Experiments with separated oscillatory fields and hydrogen masers*

Norman F. Ramsey

Department of Physics, Harvard University, Cambridge, Massachusetts 02138

I am honored to receive the Nobel prize, which I feel is also an honor to the physicists and engineers in many countries who have done beautiful experiments using the methods I shall be discussing. In particular, I am grateful to my great teacher, I. I. Rabi, to my eighty-four wonderful Ph.D. students and to Daniel Kleppner and Daniel Larson, who were my close collaborators for a number of years.

THE METHOD OF SUCCESSIVE OSCILLATORY FIELDS

In the summer of 1937, following two years at Cambridge University, I went to Columbia University to work with I. I. Rabi. After I had been there only a few months, Rabi invented (Rabi *et al.*, 1938, 1939; Kellogg *et al.*, 1939, 1940)[1] the molecular-beam magnetic-resonance method so I had the great good fortune to be the only graduate student to work with Rabi and his colleagues on one of the first two experiments to develop and utilize magnetic-resonance spectroscopy, for which Rabi received the 1944 Nobel Prize in Physics.

By 1949, I had moved to Harvard University and was looking for a way to make more accurate measurements than were possible with the Rabi method, and in so doing I invented the method of separated oscillatory fields (Ramsey, 1949, 1950, 1956, 1980, 1983; the last paper contains an extensive list of references). In this method the single oscillatory magnetic field in the center of the Rabi device is replaced by two oscillatory fields, one at the entrance and one at the exit of the space in which the properties of the atoms or molecules are studied. As I will discuss, the separated oscillatory fields method has many advantages over the single oscillatory field method, and in subsequent years it has been extended to many experiments beyond those of molecular-beam magnetic resonance. The device shown in Figure 1 is a molecular-beam apparatus embodying successive oscillatory fields that has been used at Harvard for an extensive series of experiments.

Let me now review the successive oscillatory field

method, particularly in its original and easiest to explain application—the measurement of nuclear magnetic moments. The extension to more general cases is then straightforward.

The method was initially an improvement on Rabi's resonance method for measuring nuclear magnetic moments, whose principles are illustrated schematically in Figure 2. Consider a classical nucleus with spin angular momentum $\hbar \mathbf{J}$ and magnetic moment $\mu = (\mu / J)\mathbf{J}$. Then in a static magnetic field $\mathbf{H}_0 = H_0\mathbf{k}$, the nucleus, due to the torque on the nuclear angular momentum, will precess like a top about $\mathbf{H}_0$, with the Larmor frequency ν_0 and angular frequency ω_0 given by

$$\omega_0 = 2\pi\nu_0 = \frac{\mu H_0}{\hbar I} \ . \tag{1}$$

as shown in Figure 3. Consider an additional magnetic field $\mathbf{H}_1$ perpendicular to $\mathbf{H}_0$ and rotating about it with angular frequency ω. Then, if at any time $\mathbf{H}_1$ is perpendicular to the plane of $\mathbf{H}_0$ and $\mathbf{J}$, it will remain perpendicular to it provided $\omega = \omega_0$. In that case, in a coordinate system rotating with $\mathbf{H}_1$, $\mathbf{J}$ will precess about $\mathbf{H}_1$ and the angle ϕ will continuously change in a fashion analogous to the motion of a "sleeping top;" the change of orientation can be detected by allowing the molecular beam containing the magnetic moments to pass through inhomogeneous fields as in Figure 2. If ω is not equal to ω_0, $\mathbf{H}_1$ will not remain perpendicular to $\mathbf{J}$; so ϕ will increase for a short while and then decrease, leading to no net change. In this fashion the Larmor precession frequency ω_0 can be detected by measuring the oscillator frequency ω at which there is maximum reorientation of the angular momentum and hence a maximum change in beam intensity for an apparatus as in Figure 2. This procedure is the basis of the Rabi molecular-beam resonance method.

The separated oscillatory field method in this application is much the same except that the rotating field $\mathbf{H}_1$ seen by the nucleus is applied initially for a short time τ, the amplitude of $\mathbf{H}_1$ is then reduced to zero for a relatively long time T and then increased to $\mathbf{H}_1$ for a time τ, with phase coherency being preserved for the oscillating fields as shown in Figure 4. This can be done, for example, in the molecular-beam apparatus of Figure 2, in which the molecules first pass through a rotating field region, then a region with no rotating field, and finally a region with a second rotating field driven phase coherently by the same oscillator.

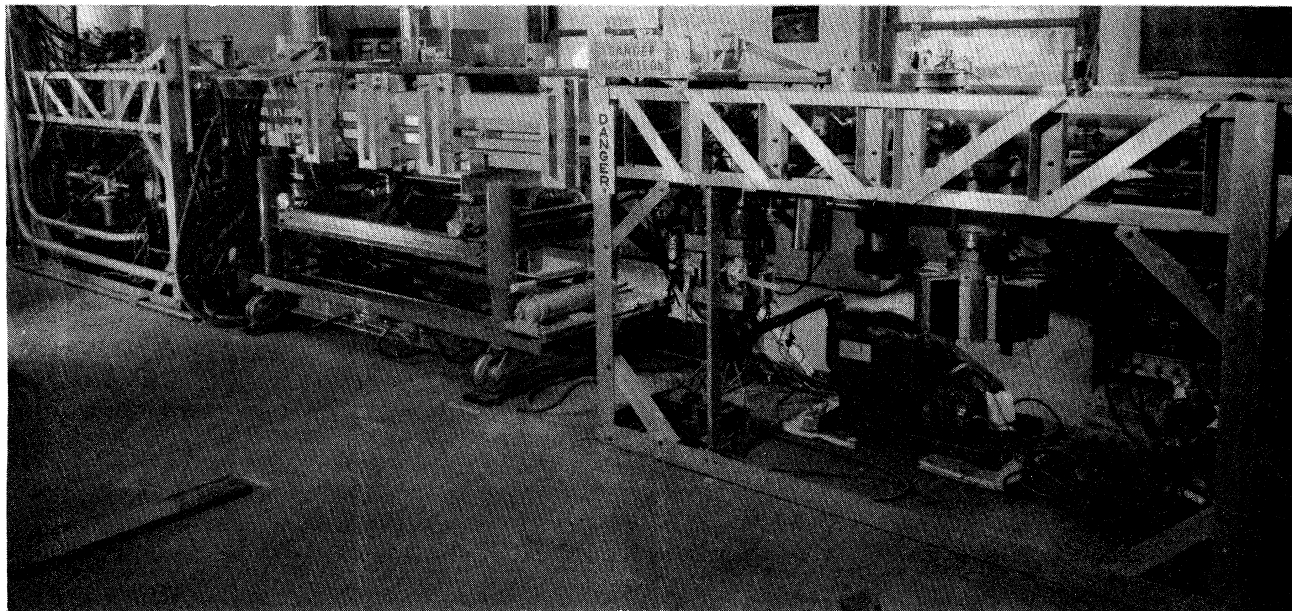If the nuclear-spin angular momentum is initially

