

ATOMIC FREQUENCY STANDARDS: SURVEY AND FORECAST

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The present paper is a concise survey giving the characteristics (mainly stability and accuracy) of state of the art Atomic Frequency Standards. A modest attempt is made at a projection in the future on improvements that could be accomplished with major research and development efforts. The reader will find well documented information on Atomic Frequency Standards in several review papers which have been published within the last ten years (1-7). The present paper is based on some of these articles, and owes much to some other publications more closely related to the question of forecast (8-10). It is also based on private communications with various scientists working actively in this field.

The Basis of Atomic Frequency Standards.

Basic concepts

An atomic frequency standard is a frequency generator in which the essential harmonic motion is produced by an atomic resonance. This resonance originates in a transition between two specific energy levels of the atom in question. The resonance frequency is given by

$$v_0 = \frac{E_2 - E_1}{h} \quad (1)$$

where E_2 and E_1 are the energy of the two levels involved and h is Planck's constant. The main reasons behind the idea of using such an atomic resonance for producing an harmonic motion are:

a) The energy difference $\Delta E = E_2 - E_1$ giving rise to v_0 is primarily a function of fields internal to the atoms and, within some specific conditions and limits, is only slightly affected by external fields or perturbations. Furthermore these perturbations in many cases can be well controlled and calculated, and corrections can be applied to the frequency observed experimentally. When the corrections are well known, one can retrace back from the frequency measured to the actual resonance frequency v_0 of the free atom at rest. This consideration is related to the accuracy of the particular standard. Here accuracy is defined as the cumulative uncertainty on the frequency corrections which must be applied to the observed frequency in order to obtain the transition frequency of the free atom at rest, divided by the nominal frequency. In practice it amounts to a measure of inaccuracy. The smallest numbers give the highest accuracy.

b) With some ensembles of atoms it is possible to obtain very narrow resonance lines. This is in fact related to the actual life time of the atoms in a particular state. The longer the lifetime of the atoms, the narrower the resonance is and one defines a line quality factor as

$$Q_L = \frac{v_0}{\Delta v} \quad (2)$$

where Δv is the width of resonance line. Although, in principle the lifetime of atoms in a given state may be years, in practice the simple process of probing the resonance limits this lifetime and a compromise must be made between signal amplitude and linewidth. Furthermore, as

will be discussed below, other perturbations which broaden the line are always present in actual devices. However, the line Q's possible in practice for some atoms are still much higher than those obtained in the best standard mechanical devices such as quartz crystals used in precision oscillators. The line quality factor relates to the precision with which the line center can be determined experimentally. The line quality factor relates also to the stability of the frequency standard. Here frequency stability is a measure of the degree to which the frequency will stay constant after the oscillator has been set in operation. In the present article we will characterize the stability either through the spectral density $S_y(f)$ of fractional frequency fluctuations or $\sigma_y(\tau)$, the two sample variance. (11)

c) It is also believed that $v_0 = \frac{E_2 - E_1}{h}$, for a given pair of levels in a specific atom, is a constant independent of time. This is in contrast with the actual rotation rate of the earth which is known to fluctuate and quartz crystals whose frequency is known to change with time. The idea is fundamental and drifts observed in actual frequency standards are assumed at present to be entirely inherent to the practical realization of the standard. The second is currently defined in terms of a particular transition in cesium, whose frequency is 9, 192, 631, 770 Hz.

Practical realization.

In order to realize an actual device based on the concepts discussed above, several practical considerations must be examined. Those considerations actually dictate the choice of atoms which is of course a main point in the whole discussion.

The problem can be best situated with a block diagram showing the two possible approaches. In general one wants an oscillator

at a given nominal frequency of, say, 10 MHz to represent as well as possible the characteristics of the atomic resonance line. It thus amounts to a question of locking a frequency generator, generally a quartz oscillator to the atomic resonance line. This can be done in the two modes illustrated in figure 1. In the active mode, the atomic ensemble, well prepared in a given state, emits energy at its resonance frequency. The quartz oscillator is locked in phase to the coherent signal emitted by the atomic ensemble; one has what is called a phase lock loop. In the passive mode, the ensemble of atoms also well prepared in a given state is questionned or interrogated with a signal derived from the crystal oscillator as to what its frequency is; we then have a frequency lock loop. Figures 2 and 3 give more details on the two approaches.

From figure 1, it is seen that one must consider as essential processes: a) the choice of the atomic ensemble, b) its storage, c) its preparation, d) its interrogation and e) the detection of the atomic signal. These points have been discussed in some details by several authors in various ways. (9, 10). It is clear that the processes are not completely independent and that interaction can exist between them. For example the choice of the atomic ensemble is dictated by practical considerations such as obtaining a decent vapor pressure at a moderate temperature, the possibility of storing it in some enclosure or forming a beam of these atoms.

a) Choice of the atomic ensembles: As a general rule and added to constraints of practicality, the AE of the atomic ensemble chosen should be affected as little as possible by external force fields and by the experimental set up used to detect the resonance. Another consideration, of course, is also that the line width should not be affected to a too great extent by the same mechanisms used to detect the resonance.

For the purpose of the discussion it is best to look at the requirements for active and passive devices separately. However on a general basis, for practical reasons, a first criterion should be fulfilled: the atomic resonance frequency should be in a useful range in order that its detection is not too difficult and that it can be used, for example, for locking a crystal oscillator. This constraint has greatly limited the choice of atoms for practical frequency standards. Of course, frequency standards exist at very high frequencies (stabilized lasers). However their signal frequency cannot be processed as easily as in the case of basic standards in the microwave region. These standards exist as a class almost by themselves and until, reliable, low cost, low complexity synthesis techniques are developed they will stay that way. This in no way, however, has inhibited research as such in this particular sector.

In active devices, such as the hydrogen and rubidium masers, where the storage principle in a bulb is used, other requirements are low atomic polarizability, in order that upon collisions ΔE and Δv are not altered too drastically, as well as absence of reaction with other atoms used as buffer gas or wall coating of the container.

In passive devices using beam techniques, the detection of particles requires a low ionization potential if the hot wire technique is used for particle detection. On the other hand if selective optical absorption is used to detect the state of the ensemble, the transition probability at the photon wavelength should be high.

Noble gases are chemically inert but they have a ground state 1S_0 , thus have no magnetic moment, have no hyperfine structure and do not possess a resonance frequency in the microwave range which

is a range of frequencies easily treated with present technology. On the other hand, hydrogen and alkali atoms (Li, K, Na, Rb, Cs, Fr) have a $^2S_{\frac{1}{2}}$ ground state and thus have a strong magnetic moment. Their hyperfine frequency is in a useful range where present day technology permits easy experimental signal processing. However they are far from being chemically inert. A compromise is thus made. They are either used as a beam or under the form of a vapor pressure contained in an enclosure coated internally with an inert film or imbedded in an inert gaseous matrix such as a buffer gas. The atoms that have been most studied for the purpose of atomic frequency standard applications are H, Rb, and Cs. Presently they form the core of the most documented atomic frequency standards. They have been used in different types of arrangements either active or passive. Unfortunately their frequency depends quite a lot on the magnetic field present and serious precautions must be taken to shield these atomic ensembles from environmental fluctuations.

Of course other atoms or molecules have been used for frequency standards applications or have been tried. For example NH_3 and Tl have interesting characteristics and have been used for implementing particular frequency standards. These will be discussed below. Other atoms have also been proposed and are presently under investigation. These are, Mg, Ca, Ag, Ba, Hg, Bi and others. However the implementation of these atoms in a given system, forms, for each one, a particular case and must be discussed on an individual basis. For frequency standard applications, they generally suffer from one of the requirements mentioned above: generally it is the frequency which is not in a useful range. Some other atoms suffer from the signal detection point of view. However, this is related to the actual experimental technique used.

Thus the main point to remember here is that the choice of the atomic ensemble for practical frequency standard applications depends largely on considerations related to the problem of "usable frequency" and this will stay true until reliable frequency synthesis from microwave to optical frequencies becomes routine work in the laboratory. On the other hand, intrinsic atomic particularities will dictate the experimental arrangement to be used.

b) The storage of the atomic ensemble: The atom must be stored somewhere to be examined as to what is its resonance frequency. Here, storage is taken in its most general sense. The most conventional storage processes are the following:

- Storage in a bulb under the form of a vapor.
 - . The inside of the bulb may be coated with an inert film. In that case the atom is free to move inside the bulb and makes collision with the wall. This permits long observation times; for hydrogen a lifetime in excess of one second is possible. However collisions with the wall create perturbations in the atomic wave function, causing relaxation and an average frequency shift.
 - . The bulb is not coated, but an inert gas is introduced by which the motion of the studied atoms is inhibited to some extent and can take place mainly through diffusion. This technique is used presently in Rb and Cs vapor frequency standards (passive and active). The buffer gases commonly used are N₂, Ar, Ne, CH₄. The atom has a lifetime limited by collision processes with the buffer gas atoms and diffusion to the wall.
 - . In some cases, it is preferable to use both techniques at the same time: wall coating and buffer gas. In this case the lifetime is

limited mostly by collision processes with the buffer gas.

- Storage through drift in a beam.

In this case the storage region is the atomic beam and the interro-gation or observation of the atomic ensemble is done at very specific regions in the path of the beam. A typical example is the case of the NH_3 maser in which the beam of molecules passes through a microwave cavity and emits its energy inside this same cavity. The average storage time τ is simply the length of the cavity divided by the average speed of the molecules. The line width is thus of the order of $1/\tau$. Another typical example is the Cs beam tube whose observation is done at two different regions of the beam path in two microwave cavities. In this case although the observation time in each region is short, the drift time between the two regions is of fundamental importance. If L is the distance between these regions the effective time of storage is $\sim L/v$ (v =atomic speed). It should be realized that this technique can be applied at any frequencies up to the visible. Since the line width, v/L , is in principle a constant for a given L , the line Q increases with the frequency. It is also clear that this type of storage is the one that is closest to that in which the atom is observed as a "free atom". However secondary effects are always present which affect either the stability or the accuracy of a particular device.

- Other storage techniques.

There are other storage techniques possible like those used in ion traps, in which the ion is kept in a region of space by alternating electric fields.

