

USE OF SLOW MOLECULES IN A MASER

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Several methods for improving the absolute frequency stability of a maser are considered; these methods are based on the use of molecular beams in which the mean velocity is much lower than the thermal velocity at room temperature.

It has been shown experimentally¹ that a maser can have a frequency stability as high as 10^{-13} – 10^{-14} for a period of several seconds and 10^{-11} – 5×10^{-12} for a time of the order of thirty minutes. However, when a conventional system (ammonia inversion line, $J = 3$, $K = 3$) is used as an absolute standard of frequency the accuracy is 10^{-9} – 5×10^{-10} , a figure which is appreciably lower than the relative stability.²⁻⁴ For this reason it is desirable to investigate the possibility of building a system with an absolute stability of the order of 10^{-11} , even if it could not provide prolonged operation. A system of this kind could provide intermittent frequency control of an ordinary maser which would operate for extended periods; in this way it might be possible to maintain the frequency for appreciable periods of time, because of the high relative stability.

The theory of the maser^{5,6} indicates that the absolute frequency stability is inversely proportional to the line width and that the generated frequency does not differ from the frequency of the line by more than several percent of the line width. This deviation is determined by the accuracy with which the cavity is tuned to the frequency of the line, the dependence of line shape on external parameters, the line shape and so on.⁷ In particular, the better the adjustment of the cavity to the line frequency, the higher the absolute stability of the system.

In this paper we report on a new method of increasing absolute stability in which the line is narrowed by the use of "slow molecules." Three methods of obtaining slow molecules are considered: 1) removal of high-velocity molecules from the beam, i.e., a high velocity "cutoff"; 2) retardation of the molecules by an external field; 3) reduction of the temperature of the molecular beam.

1. A high velocity cutoff can be imposed on the beam by reflecting the ammonia molecules from a potential barrier. An inhomogeneous electric field serves as a convenient potential barrier.

The interaction of the ammonia molecules with the field is such that the energy of molecules in the upper inversion state is increased while the energy of molecules in the lower inversion state is reduced. Hence, if an ammonia molecule encounters a potential barrier in the form of an electric field it is reflected from the barrier if it is in the upper state and passes through the barrier if it is in the lower state.

Reflection from a barrier is an effective means of separating molecules. The potential barrier can be chosen in such a way that only low-velocity molecules are reflected, since the velocity of the reflected molecules must satisfy the relation

$$\frac{1}{2} m (vn)^2 \leq W, \quad (1)$$

where \mathbf{n} is a unit vector in the direction of $\text{grad } W$. In this case the field must be such that the isoenergetic surfaces $W(\mathbf{r}) = \text{const.}$ act as focusing surfaces. By placing the source at the focus it is then possible to obtain a reflected beam with a small aperture, starting with an incident beam of large aperture.

A maser of this type can be realized most easily with deuterated ammonia (ND_3) because in this case the input aperture in the cavity can be large.⁸ A potential barrier of the type discussed above can be produced by means of two flat plates, one of which has an aperture (fig. 1). The plates are arranged so that optimum focusing is achieved. A rough calculation, in which ideal reflection is assumed, indicates that when deuterated ammonia is used the excitation factor remains the same as in

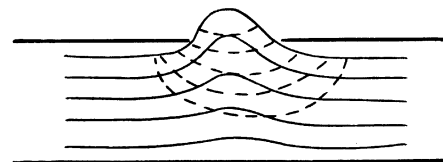


FIG. 1. The solid lines are equipotentials of the fields. The dashed lines are lines of equal potential energy for the interaction of molecules with the field.

a conventional system⁸ if the mean velocity of the molecules in the beam is reduced by a factor of 10.

2. A potential barrier can also be used to retard molecules. If a molecule in the upper inversion state enters an electric field in which the interaction energy is W , then its velocity is reduced from v_0 to v so that the total energy is conserved,

$$\frac{1}{2} m v_0^2 = \frac{1}{2} m v^2 + W. \quad (2)$$

The maximum possible number of molecules is obtained if molecules whose velocities are close to the most probable velocity are slowed down. This velocity is given approximately by $(kT/m)^{1/2}$.

But electric fields which can be realized in practice are such that $W_{\max} = 0.01 kT$ for $T \approx 300^\circ K$. Hence, a single retardation interaction does not reduce the velocity of the molecules appreciably. However, it is possible to use a multiple retardation system, in which regions of electric field are separated by regions of zero field. A molecule in the upper inversion state is decelerated on entering the field. On leaving the field it is accelerated to its original velocity. If, however, the molecule is caused to make a transition to the lower state by means of external radiation while it is in the field, it is again decelerated when it leaves the field. An external radiation field can then be used to raise the molecule to the upper level again and the process can be repeated.