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FIG. 1. Molecular-beam apparatus with separated oscillatory fields. The beam of molecules emerges from a small source aperture in the left third of the apparatus, is focussed there, and passes through the middle third in an approximately parallel beam. It is focussed again in the right third to a small detection aperture. The separated oscillatory electric fields at the beginning and end of the middle third of the apparatus produce resonance transitions that reduce the focussing and therefore weaken the detected beam intensity.

parallel to the fixed field (so that ϕ is equal to zero initially), it is possible to select the magnitude of the rotating field so that ϕ is 90° or $\pi/2$ radians at the end of the first oscillating region. While in the region with no oscillating
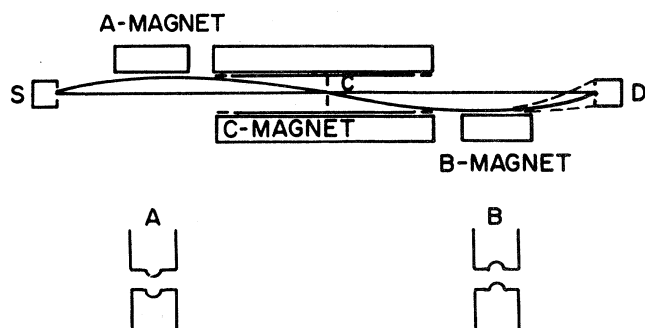

FIG. 2. Schematic diagram of a molecular-beam magnetic-resonance apparatus. A typical molecule which can be detected emerges from the source, is deflected by the inhomogeneous magnetic field A, passes through the collimator, and is deflected to the detector by the inhomogeneous magnetic field B. If, however, the oscillatory field in the C region induces a change in the molecular state, the B magnet will provide a different deflection and the beam will follow the dashed lines with a corresponding reduction in detected intensity. In the Rabi method, the oscillatory field is applied uniformly throughout the C region, as indicated by the long rf lines, whereas in the separated oscillatory field method the rf is applied only at the beginning and end of the C region.

field, the magnetic moment simply precesses with the Larmor frequency appropriate to the magnetic field in that region. When the magnetic moment enters the second oscillating field region, there is again a torque acting to change ϕ. If the frequency of the rotating field is exactly the same as the mean Larmor frequency in the intermediate region there is no relative phase shift between the angular momentum and the rotating field.

Consequently, if the magnitude of the second rotating field and the length of time of its application are equal to those of the first region, the second rotating field has just the same effect as the first one—that is, it increases ϕ by
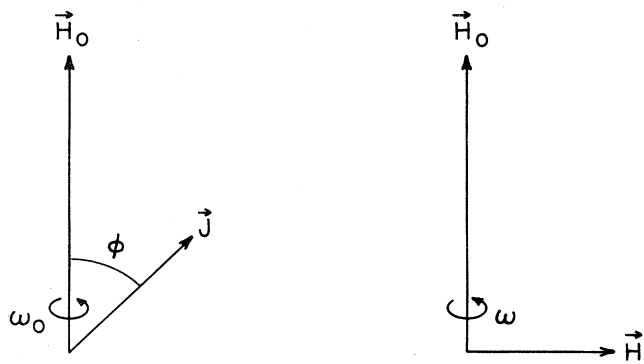

FIG. 3. Precession of the nuclear angular momentum **J** (left) and the rotating magnetic field $\mathbf{H}_1$ (right) in the Rabi method.
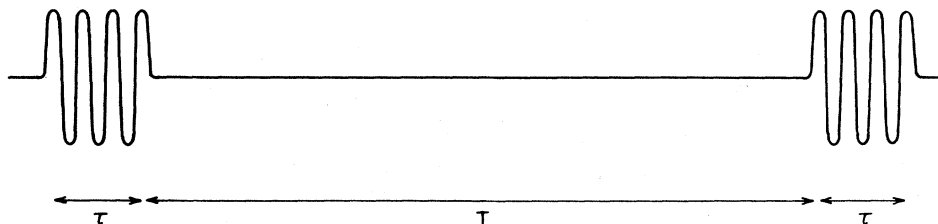
FIG. 4. Two separated oscillatory fields, each acting for a time τ, with zero-amplitude oscillating field acting for time T. Phase coherency is preserved between the two oscillatory fields, so it is as if the oscillation continued, but with zero amplitude for time T.

another $\pi/2$, making $\phi = \pi$, corresponding to a complete reversal of the direction of the angular momentum. On the other hand, if the field and the Larmor frequencies are slightly different, so that the relative phase angle between the rotating field vector and the precessing angular momentum is changed by π while the system is passing through the intermediate region, the second oscillating field has just the opposite effect to the first one; the result is that ϕ is returned to zero. If the Larmor frequency and the rotating field frequency differ by an amount such that the relative phase shift in the intermediate region is exactly an integral multiple of 2π, ϕ will again be left at π just as at exact resonance.

In other words, if all molecules had the same velocity, the transition probability would be periodic as in Fig. 5. However, in a molecular-beam resonance experiment one can easily distinguish between exact resonance and the other cases. In the case of exact resonance, the condition for no change in the relative phase of the rotating field and of the precessing angular momentum is independent of the molecular velocity. In the other cases, however, the condition for an integral multiple of 2π relative phase shift is velocity dependent, because a slower molecule is in the intermediate region longer and so experiences a greater shift than a faster molecule. Consequently, for the non-resonance peaks, the reorientations of most molecules are incomplete, so the magnitudes of the non-resonance peaks are smaller than at exact resonance and one expects a resonance curve similar to that shown in

Figure 6, in which the transition probability for a particle of spin 1/2 is plotted as a function of frequency.

Although the above description of the method is primarily in terms of classical spins and magnetic moments, the method applies to any quantum-mechanical system for which a transition can be induced between two energy states W_i and W_f which are differently focussed. The resonance frequency ω_0 is then given by

$$\omega_0 = (W_i - W_f)/\hbar , \qquad (2)$$

and one expects a resonance curve similar to that shown in Figure 6, in which the transition probability for a particle of spin 1/2 is plotted as a function of frequency.