Associated with storage the main effects on frequency affecting stability or accuracy can be, collisions with the buffer gas or the

vessel wall, storage time (line Q), electric and magnetic fields, first and second order Doppler effects. The first order Doppler effect is mostly a design problem and is generally eliminated through a suitable experimental arrangement.

c) Preparation of the ensemble: Atoms are distributed among the energy levels according to the Boltzmann distribution law. This means that for temperatures above 300°K, where the frequency standards normally operate, all levels are equally populated even for energy differences corresponding to the microwave region. This is of fundamental importance for the following reason. The detection of the resonance signal is done through the excitation of transitions among a pair of levels. It is clear that if the levels are equally populated, the excitation of transitions has no effect on the populations themselves as well as on the excitation field itself. Consequently no effect is observed. However if there is a difference of population between the levels, the excitation field may either be enhanced or decreased, depending on the sign of the population difference. On the other hand the atomic distribution among the levels may be altered by the excitation and this effect, in a beam for example, may be detected through proper techniques like the counting of particles.

There are several methods that can be used to alter the population of the levels. Among those, the most common are spatial selection through magnetic field gradients (electric field gradients in the case of molecules with electric dipole moments) and optical pumping. Other mechanisms that can be used for state selection are also spin exchange, chemical reaction, electronic collisions, temporal decay from one state to another along a beam. However, they are not used in the traditional frequency standards as such, and will be mentioned at the time of discussion of some particular devices.

The process of preparation or state selection, has in some cases profound influence on the frequency of the atomic ensemble. For example we may mention light shifts in the case of optical pumping and creation of magnetic field inhomogeneities in the system where spatial state selection is done with magnets.

d) Interrogation of the ensemble: In order to know the actual atomic resonance frequency, a signal at that frequency is used. In the case of masers, a signal is generated inside the device which in a sense, interrogates itself. This is the case of the H, Rb and NH₃ masers for example. Lasers fall also in this class. It should be noted that in all these cases the signal is affected by various effects inherent to the process of emission in the resonance structure which is necessary to obtain maser action. This is the so-called cavity pulling effect and it affects both accuracy and stability.

In the case of passive devices the interrogation of the ensemble is done also at the resonance frequency. The atomic resonance signal, however, is observed through particular techniques which will be discussed below. However, here again the process of ensemble interrogation has very profound effects on the resonance signal. It can affect its frequency (stability or accuracy) through unsymmetrical interrogating spectrum, power shift through an unsymmetrical resonance line, a phase shift associated with cavity construction in the case of separated interrogating regions and cavity pulling. The interrogating signal used may also cause broadening of the line which may decrease the ability to find its center and consequently influence the accuracy and stability of the system.

c) Detection: In the case of masers, and some passive devices the signal to be detected is in the microwave range and normally a

superheterodyne technique is used. Although very efficient and sensitive, the technique adds noise to the signal and decreases the observed stability of the device. In passive devices the population difference of the particles having made a transition is monitored. These techniques are also very efficient but, of course, noise limits their sensitivity. In optically pumped devices, the noise comes from the photovoltaic device itself or the detected light. In some beam devices the counting of particles creates shot noise. Thus, detection in general is plagued with the problem of noise inherent either to the detector itself or the signal being detected and it is a major subject of research. Improvements in noise levels associated with detection means generally improvements in short term stability.

Survey.

Traditional frequency standards.

Three atomic frequency standards have known a rather intensive development phase. Those are the Hydrogen Maser, the Rubidium Gas Cell Frequency Standard and the Cesium Beam Frequency Standard. Their physical characteristics are given in table I.

Hydrogen Maser: (13, 14, 15)

An energy level diagram of the 2S_1 ground state of H is shown in figure 4 along with a schematic diagram showing the principles of operation of the device. The maser operates between levels $F = 1$, $m_F = 0$ and $F = 0$, $m_F = 0$. Atomic hydrogen is produced in a dissociator and sent through a selector (hexapole magnet) under the form of a beam. Atoms in the upper state tend to focus on the axis of symmetry of the magnet while those in the lower states have a trajectory which

diverges from the axis. Thus, atoms in state $F = 1$ and $m_F = 0$ enter the storage bulb which is coated internally with teflon and placed inside a high Q cavity. When the flux is strong enough oscillation takes place and the signal is observed with a small loop inside the cavity coupled to a superheterodyne receiver. Thus the preparation of the states is a spatial selection, the storage is done in a coated cell and the interrogation is done in a microwave cavity (self oscillation). The detection of the signal is done with a superheterodyne receiver.

The principal characteristics of such a device are its unsurpassed short, medium and long term stability. The spectral purity of a typical II maser is approximately:

$$S_y(f) = 5 \times 10^{-25} f^2 + 10^{-27}/f^2 \quad (3)$$

with a flicker floor probably well below $10^{-30} f^{-1}$. Its stability $\sigma(\tau)$ in the time domain is shown in figure 7. The characteristics given here are those of the maser itself. The stability is limited by temperature fluctuations through cavity pulling, magnetic field fluctuations, effects of load fluctuations on the cavity tuning and by the low power output which limits the short term stability. It is believed that the flicker floor has not been observed yet. The accuracy of the device is limited to about 2×10^{-12} due to uncertainties in the wall shift (16). The tuning accuracy through classical spin exchange broadening is of the order of a few parts in 10^{14} (17). Other secondary effects are also present, but they can be calculated or measured to a precision that is not a limiting factor (18, 17).

The device is not available commercially but is fabricated in various institutions, research laboratories and universities.

Cesium Beam Frequency Standard: (19, 20, 21, 22, 23)

A diagram showing the principles of operation of the cesium beam frequency standard is shown in figure 5 along with an energy level diagram of the atom. The drift region between the two cavities acts as the storage region. In the diagram shown, atoms in the $F = 3$, $m_F = 0$ state are those which strike the detector. Thus when the atoms, which are selected by magnet A, are induced to make a transition from the level $F = 4$, $m_F = 0$ to $F = 3$, $m_F = 0$ in the microwave cavity, the beam becomes populated with atoms in the $F = 3$, $m_F = 0$ level at the exit of region C. The flux of atoms reaching the detector is then a critical function of the frequency applied. The signal observed is also shown in figure 5. The application of r.f. radiation at two separate interrogation regions allows a considerable reduction in line width. It is in fact a function of the distance L between the two arms of the cavity: $\Delta\nu \approx 0.65 V_p/L$, where V_p is the most probable velocity.

The cesium beam tube, is a frequency discriminator and must be incorporated in an electronic servo system which locks a crystal oscillator to the resonance line. The spectral purity of a typical laboratory frequency standard is given by:

$$S_y(H) = 5 \times 10^{-29} / f \pm 10^{-24} \quad (4)$$

Its stability $\sigma(\tau)$ in the time domain is given in figure 7. In the short term region the stability is limited by shot noise at the detector. In laboratory devices, a flicker floor is observed at a level of the order of 10^{-14} or lower. The accuracy is about 1×10^{-13} in long laboratory models and is limited by magnetic field, cavity phase shift and 2nd order Doppler effects.

The cesium beam frequency standard exists as a primary standard in several laboratories and small units are available commercially.

Rubidium vapor frequency standard: (24, 25, 26, 27, 28, 29)

Figure 6 shows a diagram of the passive, optically pumped, rubidium 87 frequency standard (optical package). The basic principles are the following. Light from a rubidium 87 lamp is filtered by a so called hyperfine filter made of an ampoule containing rubidium 85 and a buffer gas such as argon. This arrangement shapes the spectrum of the light in such a way that a population inversion is obtained in the cell which follows and which contains rubidium 87 and a buffer gas. This cell becomes transparent through this optical pumping effect. If it is exposed to radiation at the proper frequency, corresponding to that which is necessary to excite transitions between the states $F = 3, m_f = 0$ and $F = 2, m_f = 0$ of the ground state, this same cell becomes opaque again and this effect can be detected through a decrease in light intensity reaching the silicon solar cell detector. Thus, in this present case, storage is done in a cell with a buffer gas, interrogation is done in the same region, preparation is accomplished through optical pumping and the detection of the signal is done with a solar cell.

This optical package is used as a frequency discriminator to lock a crystal oscillator to the resonance frequency. The spectral purity of the system is typically

$$S_y(f) = 10^{-26}/f + 5 \times 10^{-23}, \quad (5)$$

and its stability in the time domain is shown in figure 7. The principal factors limiting its stability are magnetic field, 2nd order Doppler effects, buffer gas and light shifts. All these effects may vary in time depending on the environmental conditions. Furthermore, there is generally a drift associated with the absorption cell which can be due to a reaction of rubidium with the glass of the enclosure. The accuracy of this device is not competitive with the two previous standards, mainly because of the very large buffer gas shift present.

The rubidium vapor frequency standard is available commercially under various models.

Other well documented frequency standards.

The physical characteristics of some other well documented frequency standards are summarized in table 2.

$^{15}\text{NH}_3$ maser: (30, 31)

In this maser the storage is accomplished through a single passage of a beam in a microwave cavity. The preparation is spatial electrostatic deflection. Interrogation is, as in the hydrogen maser, in a cavity, and the detection is done with a superheterodyne receiver. The short interaction time limits the stability of the maser to about 5×10^{-11} (cavity pulling). The accuracy is limited to the same value by collisions between molecules in the beam (32). The ammonia maser has played a very important role in the history of the development of presently available frequency standards.

Rb maser: (33, 34, 35, 36, 37).

Masers using either of the two isotopes Rb⁸⁵ or Rh⁸⁷

have been realized. Figure 8 is a block diagram of a Rb⁸⁷ maser. The principles of operation are the same as in the passive Rb gas cell; but the cavity is designed to have a very high quality factor and self oscillation is obtained. Thus the storage is done in a cell with a buffer gas, the preparation is done through optical pumping, the interrogation is made in a microwave cavity (self oscillation), and the detection is done with a superheterodyne receiver. The main advantage of the rubidium maser is its small size and its unsurpassed short term stability. However its long term stability is limited by cavity pulling effects and by the same factors as in the passive rubidium gas cell. The short term stability in present devices, is described by

$$\sigma(\tau) = 1 \times 10^{-13} \tau^{-1},$$

and it is not believed that the flicker floor has been observed yet.

