

**Zeitschrift:** Helvetica Physica Acta  
**Band:** 37 (1964)  
**Heft:** III  
  
**Artikel:** Multiple cavity molecular beam maser  
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**DOI:** <https://doi.org/10.5169/seals-113482>

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## Multiple Cavity Molecular Beam Maser

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*Abstract:* The theory of multiple cavity molecular beam oscillators is developed. It is shown that the frequency stability of the oscillator can be increased to any required degree by coupling empty resonators to the cavity containing the beam. The coupling constants are determined. The beam intensity required to operate the device increases proportionally to the number of coupled cavities, thus the frequency stability is ultimately limited by the maximum available beam intensity.

*Zusammenfassung:* Die Theorie der Molekularstrahl-Oszillatoren mit mehreren Hohlräumen wird entwickelt. Es wird gezeigt, dass die Frequenzstabilität der Oszillatoren beliebig verbessert werden kann durch Ankoppeln an weitere Hohlräume. Die Kopplungskonstanten werden bestimmt. Die Strahlintensität, welche nötig ist, um den Maser anzutreiben, nimmt proportional zur Zahl der Hohlräume zu. Die Frequenzstabilität wird letzten Endes durch die verfügbare Strahlintensität bestimmt.

### Introduction

Molecular beam masers have been developed for use as high stability frequency etalons. It is also intended to use them as primary time standards. For this purpose the maser frequency must be stable to  $10^{-11}$  parts or better over extended periods of time.

Although the natural width of the molecular lines used to operate the maser (e.g., an inversion line of ammonia) would permit a frequency stability of  $10^{-18}$ , pulling effects reduce the frequency stability of the maser oscillator below the required limit.

Bx 'pulling' one denotes the shift of the maser frequency due to the detuning of the cavity, to the change in the number and velocity of the molecules entering the cavity in unit time, and possibly to other effects.

These effects are, in turn, due to the changes in cavity temperature, in focuser voltage, in source pressure, etc. To limit these changes, many physical parameters of the maser have to be kept under severe control. Several laboratories are engaged in the construction of high stability molecular beam oscillators<sup>1)</sup>.

Hence, it is desirable to employ methods which generally reduce the dependence of the maser frequency on these parameters.

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\*) The major part of this work was completed while the author was on leave at the Florida State University, Tallahassee, Florida.

It has been discovered<sup>2)</sup> that a twin cavity maser exhibits considerably less pulling than a single cavity system, if the coupling between the two cavities is unity. The twin cavity maser consists of two identical cavities coupled by an iris in their common wall. The beam is directed through one of the cavities; no beam is sent through the other cavity.

In view of the success of the two cavity maser the question arose whether a multiple cavity system would be even more advantageous to reduce pulling. The theory of multiple-cavity systems is developed in sections 3 and 4. The results of this theory answer the question in the affirmative, as shown in sections 5 and 6.

## 2. Description of the Molecular Beam Maser

The molecular beam maser, as developed by GORDON, ZEIGER and TOWNES<sup>3)</sup> operates on the following principle. A beam of molecules in an excited energy state is introduced into a cavity tuned to the transition frequency between the excited and a lower state. Interacting with the electromagnetic field in the cavity, the molecules are stimulated to emit radiation. This radiation, in turn, stimulates emission from other molecules. Hence, nearly monochromatic power is generated in the cavity, part of which is extracted through irises to be used for the frequency standard.

## 3. Theory of the Cavity Containing the Beam

Molecules of ammonia in the higher inversion state enter the resonant cavity. Here their induced transition into the lower state takes place. The transition is stimulated by the interaction with an electromagnetic field in the cavity. The theory of this process was worked out by SHIMODA<sup>4)</sup> *et al.*, under certain simplifying assumptions. It has been extended to meet exact conditions by FEYNMAN<sup>5)</sup> *et al.* The latter used the 'rotating coordinate method' introduced by RABI *et al.*<sup>6)</sup>

In the following, a spinor formalism is applied to illustrate its use in the treatment of two level systems. No new results are obtained in this section except a difference in sign with respect to reference <sup>5)</sup> and the inclusion of more general types of interaction potentials than admitted by FEYNMAN.