From a quantum-mechanical point of view, the oscillating character of the transition probability in Figures 5 and 6 is the result of the cross term in the calculation of the transition probability from probability amplitudes. Let C_{iif} be the probability amplitude for the nucleus to pass through the first oscillatory field region with the ini-
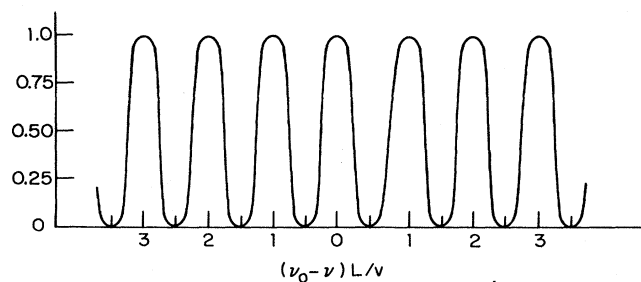

FIG. 5. Transition probability as a function of the frequency $\nu = \omega/2\pi$ that would be observed in a separated oscillatory field experiment if all the molecules in the beam had a single velocity.
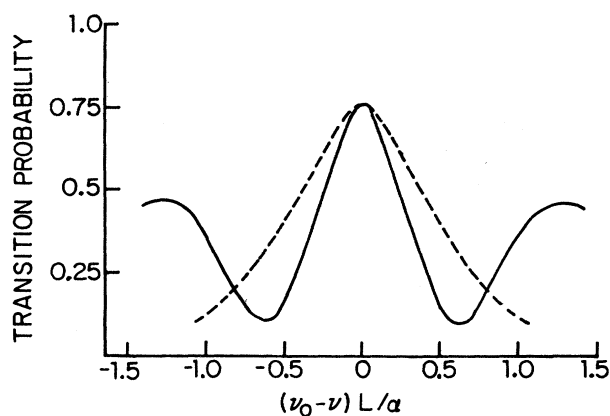

FIG. 6. When the molecules have a Maxwellian velocity distribution, the transition probability is as shown by the solid line for optimum rotating field amplitude. (L is the distance between oscillating field regions, α is the most probable molecular velocity, and ν is the oscillatory frequency $= \omega/2\pi$). The dotted line shows the transition probability for an oscillating field of one-third strength, and the dashed line represents the transition probability with the single oscillating field method when the total duration is the same as the time between separated oscillatory field pulses.

tial state i unchanged, but for there to be a transition to state f in the final field, whereas C_{iff} is the amplitude for the alternative path with the transition to the final state f being in the first field, with no change in the second. The cross term $C_{iff}C_{iff}^*$ produces an interference pattern and gives the narrow oscillatory pattern of the transition probability shown in the curves of Figures 5 and 6. Alternatively the pattern can in part be interpreted as resulting from the Fourier spectrum of an oscillating field that is on for a time τ, off for T, and on again for τ, as in Figure 4. However, the Fourier interpretation is not fully valid, since, with finite rotations of $\mathbf{J}$ the problem is a nonlinear one. Furthermore, the Fourier interpretation obscures some of the key advantages of the separated oscillatory field method. I have calculated the quantum-mechanical transition probabilities (Ramsey, 1949, 1950, 1956, 1958a, 1958b), and these calculations provide the basis for Figure 6.

The separated oscillatory field method has a number of advantages, including the following:

(1) The resonance peaks are only 0.6 as broad as the corresponding ones with the single oscillatory field method. The narrowing is somewhat analogous to the peaks in a two-slit optical interference pattern being narrower than the central diffraction peak of a single wide slit whose width is equal to the separation of the two slits.

(2) The sharpness of the resonance is not reduced by nonuniformities of the constant field, since, both from the qualitative description and from the theoretical quantum analysis, it is only the space average value of the energies along the path that enter Eq. (2) and are important.

(3) The method is more effective and often essential at very high frequencies where the wavelength of the radiation used may be comparable to or smaller than the length of the region in which the energy levels are studied.

(4) Provided there is no unintended phase shift between the two oscillatory fields, first-order Doppler shifts and widths are eliminated.

(5) The method can be applied to study energy levels in a region into which an oscillating field cannot be introduced; for example, the Larmor precession frequency of neutrons can be measured while they are inside a magnetized iron block.

(6) The lines can be narrowed by reducing the amplitude of the rotating field below the optimum. The narrowing is the result of the low amplitude's favoring slower than average molecules.

(7) If the atomic state being studied decays spontaneously, the separated oscillatory field method permits the observation of narrower resonances than those anticipated from the lifetime and the Heisenberg uncertainty principle, provided the two separated oscillatory fields are sufficiently far apart; only states that survive sufficiently long to reach the second oscillatory field can contribute to the resonance. This method, for example, has been used by Lundeen and others (Lundeen, Jessop, and Pip-

kin, 1975; Lundeen and Pipkin, 1975) in precise studies of the Lamb shift.

The advantages of the separated oscillatory field method have led to its extensive use in molecular- and atomic-beam spectroscopy. One of the best known is in atomic cesium standards of frequency and time, which will be discussed later.

Although, in most respects, the separated oscillatory field method offers advantages over a single oscillatory field, there are sometimes disadvantages. In studying complicated overlapping spectra the subsidiary maxima of Figure 6 can cause confusion. Furthermore, it is sometimes difficult at the required frequency to obtain sufficient oscillatory field strengths with two short oscillatory fields, whereas adequate field strength may be achieved with a weaker, longer oscillatory field. Therefore, for most molecular-beam resonance experiments, it is best to have both separated oscillatory fields and a single long oscillatory field available, so the most suitable method under the circumstances can be used.

As in any high-precision experiment, care must be exercised with the separated oscillatory field method to avoid obtaining misleading results. Ordinarily these potential distortions are more easily understood and eliminated with the separated oscillatory field method than are their counterparts in most other high-precision spectroscopy. Nevertheless, the effects are important and require care in high-precision measurements. I have discussed the various effects in detail elsewhere (Ramsey, 1955, 1956, 1958a, 1958b) but I will briefly summarize them here.

Variations in the amplitudes of the oscillating fields from their optimum values may markedly change the shape of the resonance, including the replacement of a maximum transition probability by a minimum. However, symmetry about the exact resonance frequency is preserved, so no measurement error need be introduced by such amplitude variations (Ramsey, 1958a, 1958b).

Variations of the magnitude of the fixed field between, but not in, the oscillatory field regions do not ordinarily distort a molecular-beam resonance provided the average transition frequency (Bohr frequency) between the two fields equals the values of the transition frequencies in each of the two oscillatory field regions alone. If this condition is not met, there can be some shift in the resonance frequency (Ramsey, 1958a, 1958b).

If, in addition to the two energy levels between which transitions are studied, there are other energy levels partially excited by the oscillatory field, there will be a pulling of the resonance frequency as in any spectroscopic study and as analyzed in detail in the literature (Ramsey, 1956, 1958a, 1958b).

Even in the case when only two energy levels are involved, the application of additional rotating magnetic fields at frequencies other than the resonance frequency will produce a net shift in the observed resonance frequency, as discussed elsewhere (Ramsey, 1956, 1958a, 1958b). A particularly important special case is the effect

identified by Bloch and Siegert (1940), which occurs when oscillatory rather than rotating magnetic fields are used. Since an oscillatory field can be decomposed into two oppositely rotating fields, the counter-rotating field component automatically acts as such an extraneous rotating field. Another example of an extraneously introduced rotating field is that which results from the motion of an atom through a field H_0 whose direction varies in the region traversed. The theory of the effects of additional rotating fields at arbitrary frequencies has been developed by Ramsey (1955, 1958a, 1958b), Winter (Ramsey, 1955), Shirley (1963), Code (Code and Ramsey, 1971), and Greene (1970).