Tl beam: (38, 39, 40, 41)

This device works essentially in the same way as the Cs beam tube, the only difference being the atom used and the frequency involved. The main problem in its realization has been a detection problem. The advantage over Cs would be a low magnetic field sensitivity. No work is presently being done on the device.

Rb⁸⁷ beam: (42, 43)

The implementation that has been studied in some detail is shown in figure 9. Here the only difference from the cesium beam is the method of preparation and of detection. This is done through optical pumping. One advantage of this method over the magnetic selection could be the realization of a better homogeneity of velocities across the beam. This could make cavity phase shift evaluation more

rigorous. This selection method could also be used with a Cs beam.

Saturated absorption stabilized lasers. (44, 45, 46, 47, 18, 49, 50, 51, 87)

This class of frequency standards falls in the passive type, the laser radiation being used as the interrogation signal generator and thus being slaved to a resonance appearing in an absorption cell. These frequency standards form a class by themselves operating at very high frequencies, in the terahertz region, where frequency comparisons are still an art. A schematic diagram of the device is shown in figure 10. Here the storage is done in a cell and actual preparation is not necessary. Due to the high frequency involved, the Maxwell Boltzmann law provides sufficient population difference between the levels of interest. The molecules commonly used are CH₄, N₂ and CO₂, with the proper laser providing the power for interrogation. The main particularity of the device resides in the method of interrogation called saturated absorption. The beam of light traverses the absorption cell twice in opposite directions, produces transitions and creates saturation only for those atoms having no first order Doppler effect or in other words crossing the beam at right angle. Thus, at the resonance frequency a slight increase in transmission is observed and this is the signal used to lock the laser to this same resonance frequency. The detector can be a PIN photodiode or an infrared device such as an In Sb detector. The stability $\Delta f/f$ of the methane stabilized He Ne laser is shown in figure 7 along with the traditional frequency standards. The accuracy of stabilized lasers is in the range of 10^{-11} to 10^{-12} and is limited by hyperfine structure, pressure shift, wave front curvature, 2nd order Doppler effect and recoil effect. (52).

Proposed atomic frequency standards.

Several other atomic frequency standards have been proposed. Their principal physical characteristics are shown in table 3.

Cs maser (53, 54)

This device works on the same principles as the Rb maser. Figure 8 in which rubidium is replaced by cesium applies in principle. The main advantage would be its good short term stability due to its expected high power output.

H beam tube (55, 56) and Ag beam device (57).

These devices work on the same principles as the Cs and Tl beam tubes. It is not evident that there are definite, theoretical or practical advantages in the proposed devices.

Cs cell (optically pumped) (58, 59, 60, 61)

This device works on the same principles as the Rb passive gas cell. The main advantage would lie in the weak interaction of Cs with glass, probably improving long term stability. However a bulky hyperfine filter must be used to filter the light for efficient optical pumping.

Magnesium and calcium beam. (62, 63, 64)

This proposal is based on temporal state selection in a beam. Magnesium and calcium show a metastable 3P level with fine structure. The 3P states are populated through electron collisions and a beam is formed. The 3P_1 level then decays to the ground state by emitting visible light with a lifetime of the order of 1 ms. A

submillimeter wavelenght radiation exciting the transition 3P_0 to 3P_1 , for instance, replenishes the 3P_1 level. Its decay to the ground state gives an added emission of light which allows the detection of the transition. The main advantage would reside in a very high line Q, due to the long lifetime involved and the high frequency of the transition. Major problems reside in the generation of the submillimetre wave for interrogating the resonance.

Ion storage (65, 66, 67, 68, 69, 88)

This device is based on the storage of ions in a trap formed by non uniform alternating electric fields without the need of any applied magnetic fields. The stored ions do not collide with any surfaces and storage times longer than several hours have been reported for sufficiently low background pressures. If the stored ions possess a hyperfine structure, one could then think of state preparation, interrogation and detection in the same way as in conventional passive frequency standards. The hyperfine splitting of $^3\text{He}^+$ has been precisely measured and the sources of frequency offsets analysed (65). However light ions have a large second order Doppler frequency shift and are not suited for frequency standard applications. Heavy ions such as mercury appear to be more convenient. The hyperfine splitting of $^{199}\text{Hg}^+$ has been observed with a fractional linewidth of 2×10^{-10} and has been precisely measured. This result is attractive for frequency standards application. The proposed scheme for detection of the resonance is similar to what is used in the passive optically pumped standard. Light from a $^{202}\text{Hg}^+$ lamp is selectively absorbed by one of the hyperfine components of the $^{199}\text{Hg}^+$ ions. The hyperfine transition at 40.5 GHz is thus detected by the change in the intensity of the resonance fluorescence. One main difficulty in this technique resides in the low signal to noise ratio available due to the low ion density at which the trap operates.

H beam storage (70, 71)

The basic idea behind the operation of this device is the same as in the operation of the hydrogen maser, except that it is operated in the passive mode. A microwave signal is used to interrogate the atoms stored in a coated bulb and the effect of the atomic transitions on the interrogation signal is used to lock a crystal oscillator to the atomic resonance. In one version, the modulation technique used is one in which the intensity of the beam is altered. Otherwise the system works in a way similar to passive frequency standards. The main advantage is the reduction of the effect of cavity pulling through the possibility of using very low Q cavities since the oscillation condition does not need to be satisfied. However the wall shift problems remains and should be the principal limit to the accuracy of the device.

NH₃ absorption cell (72, 73)

This is one of the simplest versions of an atomic frequency standard and is based on one of the oldest ideas. The storage is done in a cell and the full Doppler broadened absorption line is interrogated. The states are not prepared and for the absorption, one relies on the Maxwell Boltzman distribution law of the populations among the levels. The microwave absorption is detected through standard techniques and a crystal oscillator is locked to the atomic resonance signal. The accuracy is low ($\sim 10^{-9}$) and the stability is given by

$$\sigma(\tau) = 6 \times 10^{-10} \tau^{-\frac{1}{2}},$$

with a flicker floor in the 10^{-10} range. The main purpose is the realization of a small, rugged, fast warm up device.

Atomic beam stabilized lasers (74, 75, 76, 77)

This is a class of devices in which a laser is stabilized through its interaction with a resonance frequency that can exist in an atomic beam as shown in Fig. 11. In this case the atoms which are used as a reference for stabilizing the laser can be considered free in the same sense as in the passive cavity frequency standard. Several implementations are possible. In the simplest approach the absorption of the laser light by the atomic beam is detected and is used as the resonance signal for locking the laser. More recently the technique of separated Ramsey interrogation regions has been proposed for the optical range. In principle it should provide extremely high line σ (10^{11} - 10^{12}). This proposal coupled with the two photon absorption technique (78, 79) shows promises in stability and accuracy. Atomic beams which appear to attract much attention are rubidium, cesium and iodine.

Forecast

Traditional devices

Figure 7 and equations 3, 4 and 5 give the present state of the art stability of the three most documented and most utilized frequency standards: H maser, atomic tube, Rb gas cell. An analysis over the past years shows that the stability of these devices, especially in the long term region has been increasing continuously. Better understanding of the physical processes involved and of the sources of noise has provided a basis for a better design of the devices themselves. The H maser has now passed the 10^{-15} level in the 10,000 second region and it appears that the limitation is still a question of cavity temperature control and magnetic shielding. It is thus probable that with some reasonable effort, further improvements will be realized. It should be mentioned that the order of magnitude of

stability mentionned here is not common laboratory practice. Care should be taken in the transfer of this kind of stability to quartz crystal oscillators. It appears that one must be extremely cautious about time varying phase shifts being generated in the detection components. However it is not unlikely that through better cavity design and magnetic shielding and through improved detection systems a flicker floor below the 10^{-15} level would be observed. The laboratory Cs beam tube has reached a level of flicker floor of the order of 7×10^{-14} . In the short term region, better stability could be achieved with more intense beams but this could affect the lifetime of the device.

The accuracy of the hydrogen maser is still limited at the time of writing by the wall shift, to about 2×10^{-12} . This is two orders of magnitude worse than any other uncertainties related, for example, to the magnetic field determination or cavity tuning. It is not possible at this time to tell exactly what kind of improvement we can hope for. However there is intensive work being done on wall coatings and bulbs with variable geometries. It appears that operation at a temperature close to the point where the wall shift goes to zero may be a proper avenue of research. The accuracy of the laboratory cesium beam device has passed the 10^{-13} level. The main uncertainties are originating in the magnetic field and the cavity phase shifts. One problem lies on the non-uniform distribution of velocities across the beam. This makes the actual evaluation of the cavity phase shift difficult. It is possible that through homogeneous state preparation a better evaluation of the cavity phase shift could be done. People working in this field believe that an accuracy of 1×10^{-14} can effectively be achieved. These comments are summarized in table 4 along with some projections on the rubidium gas cell device. The numbers in circles are the author's evaluation of the possibility that the projection will be realized, 0 meaning no confidence at all and 5 meaning

certainty. This evaluation is based on many intangible factors and is entirely subjective.

Weight and size: The hydrogen maser has been reduced considerably in size during the last several years and it appears that further reduction could be realized by using cavities in the TE111 mode (80). Present state of the art masers occupy a volume of the order of 100 liters and weigh 45 kg. In the case of commercial cesium beam frequency standards it appears that further progress in size and weight could be gained through a reduction in size of the tube but with a parallel degradation in stability and accuracy. On the other hand it appears that further improvements are possible in the field of passive rubidium frequency standards. The optical package is gaining more and more in simplicity and the use of integrated circuits and modern technology has made possible real gain in size. At present, units 2 liters in volume and weighing 2 kg are available commercially.

Other devices.