If the frequency of the molecular transition is the same as the frequency of the external radiation, then the probability w of a transition from the upper level to the lower level (which is equal to the probability in the reverse direction) is given by⁵

$$w = \sin^2(\gamma\tau_v/2), \quad \gamma = |d_{12}| E/\hbar, \quad (3)$$

where τ_v is the time of flight of a molecule with velocity v and E is the amplitude of the external radiation. Hence, the total number of retarded molecules at the output of the system is given by

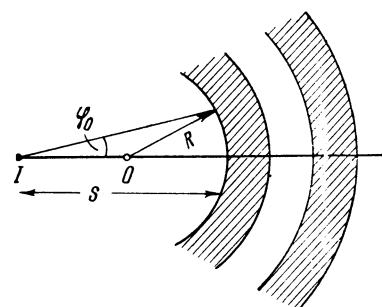
$$N = N_0 \int_{v_0-\Delta}^{v_0+\Delta} \left(\prod_{k=1}^n \sin^2 \frac{\gamma_k \tau_{vk}}{2} \right) F(v) dv \Big/ \int_0^\infty F(v) dv, \quad (4)$$

where $F(v)$ is the velocity distribution of the molecules, v_0 is the most probable velocity in $F(v)$, 2Δ is the velocity range of the retarded molecules and N_0 is the original number of molecules.

If we wish to make the mean velocity in the retarded beam one-tenth the mean thermal velocity (roughly v_0), $\Delta/v_0 \approx 0.1$. In order to achieve this reduction it is necessary to have approximately fifty retardation sections, since $n \approx 100$. If the amplitude of the auxiliary field is chosen to

make $\sin^2(\gamma_k \tau_{vk}/2) = 1$ for the mean velocity (which varies from section to section because of the retardation effect), a calculation of the integral in Eq. (4) with an electronic computer (Maxwellian velocity distribution) indicates that $N \approx 0.01 N_0$. The use of retarding sections with uniform fields (parallel plates) is not desirable because these reduce the velocity component perpendicular to the plane of the plate and defocus the molecular beam itself. However, if the retarding sections are spherical mirrors, the system can be set up in such a way as to focus the beam, as can be seen from Fig. 2.

FIG. 2. To obtain focusing the distance S must be larger than the distance R . The point denoted by O is the center of the spherical focusing mirrors; I is the source of the molecular beam and regions with electric field are denoted by cross-hatching.



In this system the initial flight angle ϕ_0 is changed to $\pi - \phi_0$ according to the retardation. As soon as the exit angle became equal to $\pi - \phi_0$ the retardation process must be repeated identically if the first retardation is inadequate.

3. We now consider a method for reducing the temperature of the molecular beam. It is not feasible to reduce the temperature by cooling the source because a small reduction in source temperature has very little effect (the velocity is proportional to $T^{1/2}$). On the other hand, a large reduction in the source temperature causes the ammonia to freeze. It is possible, however, to reduce the ammonia temperature by allowing the ammonia molecules to collide with a cold gas. If the ammonia interacts with the cold gas solely through collisions the kinetic energy of the ammonia molecules is gradually reduced to that of the cold gas. Helium at a few degrees Kelvin serves as a convenient cooling gas. In order for a system of this kind to operate effectively, the following conditions must be satisfied: a) the time in which equilibrium is established in the helium-ammonia mixture must be shorter than the time between collisions of the ammonia molecules or collisions of the ammonia molecules with the walls; b) an effective means of heat removal must be provided so that the temperature of the helium does not rise.

Condition a) implies the relation

$$N_A \sigma_{A-A} / \sqrt{m_A} \ll N_H \sigma_{A-H} / \sqrt{2\mu_{A-H}}, \quad (5)$$

where σ_{A-A} is the cross section for a collision between ammonia molecules, σ_{A-H} is the cross section for a collision between an ammonia molecule and a helium molecule, m_A is the mass of the ammonia molecule, μ_{A-H} is the reduced mass of the system, consisting of the ammonia molecule and the helium molecule, and N_A and N_H are the ammonia and helium densities respectively. From the conditions for thermal equilibrium we can obtain the temperature of the mixture:

$$T = T_H + T_A N_A / N_H, \quad (6)$$

where T_H is the initial temperature of the helium and T_A is the initial temperature of the ammonia. Condition b) requires that the increment in the helium temperature be small ($\sim 1^\circ\text{K}$). Both of these conditions can be satisfied if N_A/N_H is about 1/100.

One important point should be noted. If the ammonia molecules are in thermodynamic equilibrium at 4–5°K they are practically all in the ground state ($J = 0, K = 0$); this state cannot be used for generation since the dipole-moment matrix element for this state is zero. However, this situation is alleviated by the fact that the equilibrium time for the rotational energy is much greater than the equilibrium time for the translational motion because transitions with $\Delta K \neq 0$ are highly forbidden, even in collisions between molecules.⁹

Removal of slow molecules from the beam by a cutoff technique or retardation by external fields disturbs the molecular velocity distribution and this effect causes a change in the line shape. Since the frequency depends on the line shape, at first glance it might be supposed that these methods of obtaining slow molecules are not very useful. An analysis of this aspect of the problem indicates, however, that, in both cases considered above the absolute frequency stability increases for a given excitation factor linearly with the mean time spent by molecules in the cavity.

It should be noted that all these methods of obtaining slow molecules imply an appreciable loss in the number of useful molecules and a consequent reduction in power. Thermal noise in the maser is characterized by a frequency spectrum whose width is inversely proportional to power.¹⁰ An appreciable loss of power can lead to a marked increase in the relative effect of this noise; hence, for effective operation the cavity should be cooled. If the mean time of flight is increased by a factor of ten through the use of a velocity cutoff the thermal noise at room temperature remains unimportant ($\Delta\omega/\omega \approx 10^{-14}$).

The considerations given above indicate that it may be possible to build a maser with an absolute stability of $10^{-11} - 10^{-12}$.

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