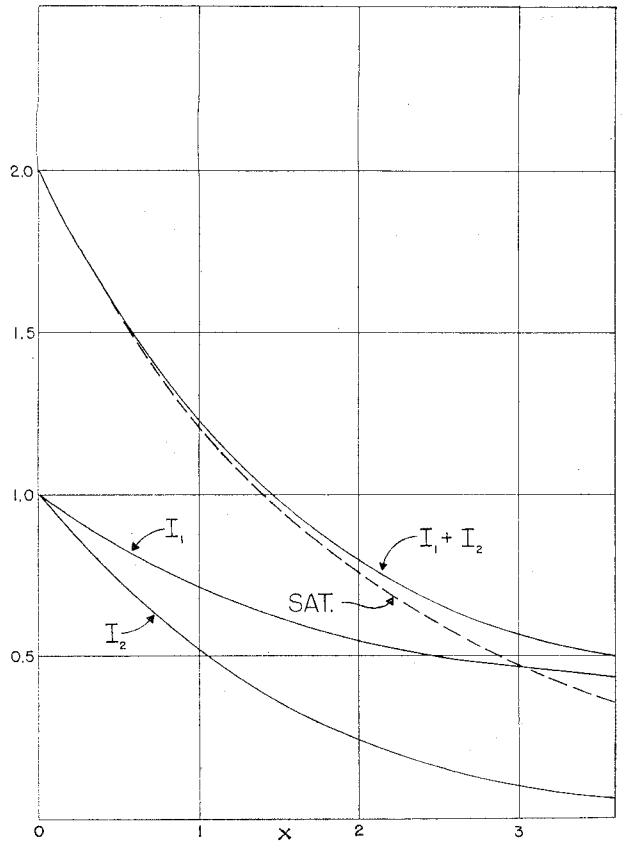

Fig. 3—Light intensity vs total absorption through the gas cell.

back and forth through the resonance at 60 cycles. The type of signal observed in this apparatus, which observes population differences rather than oscillating components of magnetization, is somewhat different from the resonance signals normally encountered. The process of producing a signal may be outlined as follows. As the frequency is swept exactly through resonance, the transient RF coupling to the spins rapidly inverts them,[6] *i.e.*, shifts the population difference. During the remaining half cycle or so when the RF is not exactly at resonance, the spins slowly return to population equilibrium with the pumping light, resulting in a relatively slow recovery of light intensity as observed by the photocell. At the end of the half cycle, the spins are back to equilibrium and ready for another inversion. The important thing which must be understood is that the "width" of the signal as observed in this system is not really a function of natural line width but is merely a function of the manner in which the sweep field operates. For this reason, in order to take full advantage of the relatively narrow line widths of these systems, it is necessary to operate with a quite narrow sweep and weak RF. However, quite satisfactory results also can be obtained with a wider sweep and more RF. The result is effectively a line with somewhat better signal-to-noise ratio and greater line width, and the system as a whole has a lower stabilization.

[6] F. Bloch, "Nuclear induction," *Phys. Rev.*, vol. 70, pp. 460–471; 1946.

Electronics

The oscillator is a conventional microwave triode of the lighthouse type, with cavities tunable about the desired frequency of resonance, 1772 mc. A fine frequency control is provided by a small variation of the plate supply voltage applied to the tube, which simultaneously yields a convenient method for frequency modulating the oscillator. Since the index of modulation needed is so small, no more than 1 part in 10^5, it was merely necessary to superimpose a small 60-cycle ripple on the output of the regulated power supply to sweep the oscillator through the sodium resonance.

The oscillator is loosely coupled to the cavity containing the absorption cell and provides the radiation necessary to excite the sodium hyperfine transition. The cavity is a right circular cylinder resonating in the TE_{011} mode with coaxial holes at each end to allow the light to pass in and out. The absorption cell is inside the cavity and is arranged to intercept the light beam. For a cavity in the TE_{011} mode, the magnetic lines of force of the RF field are parallel to the axis except near the ends where there are some components perpendicular to the axis. To excite the field independent transition, it is necessary that the RF field be parallel to any existing dc field such as the earth's magnetic field. By placing the absorption cell near one end of the cavity where components of the magnetic field in all directions are present, it was possible to achieve a reasonable amount of directional independence, so that the apparatus as a whole did not have to be oriented in one particular direction to give good signals.

The Q of the cavity used in optical detection does not have to be particularly high, since it is light absorption rather than electromagnetic radiation which is being detected. However, a high Q is useful in that it reduces the amount of power which is required from the oscillator to excite the cavity. The configuration employed allows a reasonably high Q (over 10,000) even though the holes at the ends are large enough to allow a beam of light about two inches in diameter to pass through. The absorption cell is a fused quartz cylinder two inches in diameter and three inches long. Quartz is used because of the reduced losses at these frequencies and thereby it does not appreciably perturb the field configuration of the cavity. The cell contains pure metallic sodium deposited at one end in a manner permitting a sufficient area to be transparent to the sodium light. The buffer gas is spectral grade argon at a pressure of about 10 centimeters of Hg, and the cell is operated at a temperature of about 135°C, which provides a vapor pressure for the sodium of about 10^{-5} millimeters of Hg.