The other devices or techniques which appear most promising are the hydrogen beam device, the ion storage technique, the magnesium beam device and atomic beam stabilized lasers using optical Ramsey resonance techniques. The hydrogen beam storage technique has proven to be feasible and reports on its stability are most encouraging. The technique will also allow an easier determination of the wall shift than in an actual maser. The ion storage technique has made some progress but it appears that serious problems are still encountered at the S/N level. Work is in progress to solve this problem (81). In the case of the magnesium beam proposal, the problem of generating an interrogating signal at the resonance frequency appears to be important. However efforts are being made with infrared lasers in this direction and we

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FIGURE CAPTIONS

- Figure 1. The two basic versions of atomic frequency standards. The continuous paths apply to the passive mode, while the dotted paths apply to the active mode.
- Figure 2. Block diagram of the phase lock loop commonly used to lock a crystal oscillator to a maser signal.
- Figure 3. Block diagram of the frequency lock loop normally used to lock a crystal oscillator to the atomic resonance line.
- Figure 4. Schematic diagram of a hydrogen maser.
- Figure 5. Schematic diagram of the cesium beam tube used in passive cesium frequency standards and using the separated Ramsey cavity technique.
- Figure 6. Schematic diagram of the optical package used in the passive rubidium 87 frequency standard. In some practical devices the isotopic filter is removed and natural rubidium is used in the storage cell. Mixtures of isotopes can be used in the lamp to reduce the light shift.

Figure 7. Time domain frequency stability of the three traditional frequency standards: H maser, Cs beam frequency standard and passive Rb gas cell. The stability of the methane stabilized laser is also shown for comparison.

Figure 8. Schematic diagram of the optical package of the Rb maser. This is similar to the diagram of figure 6 except for the detection of the resonance signal. Here the cavity Q is high enough to permit oscillation and the signal is detected with a superheterodyne receiver.

Figure 9. Passive rubidium beam frequency standard using optical pumping for state selection and detection.

Figure 10. Schematic diagram of a saturated absorption stabilized laser.

Figure 11. Basic concepts used in the stabilization of a laser with an atomic beam. (One version is shown and several other implementations can be thought of.)

TABLE I
PRINCIPLES OF OPERATION OF THE TRAJECTORY
ATOMIC FREQUENCY STANDARDS

Frequency standards	Transition frequency (Hz) (Atomic ensemble)	Basic principles (Storage + interrogation)	State selection (Preparation)	Detection system (detection)	Atoms: Fine (Δ)	Main sources of frequency offset
Cs beam	9 192 631 770 Hz defining ion of the atomic trap second	Passive atomic trap magnetic resonance cavities	Magnetic deflection	Hot wire ionizer	$\frac{2}{3} \times 10^6$ to 3×10^8	Magnetic field Doppler (2nd order) cavity phase shift
H water	1 420 405 751, 768 + 41602	Active aner Atomic beam Teflon coated storage cell	Magnetic deflection	Sinerheterodyne receiver, 16-13 K	$\frac{2}{3} \times 10^4$	Magnetic field Doppler (2nd order) wall shift
Pb cell	6 834 682 612, 8 + 0.5	Passive Storage cell Baffler pins	Optical pumping	Silicon solar cell	5×10^7	Magnetic field Doppler (2nd order) Baffler gas and light shifts

TABLE 2

 PRINCIPLES OF OPERATION OF WELL EQUIPPED
 ATOMIC FREQUENCY STANDARDS

Frequency standards (Atomic ensemble)	Transition frequency (Hz)	Basic principles (Storage + interrogation)	State selection	Detection system (detection)	Atomic line Q	Main sources of frequency offset
$^{15}\text{Mg}_3$ maser	22 789 421 701 ± 1	Active maser Atomic beam Single passage in a cavity	Electrostatic deflection	Superheterodyne receiver 10^{-10} W	5×10^6	Collisions in an intense beam
Rb maser	87 Rb: 6 834 682 612.8 ± 0.5	Active maser Storage cell buffer gas	Optical pumping	Superheterodyne receiver 10^{-10} W	5×10^7	Magnetic field Doppler (2nd order) Buffer gas and Light shifts
	$^{85}\text{Rb}:$ 3 035 732 439 ± 5					
^{87}Rb beam	6 834 682 612.8 ± 0.5	Passive beam Ramsey cavities	Optical pumping	Photomultiplier	6×10^6	Magnetic field Doppler (2nd order) Cavity phase shift
Tl beam	21 310 833 946.6 ± 0.2	Passive Atomic beam Ramsey cavities	Magnetic deflection	Oxidized hot wire ionizer	10^8	Magnetic field Doppler (2nd order) Cavity phase shift

TABLE 2 (continued)

PRINCIPLES OF OPERATION OF ATOMIC FREQUENCY
ATOMIC FREQUENCY STANDARDS

Frequency standards (Atomic ensemble)	Transition frequency (Hz)	Basic principles (Storage + interrogation)	State selection	Detection system (detection)	Atomic line Q	Main sources of frequency offset
Saturated absorption stabilized lasers	Iodine 127 R(127) 11-5 nm, 1 component (473, 612, 214, 4 ± 1.9) 10 ⁶	Passive absorption in a storage cell	Not needed (Maxwell- Beltramini line)	Silicon PIN photodiode	2×10^8	At present: hyperfine structure or pressure shift
Methylene $\text{P}(\text{7})_{\text{v}2}$ band (388, 376, 181, 627 ± 50) 10 ⁵			InAs or GaAs InSb detector	4×10^8 1.5×10^{10}	In future: wavefront curvature	
Carbon dioxide $\text{P}(\text{7}, \text{s}0)$ (29, 442, 483, 315 ± 2.5) 10 ⁵			Cooled InSb detector	5×10^7	Doppler (2nd order) recoil effect	

TABLE 3
PRINCIPLES OF OPERATION OF PROPOSED ATOMIC FREQUENCY STANDARDS

Proposed devices (Atomic ensemble)	Transition frequency (Hz)	Basic principles (Storage + interrogation)	State selection	Detection system (Detection)	Atomic line Q	Major sources of frequency offset
H beam tube	1 420 405 751.768 + 0.002	Passive Atomic beam Ramsey cavities	Magnetic deflection	Penning gauge	10^6	Magnetic field Doppler (2nd order) Cavity phase shift
H beam storage	1 420 405 751.768 + 0.002	Passive Atomic beam Teflon coated Storage cell	Magnetic deflection	Superheterodyne receiver	2×10^9	Magnetic field Doppler (2nd order) Wall shift
Silver beam	10^7 Ag : 1 712.512 111 + 18	Passive Atomic beam Ramsey cavities	Magnetic deflection	Electron bombardment ionizer	10^6	Magnetic field Doppler (2nd order) Cavity phase shift
$^{109} \text{Ag}$:	+ 976.932 075 + 17					
Stored $^{199} \text{Hg}$	40 507 348 050 + 50	Passive Storage of ions in an electric field	Optical pumping	Photomultiplier	5×10^9	Magnetic field Light shift Doppler (2nd order).

TABLE 3 (cont'd)
PRINCIPLES OF OPERATION OF PROPOSED ATOMIC
BRIDGES STANDARDS

Proposed devices	Transition frequency (Hz) (Atomic ensemble)	Basic principles (Storage + interrogation)	State selection (detection)	Detection system (detection)	Atomic line Q	Main sources of frequency offset
Calcium beam	$\{1563.6\} \times 10^9$	Passive Atomic beam Two interaction regions	Decay along the beam	Photomultiplier	10^9	Wigner effects
Magnesium beam	$\{664.52\} \times 10^9$	Passive Atomic beam Two interaction regions	Decay along the beam	Photomultiplier	10^9	Magnetic field 2nd order Wigner and 2nd order
Caesium cell	9 192 631 770	Passive Storage cell Buffer gas	Optical pumping	Silicon solar cell	5×10^7	Magnetic field Raman (2nd order) Buffer gas and light shifts
Cs waser		Active laser Storage cell Buffer gas	Optical pumping	Superluminescent receiver	5×10^7	Magnetic field 2nd order Raman (2nd order) Buffer gas and light shifts
Atomic beam Stabilized laser (Ga, Bi, I ₂ ...)		Passive 2 photon transitions saturated absorption Raman Interac- tion Regions	Decay along the beam Resonant selection	Hot-wire detector Photomultiplier	$10^8 - 10^9$ (one interac- tion region)	Light shifts 2nd order Wigner effects

TABLE 4
TRADITIONAL ATOMIC FREQUENCY STANDARDS. STABILITY AND ACCURACY,
STATE OF THE ART AND PROJECTION

Frequency Standard	Stability			Accuracy	
	Present	Projected	Projected	Present	Projected
	Short term	Flicker Floor	Short term	Flicker Floor	
<u>Cs beam</u>	$7 \times 10^{-14} \tau^{-\frac{1}{2}}$	7×10^{-15}	same	1×10^{-15} ⁽³⁾	8×10^{-14}
	$5 \times 10^{-12} \tau^{-\frac{1}{2}}$	5×10^{-14}	-----	-----	10^{-14} ⁽¹⁾
<u>Commercial (I.P.)</u>					-----
<u>H maser</u>	not observed	-----	-----	$< 10^{-15}$ ⁽⁴⁾	2×10^{-12}
	$4 \times 10^{-13} \tau^{-\frac{1}{2}}$				1×10^{-15} ⁽²⁾
<u>Rb gas cell</u>	$5 \times 10^{-12} \tau^{-\frac{1}{2}}$	1×10^{-13}	$1 \times 10^{-12} \tau^{-\frac{1}{2}}$ ⁽³⁾	1×10^{-14} ⁽²⁾	-----

TABLE 5
PREDICTING ATOMIC FREQUENCY STANDARDS: STABILITY AND ACCURACY.
STATE OF THE ART AND PROJECTION

Type of device	Stability			Accuracy	
	Present	Projected	Projected	Present	Projected
Saturated absorption stabilized laser (CH ₄)	Short term $1.5 \times 10^{-14} \text{ s}^{-1}$ $3 \times 10^{-13} \text{ s}^{-1}$	Present 8×10^{-13} Projected 3×10^{-14}	Short term $2.5 \times 10^{-12} \text{ s}^{-1}$ (4)	Present $\sim 2 \times 10^{-12}$ 10^{-11}	Present ----
Mg, Ca lasers	Projected Q _e = 5×10^6	Present 10^{-14} s^{-1} (2)	Present 10^{-14} s^{-1} (3)	Present ----	Present 10^{-14} (2)
Rb Maser	Present $1 \times 10^{-13} \text{ s}^{-1}$	not observed	Present 4×10^{-14} Projected Q _e = 3×10^{10}	Present $\sim 10^{-14}$ (3)	Present 10^{-14} (3)
Ion Storage (Hg, Bi)	Present $Q_e = 3 \times 10^9$ Projected Q _e = 3×10^{10}	Present $3 \times 10^{-12} \text{ s}^{-1}$ (1)	Present ----	Present ----	Present 10^{-13} (1)
Atomic Beam stabilized laser 2-beam absorption and separated Ramsey Interrogation Regions (Ca, Bi, Li, ...)	Present 5×10^{-12} for $\approx 0.1 \text{ s}$	Present $Q_e = 2 \times 10^8$ Projected Q _e = 10^{12}	Present one interaction region	Present Ramsey interrogation technique (3)	Present ----
H Maser (passive)	Present $1.5 \times 10^{-12} \text{ s}^{-1}$	Present $7 \times 10^{-13} \text{ s}^{-1}$ (1)	Present $\sim 10^{-15}$ (3)	Present 10^{-12} (3)	Present 10^{-13} (1)
Mg cell (passive)	Present $6 \times 10^{-10} \text{ s}^{-1}$	Present 2×10^{-10}	Present $1 \times 10^{-10} \text{ s}^{-1}$ (3)	Present 2×10^{-11} (3)	Present 5×10^{-10} (2)

should expect results soon (82). Finally the two photon absorption technique coupled with the separated Ramsey interrogation technique appears to be extremely promising. In fact a line Q of the order of 10^{12} is projected and appears realistic (83). These projections are summarized in table 5 along with projections on some other devices which were examined in this paper.