Consider a physical system capable of transitions between two stationary states of energies  $E_1$  and  $E_2$  ( $E_2 > E_1$ ). The time independent wave functions of the two states are  $\psi_1$  and  $\psi_2$ , respectively. During the transition the system will be described by the wave function:

$$\psi(t) = a_1(t) \psi_1 + a_2(t) \psi_2 . \quad (3.1)$$

It is therefore appropriate to consider  $\psi(t)$  as a spinor with the complex components  $a_1(t)$  and  $a_2(t)$ , referred to the two basis spinors  $\psi_1$  and  $\psi_2$ . The latter are orthonormal, hence, well suited as a basis for the spinor space. Because it is required that

$$\int \psi(t) \psi^*(t) d^3x = 1 , \quad (3.2)$$

it follows

$$|a_1(t)|^2 + |a_2(t)|^2 = 1 . \quad (3.3)$$

Since its magnitude remains constant, the spinor  $\psi(t)$  undergoes a rotation with respect to the basis  $\psi_1$  and  $\psi_2$  as time proceeds (see Fig. 1).

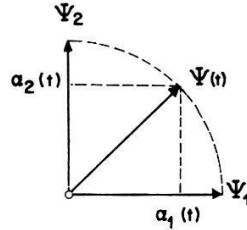


Fig. 1

Spinor representation of the wave function of the two level maser

The change of  $\psi(t)$  is determined by the equation

$$\hat{H} \psi = i \hbar \dot{\psi}, \quad (3.4)$$

where the Hamiltonian operator  $\hat{H}$  can be written as

$$\hat{H} = \hat{H}^0 + \hat{V}(t). \quad (3.5)$$

$\hat{H}^0$  is the Hamiltonian of the two-level system (which will be called 'the molecule' subsequently) in the absence of external fields. Therefore

$$\hat{H}^0 \psi_j = (-1)^j \frac{\hbar \omega_0}{2} \psi_j; \quad j = 1, 2, \quad (3.6)$$

where  $\omega_0$  is defined by

$$\hbar \omega_0 = E_2 - E_1, \quad (3.7)$$

and the zero point of the energy scale is chosen at  $(E_2 - E_1)/2$ .  $\hat{V}(t)$  is the operator of the potential of the external electromagnetic field. It will be assumed that the interaction of the molecule with the electromagnetic field occurs through its induced electric dipole moment  $\hat{\mu}$ . Consequently

$$\hat{V}(t) = - \hat{\mu} \cdot \mathbf{E}(t). \quad (3.8)$$

This assumption is physically realized in the ammonia maser. The method of calculation is the same even if the interaction is different from the above, provided that certain conditions (specified below) are fulfilled by the potential.

Expressing (3.4) in spinor form, we obtain

$$[H^0 + V(t)] \mathbf{a} = i \hbar \dot{\mathbf{a}} \quad (3.9)$$

where  $H^0$  and  $V(t)$  are the two-by-two matrices representing the operators  $\hat{H}^0$  and  $\hat{V}(t)$  and  $\mathbf{a}$  stands for the two component spinor  $\mathbf{a} = \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}$ . The explicit expression of  $H^0$  can readily be found by noticing that for  $V(t) = 0$ ;  $a_1 = a_1^0 \exp(+i \omega_0 t/2)$  and  $a_2 = a_2^0 \exp(-i \omega_0 t/2)$ , thus (3.9) for  $V(t) = 0$  reduces to

$$H^0 \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \frac{\hbar \omega_0}{2} \begin{pmatrix} -a_1 \\ +a_2 \end{pmatrix}, \quad (3.10)$$

hence

$$H^0 = \frac{\hbar \omega_0}{2} \begin{pmatrix} -1 & 0 \\ 0 & +1 \end{pmatrix} = -\frac{\hbar \omega_0}{2} \sigma_z, \quad (3.11)$$

$\sigma_z$  being the Pauli-spin matrix.