The sodium lamp supplying the pumping light is of the electrodeless discharge type. A small glass tube containing the sodium metal with argon at about 1-mm pressure is placed in the tank coil of a self-excited power oscillator running at about 50 mc. This field supplies power to heat the sodium metal to produce vapor and

at the same time excites the vapor and causes the characteristic sodium light to be emitted. It was found that if the light from the lamp was very bright, the gas cell polarization would be quite poor. This could be explained by a broadening of the sodium light spectral line because of too high a vapor pressure. Time did not permit a complete examination of phenomena connected with the lamp, so that for purposes of our experiments we determined a particular set of lamp operating conditions (power dissipation in the oscillator) for which lamp operation was satisfactory. As long as these conditions were not varied drastically, stable operation of the lamp resulted for long periods of time.

The type 6570 photocell used to monitor the light intensity passing through the absorption cell has a surface of S1 material, which has a relative efficiency of about 50 per cent for sodium light. The signal resulting from optical detection of the resonance has a signal-to-noise ratio of 10 or more, with a photocell amplifier bandpass of the order of 10^3 cycles. A photograph (1 or 2 sweep cycles) of the scope presentation is shown in Fig. 4. The vertical signal is the photocell output plotted against the 60-cycle horizontal sweep also used as the phase detector reference.

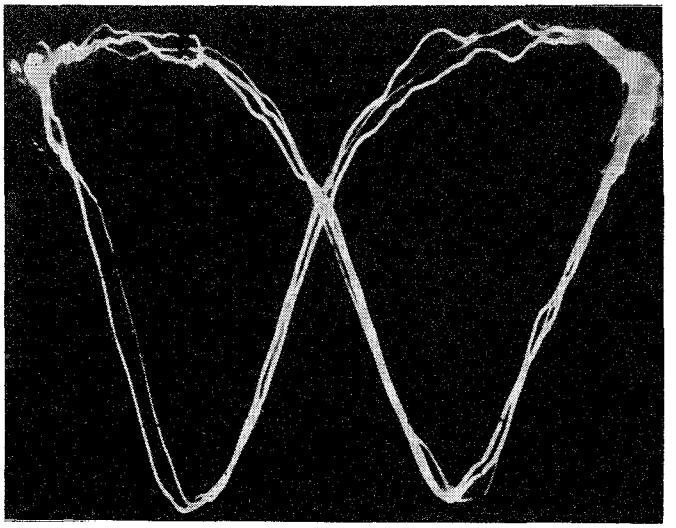

Fig. 4—Optically detected signals from sodium resonance.

Results

Time did not permit a complete examination of the parameters of this particular system; however, one particular set of operating conditions will be described. To evaluate the accuracy to which the frequency of the microwave oscillator triode is being controlled, it is necessary to measure the sweep width so that excursions of resonance within the sweep can be determined. It is fortuitous that the hyperfine structure of the sodium atom itself provides a convenient sweep calibration for these purposes, so that no additional apparatus is needed to make the calibration. If one shifts the frequency of the oscillator by about 350 kc, one can find two other resonances from the hyperfine structure, in particular, those from $F=2$, $m=0$ to $F=1$, $m=-1$ and from $F=2$, $m=-1$ to $F=1$, $m=0$. These resonances are almost coincident, and, as a matter of fact, it can be shown by theory that the separation of the resonances is almost exactly 1155 cycles in the laboratory magnetic field. Thus if the frequency modulation of the oscillator is of the order of several thousand cycles, these two resonances will appear as two separate and distinct peaks on the oscilloscope trace. Their separation relative to the total sweep can then be used to calibrate the sweep.

We operated the system with a sweep width of about 3000 cycles. Under this condition the system was quite stable, and we were able to operate for a total of about 100 hours without any major adjustments to the system. During this time, the crossover point of the oscilloscope signal remained within a factor of 10 per cent at the center of the trace. On this basis, it is clear that the oscillator had a short time stability of the order of 2 parts in 10^7 for periods corresponding to a few tenths of a second. The long-time stability, of course, was much greater than this, since at no time during a period of about 100 hours did the signal wander out of the dynamic range of the servoloop. When we attempted to narrow the sweep, the long-time stability was as good as or better than that given above. But on a short-time basis, it was observed that there were oscillations of the order of 1 cycle per second which could not be controlled by the existing time constants of the feedback loop. These oscillations are believed to be due to a hysteresis effect in the microwave triode, resulting in second derivative terms which cannot be controlled by a feedback loop consisting of simple RC time constants. Time did not permit a complete analysis of this problem. However, it is clear that a more sophisticated servomechanism controlling the feedback would enable us to control this oscillation, thus allowing one to go to much narrower sweeps and consequently much higher degrees of stability.

Theory predicts that an accuracy of possibly 1 part in 10^{10} can be achieved by systems using sodium and suitable local oscillators. It is also evident that such systems can be engineered into quite small packages, making possible many new applications of microwave oscillators stabilized to high order.