Frequency synthesis.

As was mentioned earlier, stabilized lasers form a class by themselves which has no easy connection with the traditional frequency standards such as the Cs beam frequency standards. Until reliable synthesis up to the frequencies encountered in the laser stabilization field, becomes a common laboratory technique, a gap will exist between the traditional standards and the stabilized lasers, and these last devices will have limited use. However research on this subject is being done in some laboratories. (84, 85). Presently it is possible through the use of several lasers and point contact metal diodes, to mix up and synthesize frequencies up to a corresponding wavelength of 1.5μ . (86) However, this still remains somewhat of an art and intensive work is required in this direction if substantial results are wanted.

ATOMIC FREQUENCY STANDARDS : BASIC PRINCIPLE

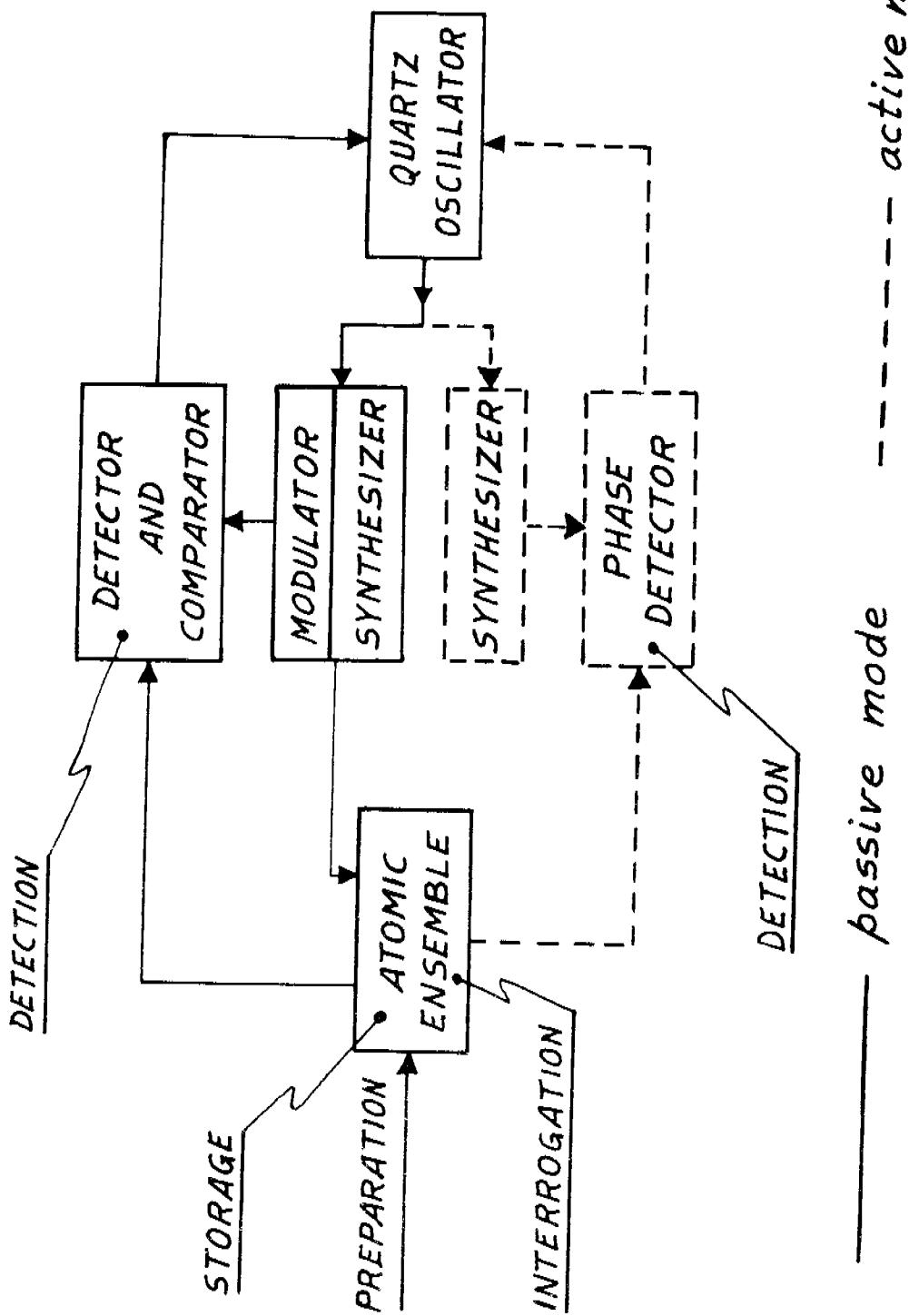


FIGURE 1

ACTIVE CRYSTAL OSCILLATOR MASER PHASE LOCK LOOP

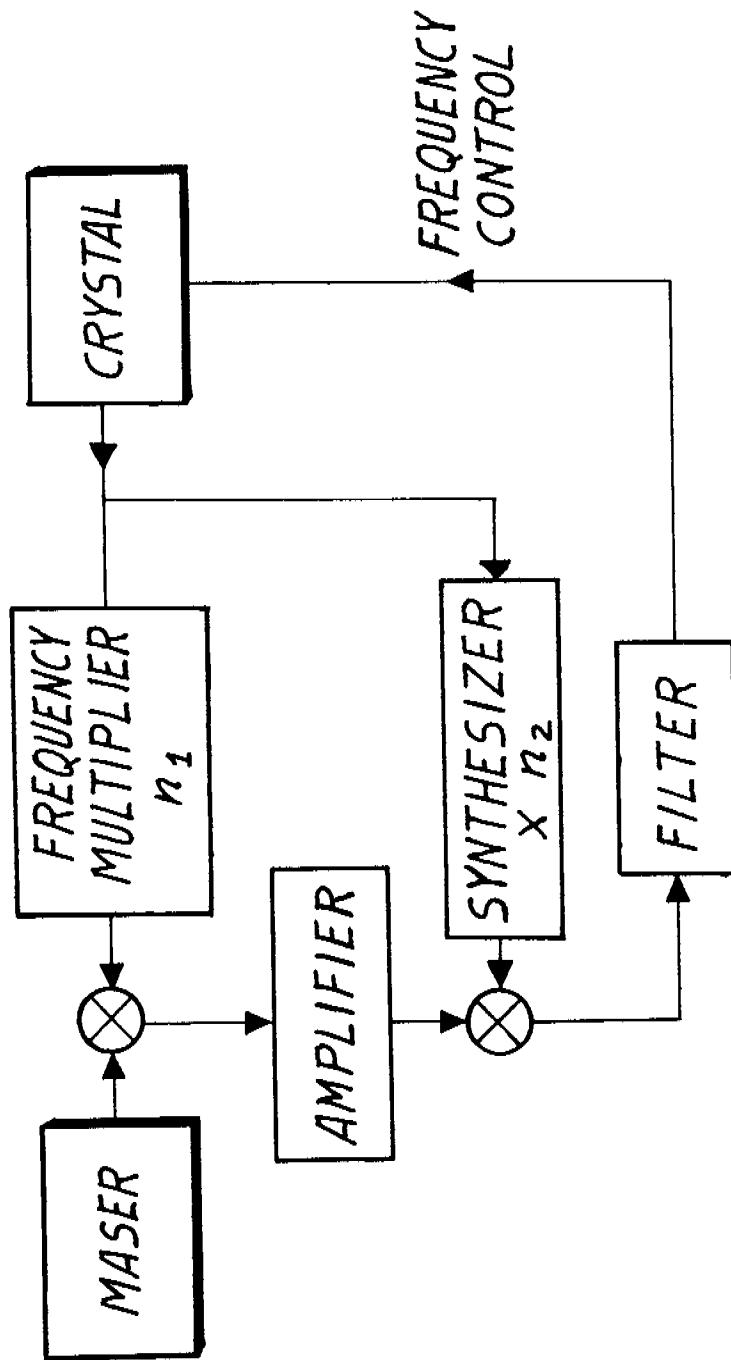


FIGURE 2

CRYSTAL OSCILLATOR PASSIVE REFERENCE FREQUENCY LOCK LOOP

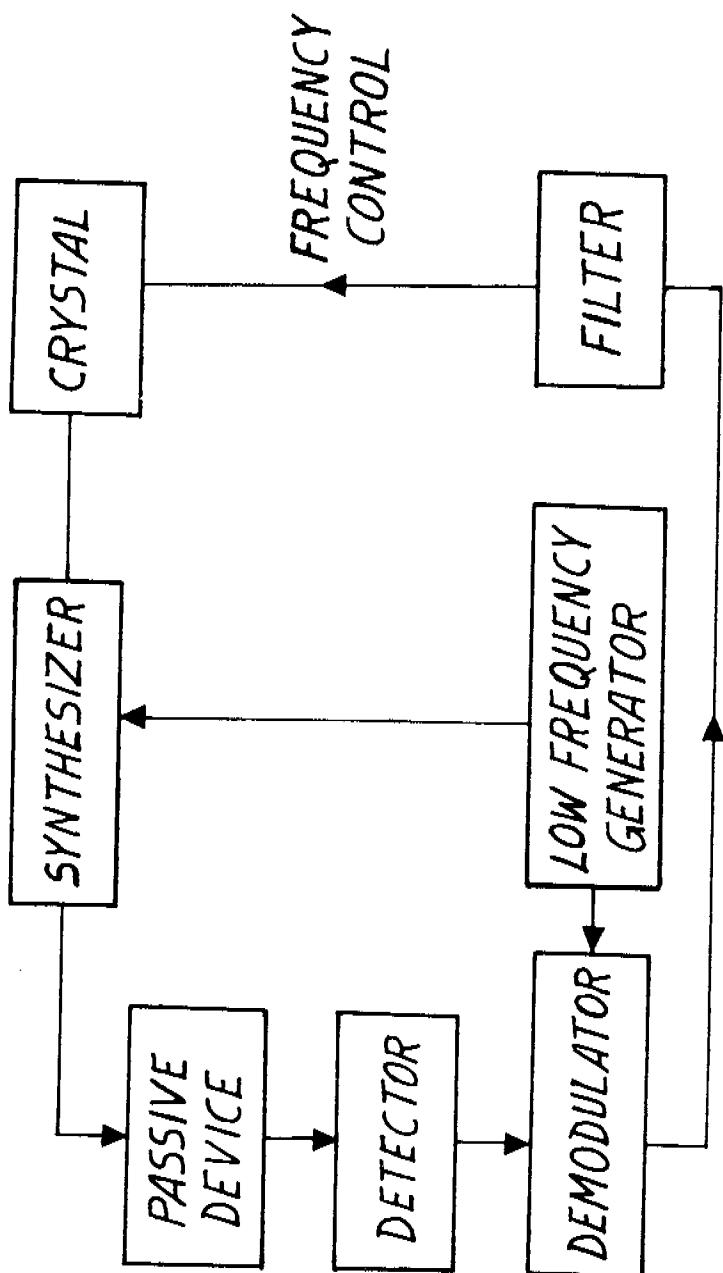


FIGURE 3

H MASER

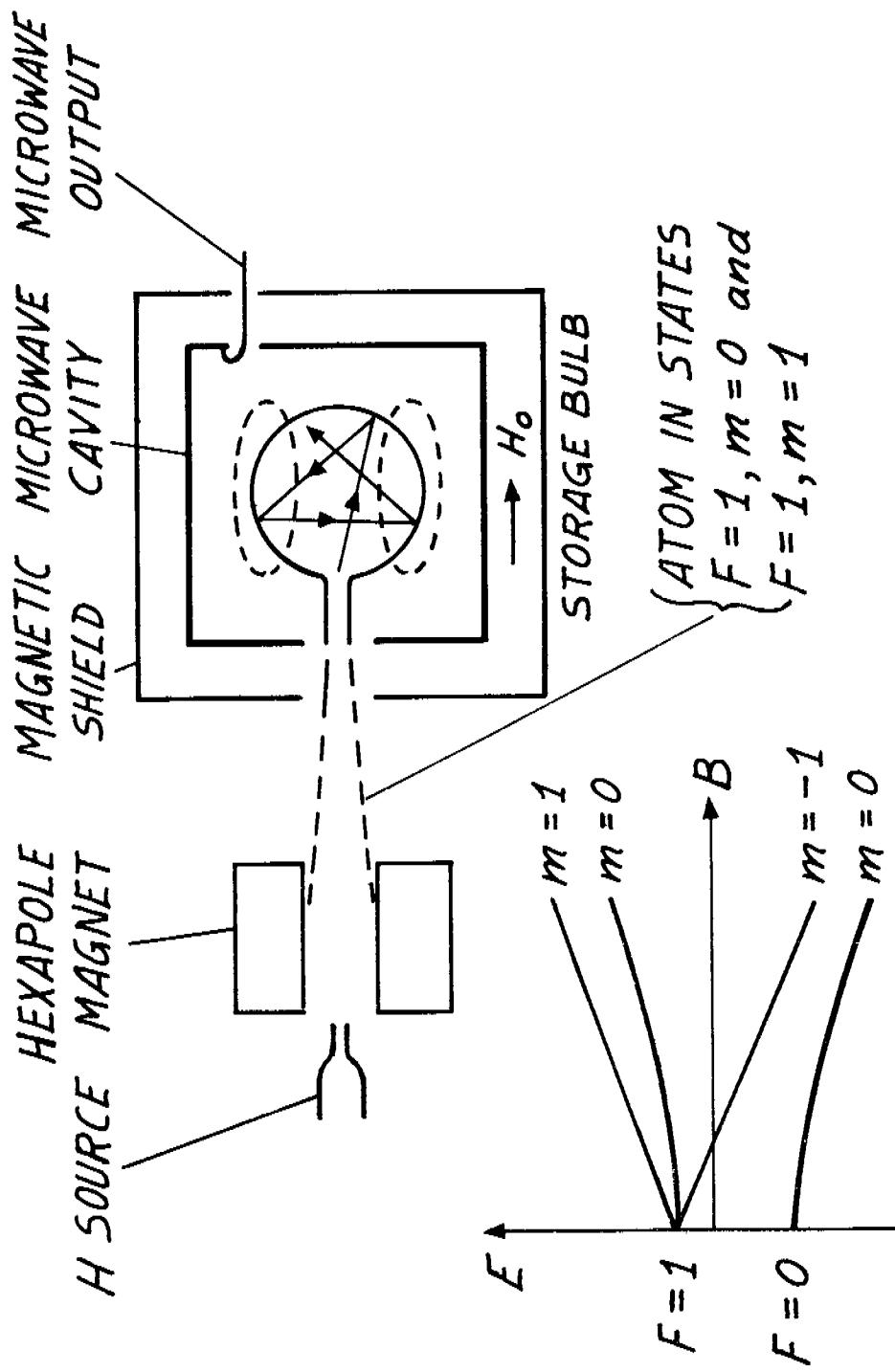
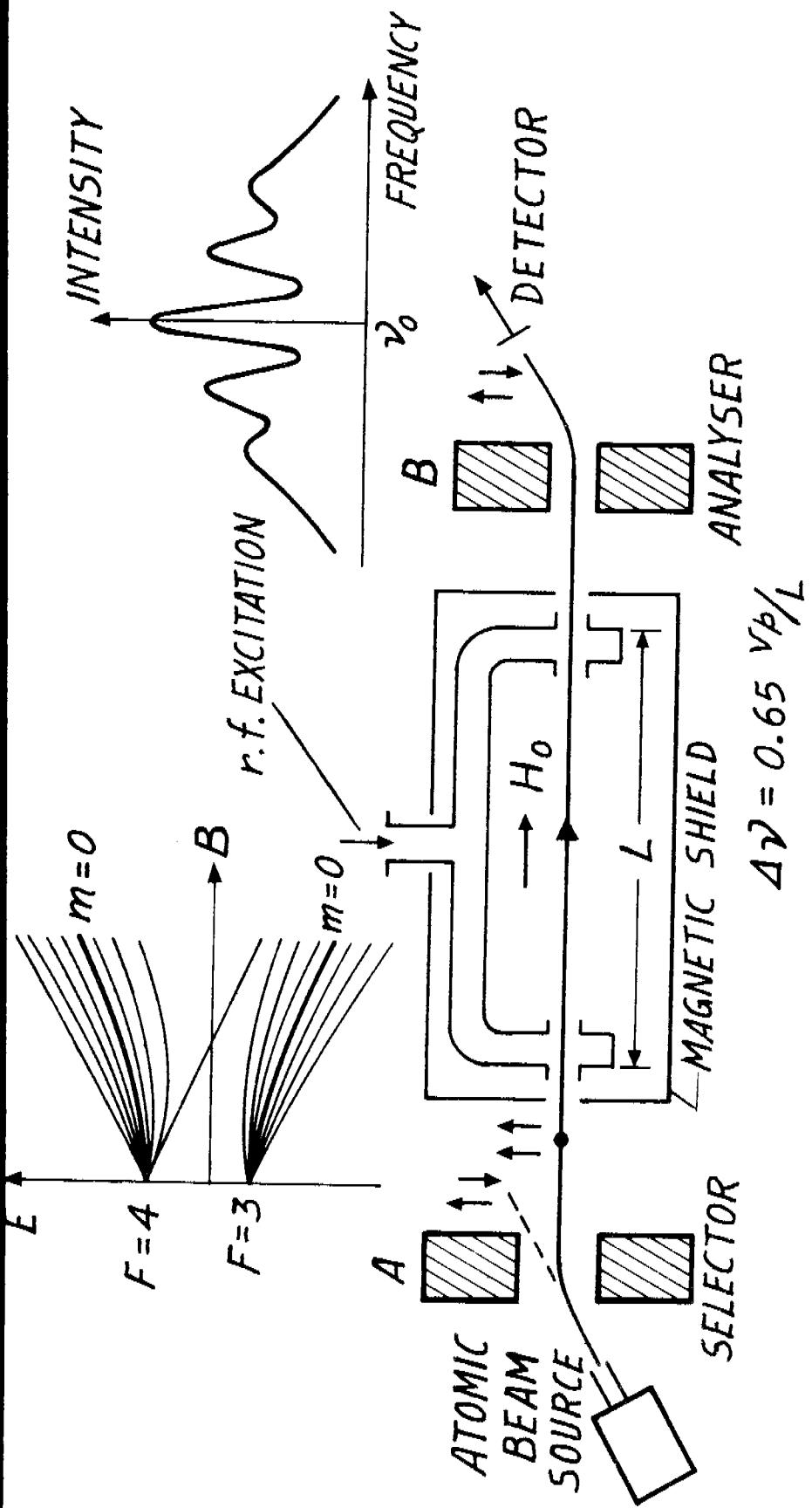