The matrix representing the potential  $V(t)$  will have the elements

$$V_{ik}(t) = \int \psi_i^* V(t) \psi_k d^3x, \quad i, k = 1, 2, \quad (3.12)$$

as can be readily seen inserting (3.1) into (3.4), multiplying with  $\psi_i^*$  and integrating over the space coordinates. This matrix can be written as a linear combination of Pauli matrices and the unit matrix  $\sigma_0$ ,

$$V = V_0 \sigma_0 + V_x \sigma_x + V_y \sigma_y + V_z \sigma_z. \quad (3.13)$$

In order that the following method is applicable, we have to assume that  $V_0 = 0$ . As will be clarified in Appendix B, this assumption means that the potential induces transitions between the two states  $\psi_1$  and  $\psi_2$ , and no others. Thus  $V_0 = 0$  is no new restriction on the potential, but equivalent to our starting assumption, that the system is capable of two linearly independent states only. (It is not necessary, however, to assume that  $V_z = 0$ , as reference <sup>5</sup>) asserts for the results to be exact.) Inserting (3.11) and (3.13) into SCHRÖDINGER's equation (3.9) we obtain

$$i \dot{\mathbf{a}} = \left[ V_x \sigma_x + V_y \sigma_y + \left( V_z - \frac{\omega_0}{2} \right) \sigma_z \right] \mathbf{a}. \quad (3.14)$$

For example, in the case of dipole interaction (3.8), the diagonal matrix elements  $V_{11}$  and  $V_{22}$  vanish because the operator of dipole moment commutes with the parity operator. Furthermore, it is always possible to choose the phases of  $\psi_1$  and  $\psi_2$  such that the matrix element  $V_{12}$  becomes real. We then define  $\mu$  by

$$V_{12} = V_{21} = - \int \psi_1^* \hat{\boldsymbol{\mu}} \cdot \mathbf{E} \psi_2 d^3x = - E \mu \quad (3.16)$$

and obtain

$$V = \begin{pmatrix} 0 & -\mu E \\ -\mu E & 0 \end{pmatrix} = -\mu E \sigma_x. \quad (3.17)$$

Inserting (3.11) und (3.17) SCHRÖDINGER's equation (3.14) becomes

$$i \dot{\mathbf{a}} = - \left[ \mu E(t) \sigma_x + \frac{\omega_0}{2} \sigma_z \right] \mathbf{a}. \quad (3.18)$$

The quantity of interest is not the solution  $\mathbf{a}$  of (3.18), but the induced dipole moment  $\mathbf{p}$  of each molecule, since the molecular beam will be considered as a polarizable medium of polarization per unit volume given by

$$\mathbf{P} = \sum \mathbf{p}, \quad (3.19)$$

where the sum is extended over all dipoles in a unit volume. The mean value of the polarization in the direction of the electric field is

$$p = \int \psi^* \mu E \psi d^3x = \mu(a_1^* a_2 + a_2^* a_1) = \mu \mathbf{a}^\dagger \cdot \sigma_x \mathbf{a}. \quad (3.20)$$

The external electric field is assumed periodic and of the form

$$E(t) = E^0 \cos \omega t.$$

To obtain the expression  $p = \mu \mathbf{a}^\dagger \cdot \sigma_x \mathbf{a}$ , it would appear convenient to perform a canonical transformation, which renders the Hamiltonian of (3.18) time independent. Geometrically this is equivalent to the introduction of a rotating coordinate system in spinor space in which the electric field would appear stationary.

It is easy to convince oneself that such a canonical transformation does not exist. A useful approximation can be obtained by the following argument: in the absence of the electric field it is  $a_1 = a_1^0 \exp(i t \omega_0/2)$  and  $a_2 = a_2^0 \exp(-i t \omega_0/2)$ ,  $a_1^0$  and  $a_2^0$  being the initial values of  $a_1$  and  $a_2$ . Hence  $a_1$  and  $a_2$  are conveniently represented by counterrotating vectors in two complex planes. It is advantageous to introduce a rotating coordinate frame in each complex plane, rotating counter-clockwise with angular velocity  $\omega/2$  and  $-\omega/2$ , respectively, through the equation

$$\mathbf{a} = R \mathbf{a}_r \quad (3.21)$$

with

$$R = \exp\left(\frac{i \omega}{2} t \sigma_z\right) \quad (3.22)$$

where  $\mathbf{a}_r$  is the spinor as expressed in the rotating frame. As will be seen subsequently, the canonical transformation  $R$  effects a reduction of the Hamiltonian to a time-independent part and a rapidly oscillating part (which will be neglected), since in the new coordinate frame the electric field will appear as consisting of a stationary part and a part oscillating with the frequency  $2\omega$ .