Unintended relative phase shifts between the two oscillatory field regions will produce a shift in the observed resonance frequency (Ramsey and Silsbee, 1951; Shirley, 1963; Greene, 1970). This is the most common source of possible error, and care must be taken to avoid it either by eliminating such a phase shift or by determining the shift—say, by measurements with the molecular beam passing through the apparatus first in one direction and then in the opposite direction.

A number of extensions to the separated oscillatory field method have been made since its original introduction:

(1) It is often convenient to introduce phase shifts deliberately to modify the resonance shape (Ramsey and Silsbee, 1951). As discussed above, unintended phase shifts can cause distortions of the observed resonance, but some distortions are useful. Thus, if the change in transition probability is observed when the relative phase is shifted from $+\pi/2$ to $-\pi/2$, one sees a dispersion curve shape (Ramsey and Silsbee, 1951) as in Figure 7. A resonance with the shape of Figure 7 provides maximum sensitivity for detecting small shifts in the resonance frequency.

(2) For most purposes the highest precision can be obtained with just two oscillatory fields separated by the maximum time, but in some cases it is better to use more than two separated oscillatory fields (Ramsey, 1983). The theoretical resonance shapes (Ramsey, 1958b) with two, three, four, and infinitely many oscillatory fields are given in Figure 8. The infinitely many oscillatory field case, of course, by definition becomes the same as the single long oscillatory field if the total length of the transition region is kept the same and the infinitely many oscillatory fields fill in the transition region continuously, as we assumed in Figure 8. For many purposes this is the best way to think of the single oscillatory field method, and this point of view makes it apparent that the single oscillatory field method is subjected to complicated versions of all the distortions discussed in the previous section. It is noteworthy that, as the number of oscillatory field regions is increased for the same total length of apparatus, the resonance width is broadened; the narrowest resonance is obtained with just two oscillatory fields separated the maximum distance apart. Despite this advantage, there are valid circumstances for using more than two os-
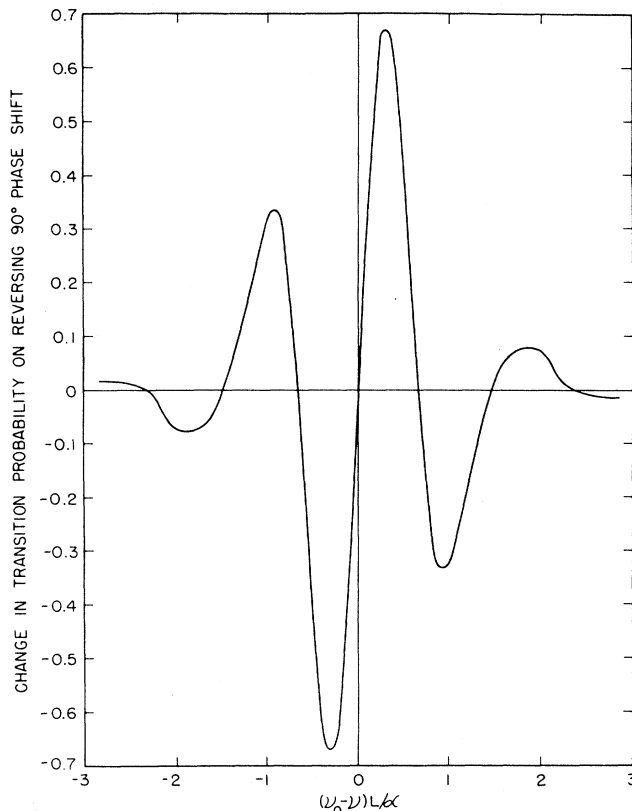

FIG. 7. Theoretical change in transition probability on reversing a $\pi/2$ phase shift. At the resonance frequency there is no change in transition probability, but the curve at resonance has the steepest slope.

cillatory fields. With three oscillatory fields the first and largest side lobe is suppressed, which may help in resolving two nearby resonances; for a larger number of oscillatory fields additional side lobes are suppressed, and in the limiting case of a single oscillatory field there are no side lobes. Another reason for using a large number of successive pulses can be the impossibility of obtaining sufficient power in a single pulse to induce adequate transition probability with a small number of pulses.

(3) The earliest use of the separated oscillatory field method involved two oscillatory fields separated in space, but it was early realized that the method with modest modifications could be generalized to a method of successive oscillatory fields with the separation being in time, say, by the use of coherent pulses (Ramsey, 1957).

(4) If more than two successive oscillatory fields are utilized, it is not necessary to the success of the method that they be equally spaced in time (Ramsey, 1983); the only requirement is that the oscillating fields be coherent—as is the case if the oscillatory fields are all derived from a single continuously running oscillator. In particular, the separation of the pulses can even be random (Ramsey, 1957), as in the case of the large box hy-
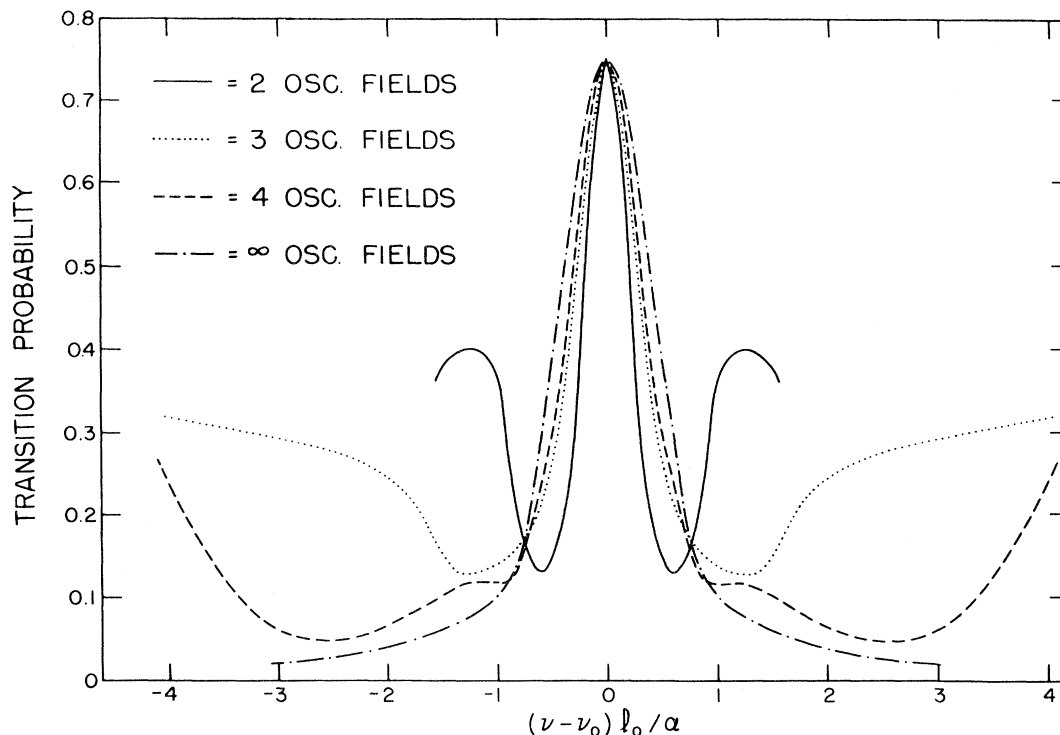
FIG. 8. Multiple oscillatory fields. The curves show molecular-beam resonances with two, three, four, and infinitely many successive oscillating fields. The case with an infinite number of oscillating fields is essentially the same as Rabi's single oscillatory field method ($l_0 = L$).