FIGURE 4



PASSIVE CESIUM BEAM FREQUENCY STANDARD

FIGURE 5

PASSIVE RUBIDIUM GAS CELL FREQUENCY STANDARD

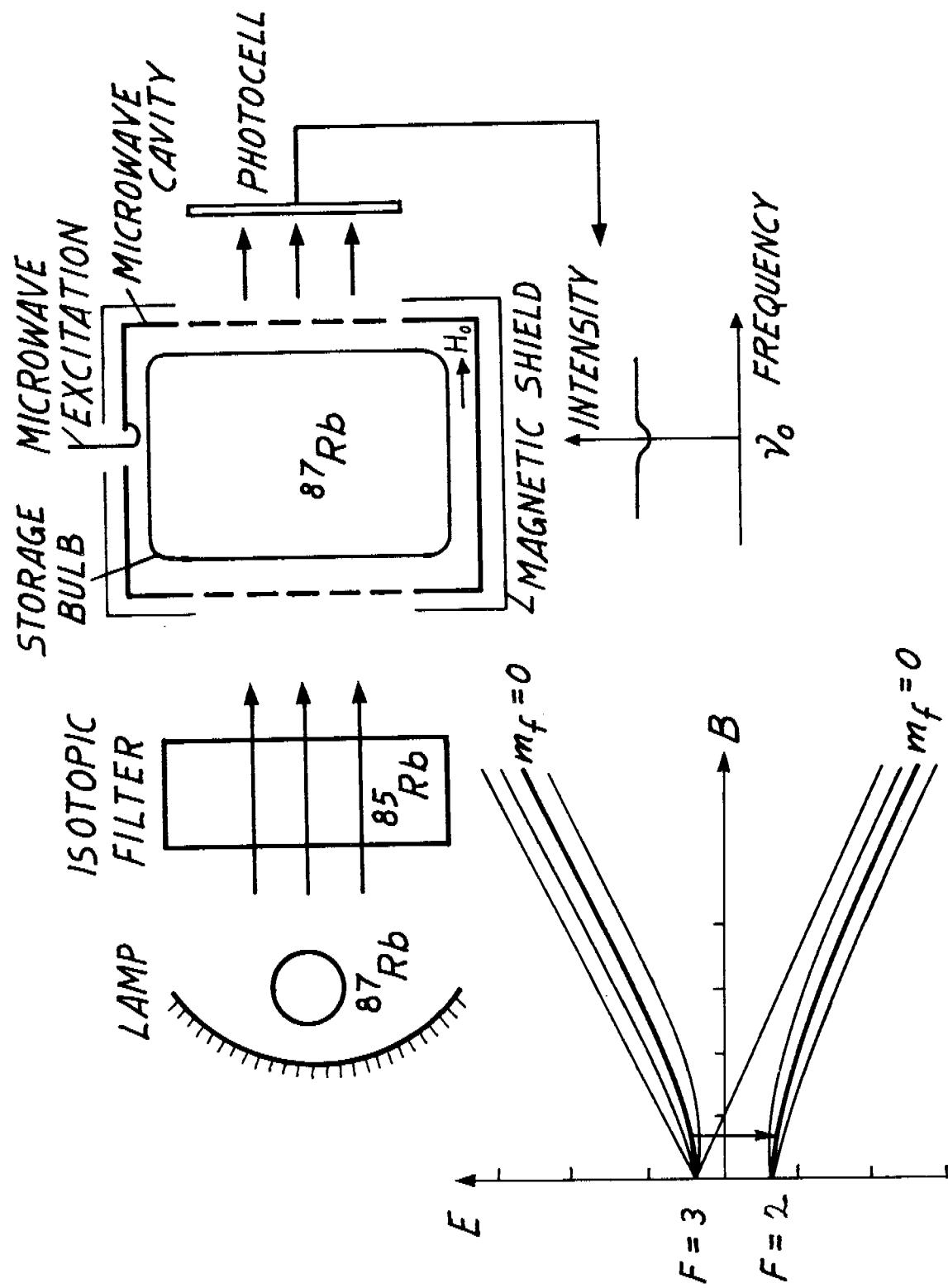


FIGURE 6

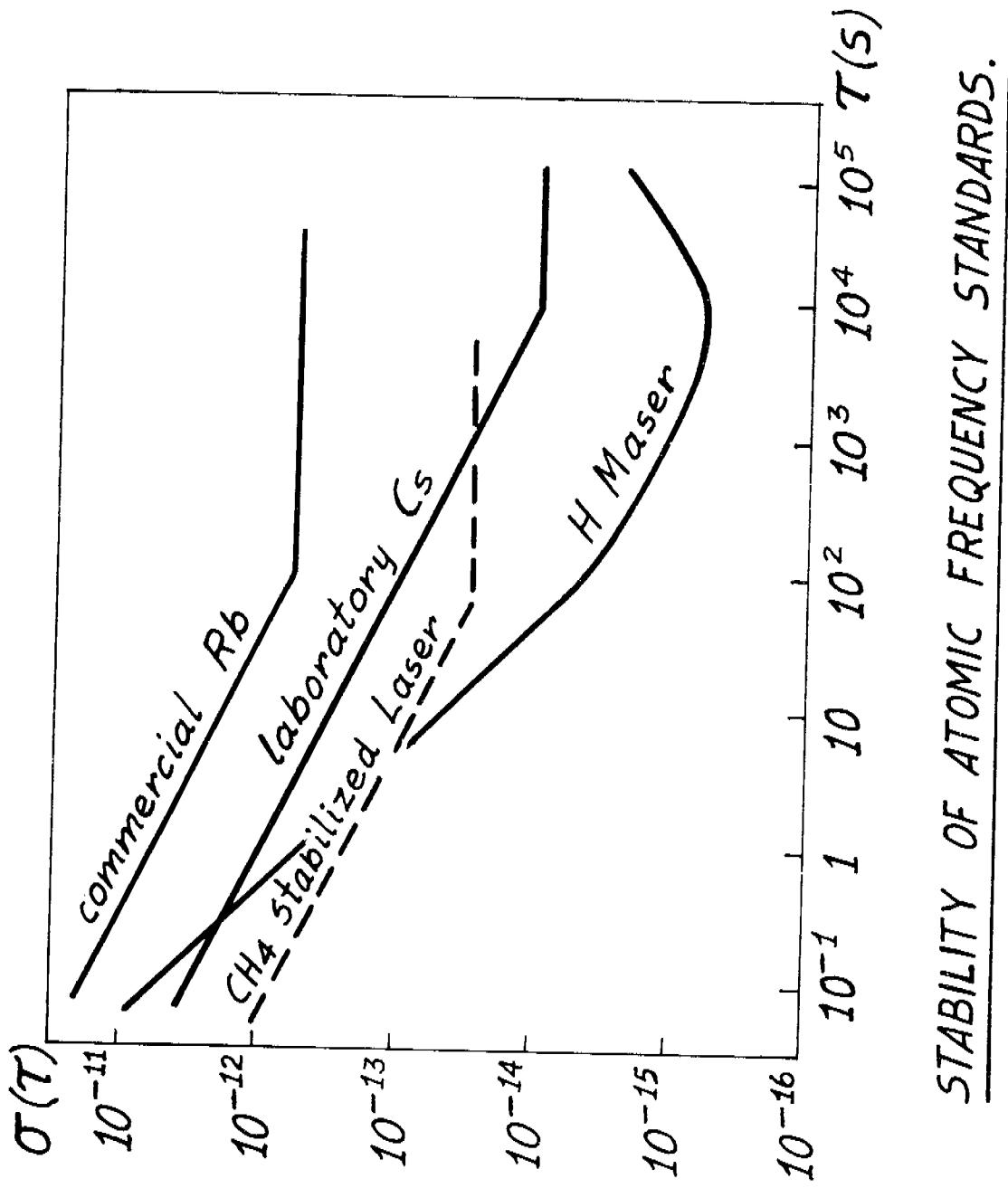


FIGURE 7

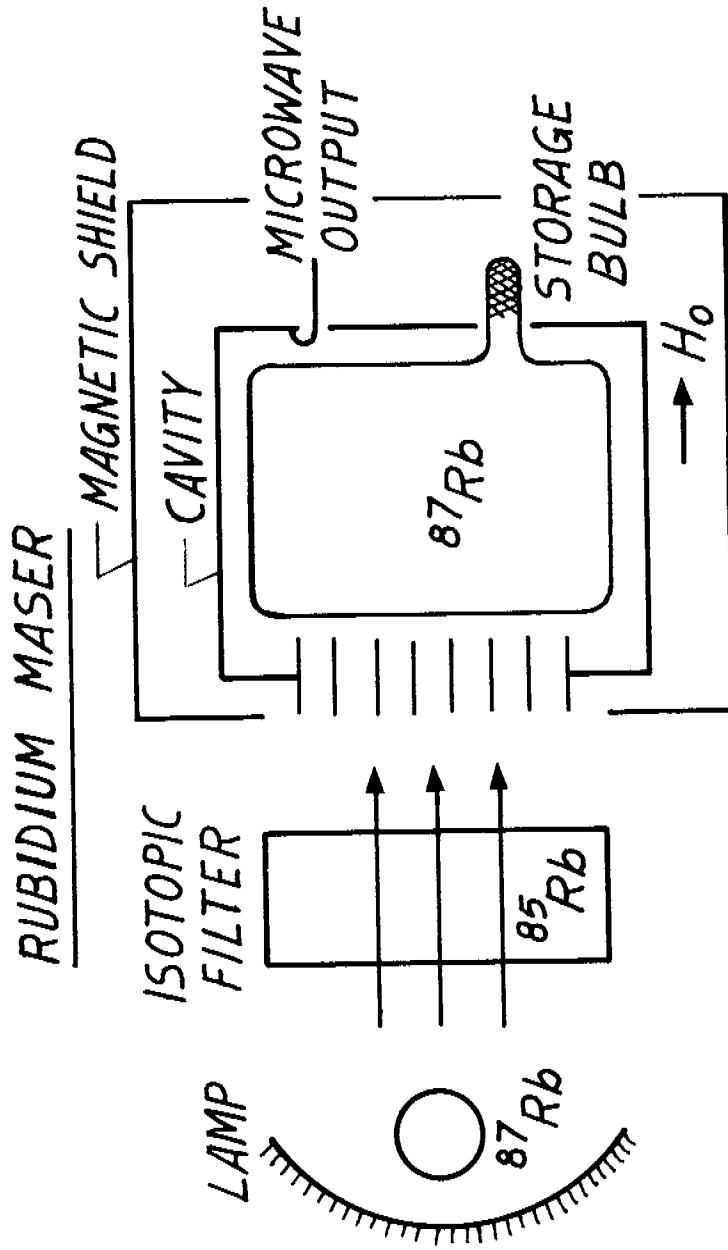


FIGURE 8

PASSIVE RUBIDIUM ATOMIC BEAM FREQUENCY STANDARD
(optically pumped)