Inserting (3.21) into (3.18), SCHRÖDINGER's equation becomes

$$i \frac{d}{dt} (R \mathbf{a}_r) = H R \mathbf{a}_r. \quad (3.23)$$

Carrying out the differentiation, and multiplying from the left with  $R^{-1}$ , we obtain

$$-\frac{\omega}{2} \sigma_z \mathbf{a}_r + i \dot{\mathbf{a}}_r = R^{-1} H R \mathbf{a}_r,$$

or

$$i \dot{\mathbf{a}}_r = \left(H_r + \frac{\omega}{2} \sigma_z\right) \mathbf{a}_r \quad (3.24)$$

with  $H_r = R^{-1} H R$ . Note the additional term  $\omega \sigma_z/2$ , which appears as a consequence of the rotation of the coordinate frame, so that the system behaves as if it had the effective Hamiltonian

$$H_{eff} = H_r + \frac{\omega}{2} \sigma_z. \quad (3.25)$$

It is

$$H_{eff} = \frac{\omega - \omega_0}{2} \sigma_z - \mu E^0 \cos \omega t e^{-i\omega t \sigma_z} \sigma_x, \quad (3.26)$$

since

$$e^{-i\omega t \sigma_z/2} \sigma_x e^{i\omega t \sigma_z/2} = e^{-i\omega t \sigma_z} \sigma_x \quad (\text{see Appendix A}).$$

To evaluate the second term on the right, we use the identity  $\cos(\omega t) e^{-i\omega t \sigma_z} = \cos \omega t (\cos \omega t - i \sigma_z \sin \omega t) = 1/2 (1 + \cos 2\omega t - i \sigma_z \sin 2\omega t)$ .

Hence

$$H_{eff} = \frac{\omega - \omega_0}{2} \sigma_z - \frac{\mu E^0}{2} \sigma_x - \frac{\mu E^0}{2} (\cos 2\omega t - i \sigma_z \sin 2\omega t) \sigma_x. \quad (3.27)$$

Since the time spent by the molecules under the influence of the electric field is large compared to the period of oscillation  $2\pi/\omega$  of the latter, it is adequate to average  $H_{eff}$  over a time  $T > 2\pi/\omega$ . The oscillating terms in  $H_{eff}$  then average to zero as compared to the constant terms, and we obtain

$$i \dot{\mathbf{a}}_r = \left( \frac{\omega - \omega_0}{2} \sigma_z - \mu E^0 \sigma_x \right) \mathbf{a}_r. \quad (3.28)$$

Integration yields

$$\mathbf{a}_r = U \mathbf{a}_r^0, \quad (3.29)$$

$$U = \exp - i t \left( \frac{\omega - \omega_0}{2} \sigma_z - \frac{\mu E}{2} \sigma_x \right). \quad (3.30)$$

Introducing (3.22) and (3.29) into the expression for the polarization (3.20), we obtain

$$p = \mathbf{a}^{0\dagger} M \mathbf{a}^0 \quad (3.32)$$

where

$$M = \mu U^\dagger R^\dagger \sigma_x R U. \quad (3.33)$$

Since all molecules enter the cavity in the higher energy state, the initial value of the spinor is

$$\mathbf{a}_r^0 = \mathbf{a}^0 = \begin{pmatrix} 1 \\ 0 \end{pmatrix}. \quad (3.34)$$

Therefore the determination of the polarization reduces to the calculation of the matrix element

$$p = M_{22}. \quad (3.35)$$

Using the rules set forth in Appendix A for the manipulation of exponential functions of Pauli matrices, a straight-forward calculation gives

$$p = a(t) \cos \omega t + b(t) \sin \omega t, \quad (3.36)$$

$$a(t) = - \frac{\mu^2 E^0 (\omega - \omega_0)}{4 \Omega^2} \sin^2(\Omega t/2) = - \frac{\mu^2 E^0}{2 \Omega^2} (1 - \cos \Omega t), \quad (3.37)$$

$$b(t) = \frac{\mu^2 E^0}{2 \Omega} \cos(\Omega t/2) \sin(\Omega t/2) = \frac{\mu^2 E^0}{\Omega} \sin \Omega t, \quad (3.38)$$