drogen maser (Uzgiris and Ramsey, 1970) discussed later. The atoms being stimulated to emit move randomly into and out of the cavities with oscillatory fields and spend the intermediate time in the large container with no such fields.

(5) The full generalization of the successive oscillatory field method is excitation by one or more oscillatory fields that vary arbitrarily with time in both amplitude and phase (Ramsey, 1958a, 1958b).

(6) V. F. Ezhov and his colleagues (Ezhov et al., 1976; Ramsey, 1980) in a neutron-beam experiment, used an inhomogeneous static field in the region of each oscillatory field such that, initially, when the oscillatory field is applied, conditions are far from resonance. Then, when the resonance condition is slowly approached, the magnetic moment that was originally aligned parallel to H_0 will adiabatically follow the effective magnetic field on a coordinate system rotating with H_1 until at the end of the first oscillatory field region the moment is parallel to H_1. This arrangement has the theoretical advantage that the maximum transition probability can be unity even with a velocity distribution, but the method may be less well adapted to the study of complicated spectra.

(7) I emphasized earlier that one of the principal sources of error in the separated oscillatory field method is that which arises from uncertainty in the exact value of the relative phase shift in the two oscillatory fields.

Jarvis, et al. (1977) have pointed out that this problem can be overcome with a slight loss in resolution by driving the two cavities at slightly different frequencies, so that there is a continual change in the relative phase. In this case the observed resonance pattern will change continuously from absorption to dispersion shape. The envelope of these patterns, however, can be observed, and the position of the maximum of the envelope is unaffected by relative phase shifts. Since the envelope is about twice the width of a specific resonance, there is some loss of resolution in this method, but in certain cases this loss may be outweighed by the freedom from phase shift errors.

(8) The method has been extended to electric as well as magnetic transitions and to optical laser frequencies as well as radio and microwave frequencies. The application of the separated oscillatory field method to optical frequencies requires considerable modifications because of the short wavelengths, as pointed out by Blaklanov, Dubetsky and Chebotsev (1976). Successful applications of the separated oscillatory field method to lasers have been made by Bergquist, Lee, and Hall (1977, 1978), Salour (1978a, 1978b, 1978e), Cohen-Tannoudji, (Salour and Cohen-Tannoudji, 1977), Bordé (1977), Hänsch (1978), Chebotayev (Chebotayev et al., 1987), and many others.

(9) The method has been extended to neutron beams

and to neutrons stored for long times in totally reflecting bottles.

(10) In a recent beautiful experiment, S. Chu and his associates (Kasevich *et al.*, 1989), have successfully used the principle of separated oscillatory fields with a fountain of atoms that rises up slowly, passes through an oscillating field region, falls under gravity, and passes again through the same oscillatory field region. This fountain experiment was attempted many years ago by J. R. Zacharias and his associates (see Ramsey, 1956, for an account), but it was unsuccessful because of the inadequate number of very slow atoms. Chu and his collaborators used laser cooling (Wineland and Dehmelt, 1973, 1975; Hänsch and Schawlow, 1975; Letokhov, 1977; Wineland and Itano, 1987) to slow the atoms to a low velocity and obtained a beautifully narrow separated oscillatory fields resonance pattern.

THE ATOMIC HYDROGEN MASER

The atomic hydrogen maser grew out of my attempts to obtain even greater stability in atomic-beam experiments. By the Heisenberg uncertainty principle (or by the Fourier transform), the width of a resonance in a molecular-beam experiment cannot be less than approximately the reciprocal of the time the atom is in the resonance region of the appratus. For atoms moving through a 1-m-long resonance region at 100 m/s, this means that the resonance width is about 100 Hz wide. To decrease this width and hence increase the precision of the measurements required an increase in this time. To increase the time by drastically lengthening apparatus or selecting slower molecules would decrease the already marginal beam intensity or greatly increase the cost of the apparatus. I therefore decided to plan an atomic beam in which the atoms, after passing through the first oscillatory field, would enter a storage box with suitably coated walls where they would bounce around for a period of time and then emerge to pass through the second oscillatory field. My Ph.D. student, Daniel Kleppner

(Kleppner *et al.*, 1958), undertook the construction of this device as his thesis project. The original configuration required only a few wall collisions and was called a broken-beam resonance experiment. Initially the beam was cesium and the wall coating was Teflon. The experiment (Kleppner *et al.*, 1958) was a partial success in that a separated oscillatory field pattern for an atomic hyperfine transition was obtained, but it was weak and disappeared after a few wall collisions. The results improved markedly when a paraffin was used for the wall coating, and a hyperfine resonance was eventually obtained after 190 collisions, giving a resonance width of 100 Hz, but with the resonance frequency shifted by 150 Hz.