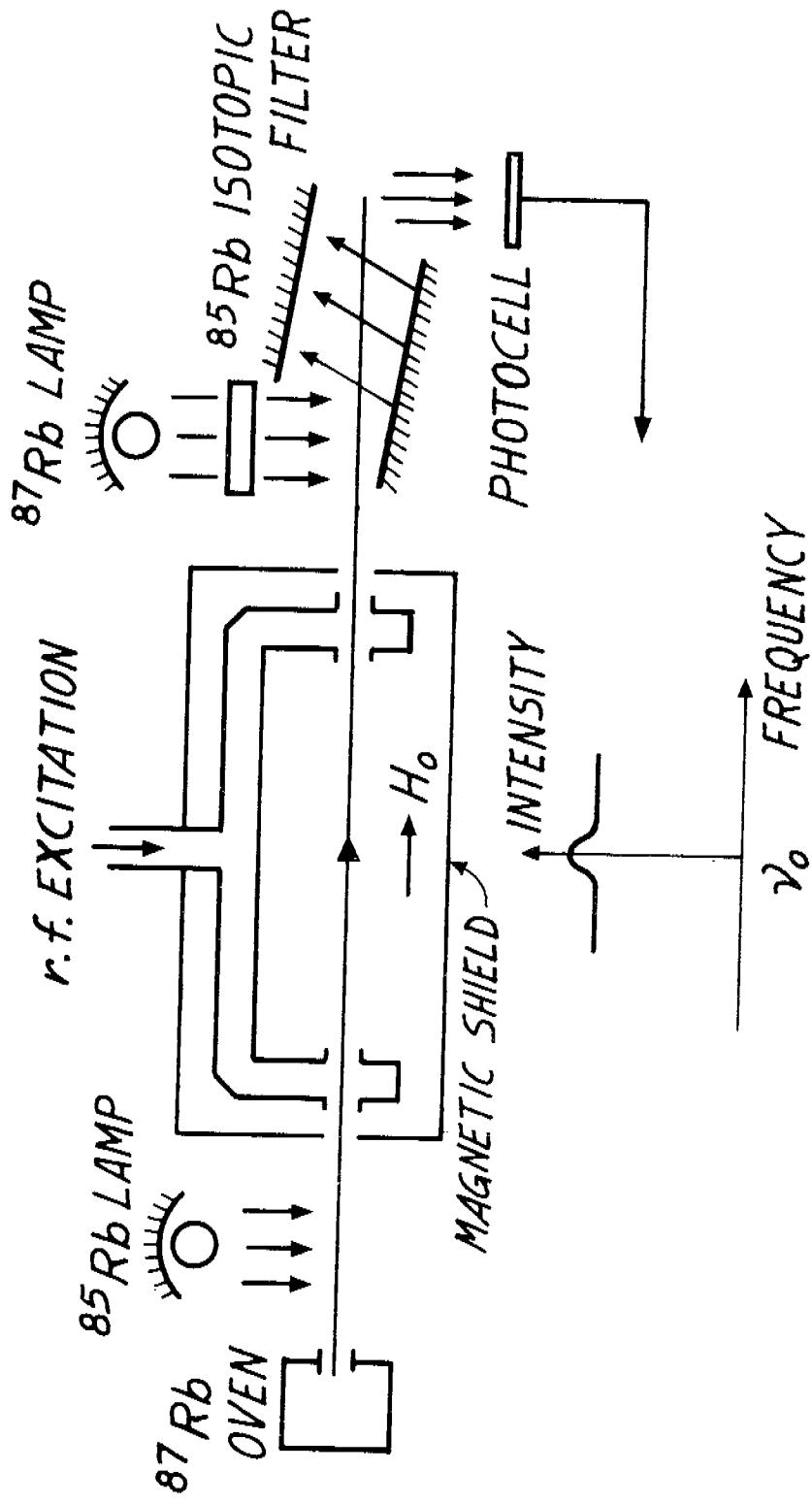


FIGURE 9

SATURATED ABSORPTION STABILIZED LASER

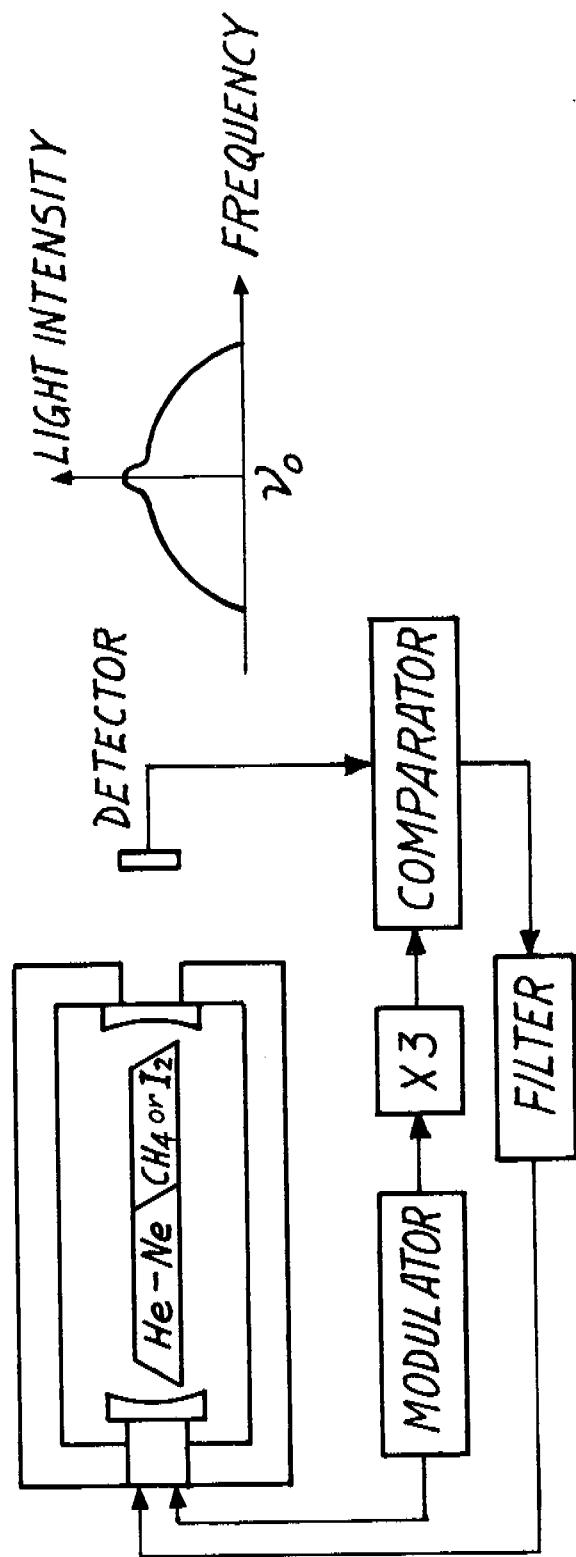


FIGURE 10

ATOMIC BEAM STABILIZED LASER (one version)

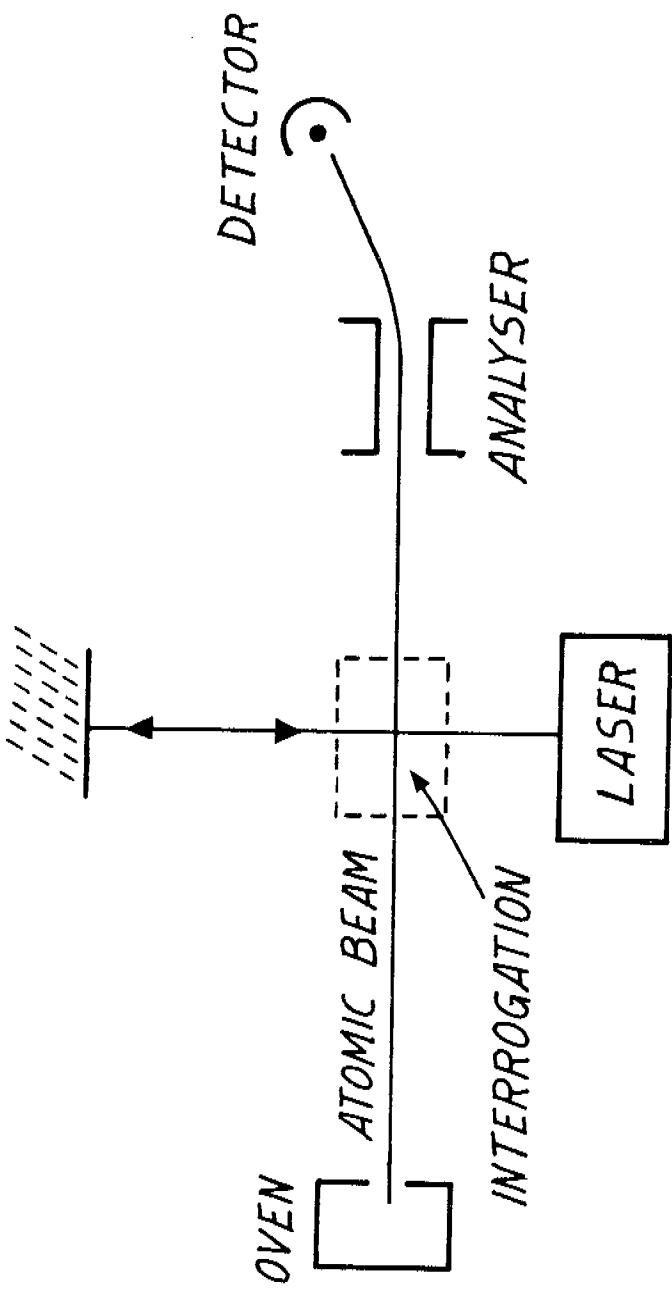


FIGURE 11