$$\Omega^2 = (\mu E^0)^2 + (\omega - \omega_0)^2. \quad (3.39)$$

In the calculations involving the cavities it is convenient to consider the electric field as the real part of a complex quantity, according to

$$E(t) = E^0 \cos \omega t = R[E_c(t) e^{i\omega t}] \quad (3.40)$$



and use  $E_c(t)$  in the calculations. Correspondingly, the polarization has to be considered as the real part of a complex quantity  $p_c$ :

$$p(t) = a(t) \cos \omega t + b(t) \sin \omega t = R[p_c e^{i\omega t}]. \quad (3.41)$$

This equation yields

$$p_c = a - i b. \quad (3.42)$$

#### 4. The Theory of Multiple Cavities

Consider  $n$  cavities coupled to each other. The maximum possible number of couplings is  $n(n-1)/2$ . Let there be an active polarization (due to the molecular beam) in one of the cavities, called cavity 1. The electric field inside the system of cavities is given by

$$\mathbf{E}(t) = \mathbf{E} e^{i\omega t} \quad (4.1)$$

and the polarization in the first cavity is

$$\mathbf{P} = \mathbf{P}_1 e^{i\omega t}. \quad (4.2)$$

The figures of merit  $Q_k$  of the cavities are assumed to be large, thus the tangential component of the electric field in the walls will be small. The electric field will be written as

$$\mathbf{E} = \sum_{k=1}^n e_k (\mathbf{E}_k + \mathbf{E}_k^1) e^{i\omega t}, \quad (4.3)$$

$\mathbf{E}_k = 0$  in all cavities except  $k$ , where  $\mathbf{E}_k$  is that normal mode of oscillation which would exist if there were no polarization present, no coupling to the other cavities and if cavity  $k$  had perfectly conducting walls.  $\mathbf{E}_k$  is given as the time independent part of the normal mode  $\mathbf{E}_k^t$ :

$$\mathbf{E}_k^t = \mathbf{E}_k e^{i\omega_k t}, \quad k = 1, \dots, n. \quad (4.4)$$

$e_k$  is the amplitude factor which is introduced since the  $\mathbf{E}_k$  are normalized to

$$\int \mathbf{E}_k^2 dV = 1, \quad k = 1, \dots, n. \quad (4.5)$$

The polarization, coupling and wall losses are treated here as perturbations and  $\mathbf{E}_k^1$  is that additional field which has to be added to  $\mathbf{E}_k$  in the cavity  $k$  to give the correct field. Notice that

$$\mathbf{E}_k^1 \ll \mathbf{E}_k$$

in the interior of the cavities, with the exception of the irises coupling the cavities.  $\mathbf{E}_k^1$  is small, but nonzero in the walls, and may be large in the iris. Fig. 2 illustrates the fields  $\mathbf{E}_k$  and  $\mathbf{E}_k^1$  for three cavities.

From MAXWELL's equations  $c \operatorname{rot} \mathbf{E} = -\dot{\mathbf{H}}$  and  $c \operatorname{rot} \mathbf{H} = \dot{\mathbf{D}}$  as well as  $\mathbf{D} = \mathbf{E} + 4\pi \mathbf{P}$  follows

$$c^2 \operatorname{rot} \operatorname{rot} \mathbf{E} + \ddot{\mathbf{E}} + 4\pi \ddot{\mathbf{P}} = 0. \quad (4.6)$$



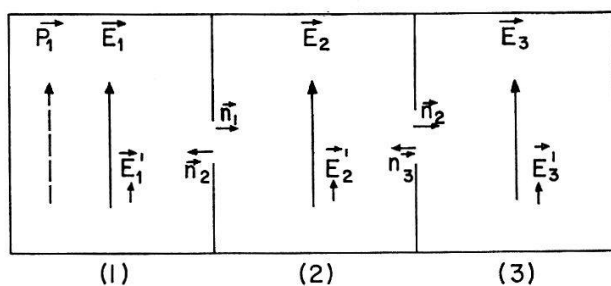


Fig. 2

Schematic diagram of coupled cavities

 $E$  electric field;  $E'$  perturbing electric field;  $P$  polarization due to molecular beam