To do much better than this, we decided we would have to use an atom with a lower mass and a lower electric polarizability to reduce the wall interactions. Atomic hydrogen appeared ideal for this purpose, but atomic hydrogen is notoriously difficult to detect. We therefore calculated the possibility of detecting the transitions through their effects on the electromagnetic radiations. Townes (Gordon, Geiger, and Townes, 1954, 1955) had a few years earlier made the first successful maser (acronym for microwave amplifier by stimulated emission of radiation), but no one had previously made a maser based on a magnetic dipole moment or at a frequency as low as that of an atomic hyperfine transition. We concluded, however, that if the resonance could be made narrow enough by multiple wall collisions, we should be able to obtain maser oscillations. The apparatus was designed and constructed by Goldenberg, Kleppner, and myself (Goldenberg *et al.*, 1960; Kleppner *et al.*, 1962), and after a few failures we obtained maser oscillations at the atomic hydrogen hyperfine frequency. Both the proton and the electron have spin angular momenta $\mathbf{I}$ and $\mathbf{J}$ as well as magnetic moments. The atomic hyperfine transitions are those for which there is a change of the relative orientation of these two magnetic moments between the initial and final states in Eq. (2). We studied H atoms in the $1^2S_{1/2}$ ground electronic state and mostly observed the transitions ($F=1$, $m=0 \rightarrow F=0$, $m=0$), where F is the
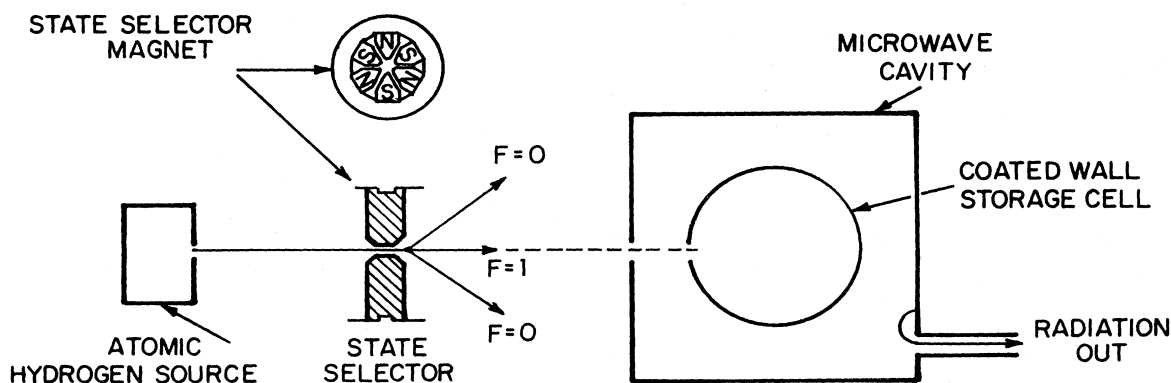

FIG. 9. Schematic diagram of atomic hydrogen maser. Only the paths of the $m=0$ atoms are shown, since the $m=1$ atoms are not involved in the $\Delta m=0$ transitions studied.

quantum number of the total angular momentum $F = I + J$ and m is the associated magnetic quantum number.

The principles of an atomic hydrogen maser are shown schematically in Figure 9. An intense electrical discharge in the source converts commericially available molecular hydrogen (H_2) into atomic hydrogen (H). The atoms emerge from the source into a region that is evacuated to 10^{-6} torr and enter a state-selecting magnet which has three north poles alternating in a circle with three south poles. By symmetry, the magnetic field is zero on the axis and increases in magnitude away from the axis. Since the energy of a hydrogen atom in the $F = 1$, $m = 0$ state increases with energy, and since mechanical systems are accelerated toward lower potential energy, an atom in the $F = 1$ state that is slightly off axis will be accelerated toward the axis, i.e., the $F = 1$ state will be focussed onto the small aperture of the 15-cm-diameter storage cell, whereas the $F = 0$ state is defocussed. As a result, if the atomic beam flows steadily, the storage bottle in equilibrium will contain more high-energy $F = 1$ atoms than low. If these atoms are exposed to microwave radiation at the hyperfine frequency, more atoms are stimulated to go from the higher-energy state to the lower one than in the opposite direction. Energy is then released from the atoms and makes the microwave radiation stronger. Thus the device is an amplifier or maser. If the storage cell is placed inside a tuned cavity, an oscillation at the resonance frequency will increase in magnitude until an equilibrium value is reached. At this level the oscillation will continue indefinitely, with the energy to maintain the oscillation coming from the continuing supply of hydrogen atoms in the high-energy hyperfine state. The device then becomes a free-running maser oscillator at the atomic hyperfine frequency.

The atomic hydrogen maser oscillator has unprecedented high stability due to a combination of favorable features. The atoms typically reside in the storage cell for 10 seconds, which is much longer than in an atomic-beam resonance apparatus, so the resonance line is much narrower. The atoms are stored at low pressure, so they are relatively free and unperturbed while radiating. The first-order Doppler shift is removed, since the atoms are exposed to a standing wave and since the average velocity is extremely low for atoms stored for 10 seconds. Masers have very low noise levels, especially when the amplifying elements are isolated atoms. Over periods of several hours the hydrogen maser stability is better than 1×10^{-15}.

The major disadvantage of the hydrogen maser is that the atoms collide with the walls at intervals, changing slightly the hyperfine frequency and giving rise to wall shifts of 1×10^{-11}. However, the wall shifts can be experimentally determined by measurements utilizing storage bottles of two different diameters or with a deformable bulb whose surface-to-volume ratio can be altered. As in all precision measurements, care must be taken in adjusting and tuning the hydrogen maser to

avoid misleading results. These limitations and precautions are discussed in a series of publications by various authors (Goldenberg et al., 1960; Kleppner et al., 1962, 1965; Koelman et al., 1988[2]). The designs of hydrogen masers have been modified in many ways either for special purposes or for increased stability and reliability. For example, different hyperfine transitions have been used and masers have been operated in relatively strong magnetic fields. A hydrogen maser has also been operated (Uzgiriś and Ramsey, 1970) with a storage bottle that is much larger than the wavelength of the stimulating radiation by confining the microwave power to two small cavities, so that it functions as a separated oscillatory field device. As shown in Figure 10, the atoms that are stimulated to emit radiation move randomly into and out of the two oscillatory field cavities and spend the intermediate time in the large container, where there is no oscillatory field. Due to the large size of the storage box, there are longer storage times and less frequent wall collisions, so the resonances are narrower and the wall shifts are smaller than for a normal hydrogen maser.

PRECISION SPECTROSCOPY

Now that I have discussed extensively the principles of the separated oscillatory field method and of the atomic maser, I shall give some illustrations as to their value. One major category of applications is to precision spectroscopy, especially at radio and microwave frequencies. Another category of applications is to atomic clocks and frequency standards.

It is difficult to summarize the spectroscopic applications, since there are so many of them. Many beautiful experiments have been done by a large number of scientists in different countries, including Sweden. I shall, therefore, use just a few illustrations from experiments in which I have been personally involved.

My graduate students have made precision measurements of the radiofrequency spectra of different molecules in various rotational states. For each of these states more than seven different molecular properties can be inferred, and thus the variations of the properties with changes in the rotational and vibrational quantum numbers can be determined. These properties include nuclear and rotational magnetic moments, nuclear quadrupole interactions, nuclear spin-spin magnetic interactions, spin rotational interactions, etc. I shall illustrate the accuracy and significance of the measurements with a single example. With both D_2 and LiD we have accurately measured (Code and Ramsey, 1971; Freeman et al., 1975) the deuteron quadrupole interaction eqQ, where e is the pro-

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[2]This paper contains an extended series of references to other papers on hydrogen maser limitations, principles, and practices.
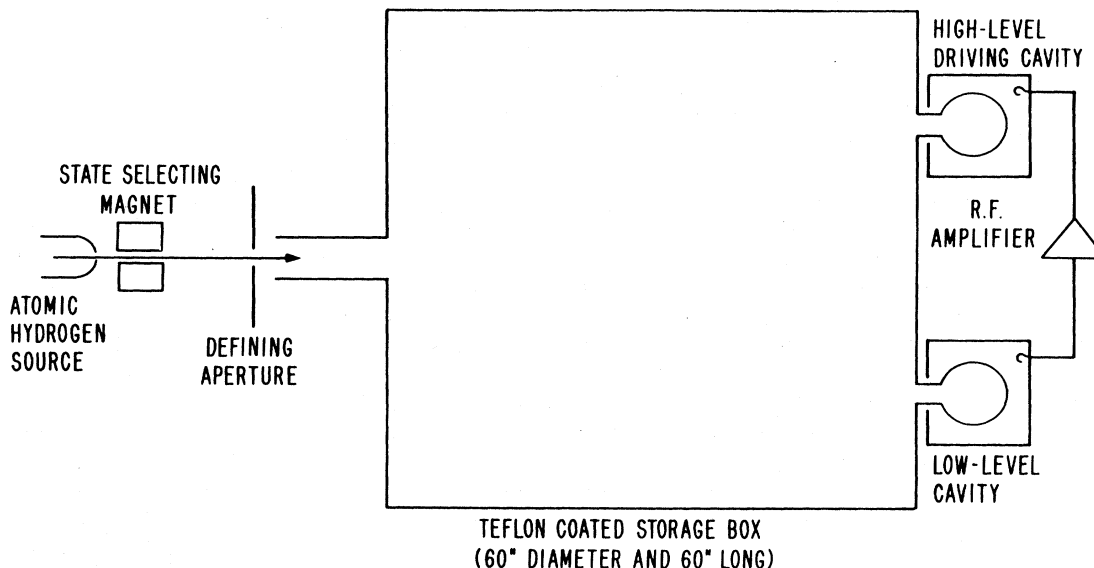
FIG. 10. Schematic diagram of a large box hydrogen maser. The two cavities on the right act as two separated oscillating fields with that of the high-level cavity being obtained by amplification from the low.

ton electric charge, q is the gradient of the molecular electric field at the deuteron, and Q is the deuteron quadrupole moment which measures the shape of the deuteron and in particular its departure from spherical symmetry. These measurements were made with a high-resolution molecular-beam apparatus based on the method of separated oscillatory fields. We found for eqQ the value $+225\,044\pm20$ Hz in D_2 and $+34\,213\pm33$ Hz in LiD. Since q has been calculated (Reid and Vaida, 1973; Docken and Freeman, 1974) for each of these quite different molecules, two independent values of Q can be calculated. The results agree to within 1.5%, which confirms the validity of the difficult calculation; with it we find $Q = 2.9 \times 10^{-27}$ cm^2.

In an experiment with collaborators (Green et al., 1979) at the Institut Laue-Langevin at Grenoble, France, we have used the separated oscillatory field method with a beam of slow neutrons to make an accurate measurement of the neutron magnetic moment and found (Reid, and Vaida, 1973; Cohen and Taylor, 1987) it to be $-1.913\,042\,75\pm0.000\,000\,45$ nuclear magnetons. In a somewhat different experiment with neutrons moving so slowly that they can be bottled for more than 80 s in a suitable storage vessel, we have used the method of successive oscillatory fields with the two coherent radiofrequency pulses being separated in time rather than space. In this manner and as a fundamental test of time reversal symmetry, we have recently set a very low upper limit for the neutron electric dipole moment by finding (Smith et al., 1984, 1989) its value to be $(-3\pm5)\times10^{-26}$ e cm.

The atomic hydrogen maser gives very accurate data on the microwave spectrum of the ground electronic state of the hydrogen atom. The hyperfine frequency Δv for atomic hydrogen has been measured in our laboratory

and in a number of other laboratories. The best value (Hellwig et al., 1970; Essen et al., 1971) is

$$\Delta v_H = 1\,420\,405\,751.7667\pm0.0009 \text{ Hz .}$$

This value agrees with present quantum-electromagnetic theory (Baldwin and Yennie, 1988) to within the accuracy of the theoretical calculation and can be used to obtain information on the proton structure. Similarly accurate values have been found for atomic deuterium and tritium, and the dependence (Gibbons and Ramsey, 1972) of these results on the strengths of externally applied electric fields have been measured. With a modified form of the hydrogen maser designed to operate at high magnetic fields, the ratio of the magnetic moment of the electron to that of the proton is found (Winkler et al., 1972; Cohen and Taylor, 1987; Particle Data Group, 1988) to be $-658.210\,688\pm0.000\,006$. Incidentally, when this result is combined with the beautiful electron measurements from Professor Dehmelt's laboratory (Van Dyck, Schwinberg, and Dehmelt, 1984; Van Dyck et al., 1986; Cohen and Taylor, 1987), we obtain the best values for the free proton magnetic moment in both Bohr and nuclear magnetons.

ATOMIC CLOCKS

In the past 50 years there has been a major revolution in time keeping, with the accuracy and reproducibility of the best clocks at the end of that period being approximately a million times those at the beginning. This revolution in time keeping and frequency control is due to atomic clocks.

Any clock or frequency standard depends on some reg-

ular periodic motion, such as the pendulum of the grandfather's clock. In the case of atomic clocks the periodic motion is internal to the atoms and is usually that associated with an atomic hyperfine structure, as discussed in the section on the atomic hydrogen maser.

In the most widely used atomic clocks, the atom whose internal frequency provides the periodicity is cesium, and the usual method of observing it is with a separated oscillatory field magnetic-resonance apparatus as in Figure 2. The first commercial cesium-beam clock was developed in 1955 by a group led by J. R. Zacharias (see Ramsey, 1983), and in the same year L. Essen and V. L. Parry (see Ramsey, 1983) constructed and operated the first cesium-beam apparatus that was extensively used as an actual frequency standard. Subsequently many scientists and engineers throughout the world contributed to the development of atomic clocks, as discussed in greater detail elsewhere (Ramsey, 1983; this paper contains an extensive list of references).

Cesium atomic clocks now have an accuracy and stability of about 10^{-13}, which was so far superior to all previous clocks that in 1967 the internationally adopted definition of the second was changed from one based on motion of the earth around the sun to 9,192,631,770 periods of the cesium atom.

For many purposes even greater stability is required over shorter time intervals. When such stability is needed, the hydrogen maser is frequently used with a stability of 10^{-15} over periods of several hours.

Atomic clocks based on the above principles have for a number of years provided clocks of the greatest stability and accuracy, and these are sufficiently great that further improvements might seem to be neither desirable nor feasible. But as we shall see in our final section, there are applications that already push atomic clocks to their limits, and there are many current developments with great promise for the future. These include improvements to the existing devices, use of higher frequency, use of lasers, electromagnetic traps for storing both ions and atoms, laser cooling, etc.

APPLICATIONS FOR ACCURATE CLOCKS

Accurate atomic clocks are used for so many different purposes that a list of them all is tediously long, so I shall here just briefly mention a few that push clock technology to its limit.

In radio astronomy one looks with a parabolic reflector at the radio waves coming from a star, just as in optical astronomy one looks with an optical telescope at the light waves coming from a star. Unfortunately, in radio astronomy the wavelength of the radiation is about a million times longer than the wavelength of light. The resolution of the normal radio telescope is therefore about a million times worse, since the resolution of a telescope depends on the ratio of the wavelength to the telescope aperture. However, if there are two radio telescopes on opposite sides of the earth looking at the same star and if the radio waves entering each are matched in time, it is equivalent to a single telescope whose aperture is the distance between the two telescopes, and the resolution of such a combination exceeds that of even the largest single optical telescope. However, to do such precise matching in time, each of the two radio telescopes needs a highly stable clock, usually an atomic hydrogen maser.

One of the exciting discoveries in radio astronomy has been the discovery of pulsars, that emit their radiation in short periodic pulses. Precision clocks have been needed to measure the pulsar periods and the changes in the periods with time; these changes sometimes occur smoothly and sometimes abruptly. Of particular interest from the point of view of time measurements are the millisecond pulsars, which have remarkable constancy of period, rivaling the stability of the best atomic clocks. (Taylor et al., 1979; Davis et al., 1985). Another millisecond pulsar is part of a rapidly rotating binary star that is slowly changing its period of rotation (Taylor et al., 1979; Davis et al., 1985). This slow change in rotation can be attributed to the loss of energy by the radiation of gravity waves—the first experimental evidence for the existence of gravity waves.

Time and frequency can now be measured so accurately that wherever possible other fundamental measurements are reduced to time or frequency measurements. Thus the unit of length by international agreement has recently been defined as the distance light will travel in a specified time, and voltage will soon be represented in terms of frequency measurements.

Accurate clocks have provided important tests of both the special and the general theories of relativity. In the experiment of Vessot et al. (1980), a hydrogen maser was shot in a rocket to a 6000-mile altitude, and its periodic rate changed with speed and altitude just as expected by the special and general theories of relativity. In other experiments, observers have measured the delays predicted by relativity for radio waves passing near the sun.

Precision clocks make possible an entirely new and more accurate navigational system, the global positioning system or GPS. A number of satellites containing accurate atomic clocks transmit signals at specific times, so any observer receiving and analyzing the signals from four such satellites can determine his position to within ten yards and the correct time within one hundreth of a millionth of a second (10^{-8} s).

A particularly fascinating navigation feat dependent on accurate clocks was the recent and highly successful tour of the Voyager spacecraft to Neptune. The success of this mission depended upon the ground controllers' having accurate knowledge of the position of the Voyager. This was accomplished by having three large radio telescopes at different locations on the earth, each of which transmitted a coded signal to Voyager, which in turn transmitted the signals back to the telescopes. The distances from each telescope to Voyager could be determined from the elapsed times, and thus Voyager could be

located. To achieve the required timing accuracy, two hydrogen masers were located at each telescope. Due to the rotation of the earth in the eight hours required for the electromagnetic wave to travel from the earth to *Voyager* and back again at the speed of light, the telescope transmitting the signal in some cases had to be different from the one receiving; this placed an additional stringent requirement on the clocks. Thus the spectacular success of the *Voyager* mission was dependent on the availability of highly stable clocks.

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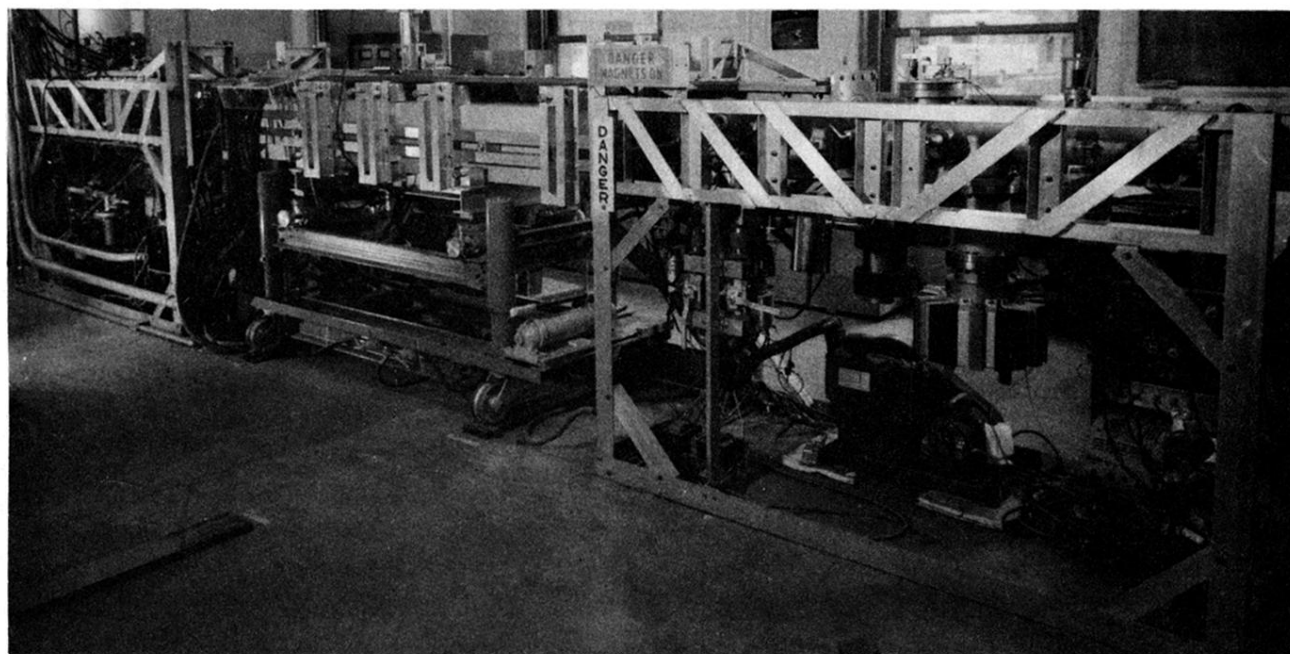
FIG. 1. Molecular-beam apparatus with separated oscillatory fields. The beam of molecules emerges from a small source aperture in the left third of the apparatus, is focussed there, and passes through the middle third in an approximately parallel beam. It is focussed again in the right third to a small detection aperture. The separated oscillatory electric fields at the beginning and end of the middle third of the apparatus produce resonance transitions that reduce the focussing and therefore weaken the detected beam intensity.