Multiplying through by  $E_1$  and integrating over the volumes of all cavities, we get after utilizing common vector identities

$$\left. \begin{aligned} -\omega^2 \left( e_1 + 4\pi \int_v E_1 \cdot P_1 dv \right) - i e_1 \omega_1 c \int_s H_1 \cdot n_1 \times E_1^1 ds + \omega_1^2 e_1 \\ - i c \omega_1 \sum_{j=2}^n e_j \int_s H_1 \cdot n_1 \times E_j^1 ds = 0 ; \end{aligned} \right\} \quad (4.7)$$

$S$  is the surface bounding the volume of the cavities. Similarly, multiplying by  $E_k$  and integrating over the entire volume we obtain the  $n - 1$  equations

$$\left. \begin{aligned} -\omega^2 e_k - i e_k \omega_k c \int_s H_k \cdot n_k \times E_k^1 ds + \omega_k^2 e_k \\ - i c \omega_k \sum_{\substack{l=1 \\ l \neq k}}^n e_l \int_s H_k \cdot n_k \times E_l^1 ds = 0, \quad k = 2, \dots, n. \end{aligned} \right\} \quad (4.8)$$

The outward normal to the surface of cavity  $k$  is denoted by  $n_k$ .

Using the results of the previous sections, where an expression for the polarization was derived, it follows for a thin molecular beam:

$$4\pi \int_v E_1 \cdot P_1 dv = \frac{n}{n_{th}} \frac{e_1}{Q_1} \left( Q_B \frac{\omega_0 - \omega}{\omega_0} \frac{\Theta}{2\pi} - i\Phi \right), \quad (4.9)$$

where

$n$  = number of molecules entering the cavity per second,

$n_{th}$  = minimum number of molecules entering the cavity per second necessary for self-sustained oscillation,

$Q_k$  = figure of merit of cavity  $k$ ,

$Q_B = 2\pi(\omega_0/v)L$ ,

$\omega_0$  = molecular transition frequency,

$v$  = molecular velocity,

$L$  = length of molecular beam within cavity.

Furthermore we have set

$$\Theta = \frac{(1 - \sin \theta / \theta)}{\theta^2} \quad \text{and} \quad \Phi = \frac{(1 - \cos \theta)}{\theta^2}$$

where  $\theta = \Omega t$ .  $\Omega$  was defined in the previous section.

From (4.7) we see, that the first surface integral corresponds to the wall losses in cavity 1, whereas the surface integrals summed over in (4.8) represent the losses of cavity 1 through coupling to the other individual cavities. Hence, integrals of the second type define the transfer constants  $M_k$

$$\left. \begin{aligned} \frac{1}{Q_k} &= \frac{c}{\omega_k} \int_s \mathbf{H}_1 \cdot \mathbf{n}_1 \times \mathbf{E}'_1 ds, \quad k = 1, \dots, n. \\ M_{kl} &= \frac{c}{\omega_k} \int_s \mathbf{H}_k \cdot \mathbf{n}_k \times \mathbf{E}'_l ds, \quad M_{lk} = - \frac{c}{\omega} \int_s \mathbf{H}_l \cdot \mathbf{n}_l \times \mathbf{E}'_k ds. \end{aligned} \right\} \quad (4.10)$$

The choice of the signs of  $M_{kl}$  and  $M_{lk}$  is evident from the fact that any energy lost by cavity  $k$  to cavity  $l$  by coupling must appear as a gain in the equation of cavity  $l$ .

Inserting these definitions into (4.7) and (4.8) and rearranging, we arrive to

$$\left. \begin{aligned} e_1 \left( \frac{\omega_1}{\omega} - \frac{\omega}{\omega_1} \right) - i e_1 \frac{\omega_1}{\omega} \frac{1}{Q_1} - i \frac{\omega_1}{\omega} \sum_{j=2}^n e_j M_{1j} \\ - \frac{\omega}{\omega_1} \frac{n}{n_{th}} \frac{e_1}{Q_1} \left( \frac{Q_B \Theta}{2 \pi} \frac{\omega_0 - \omega}{\omega_0} - i \Phi \right) = 0, \end{aligned} \right\} \quad (4.11)$$

$$e_k \left( \frac{\omega_k}{\omega} - \frac{\omega}{\omega_k} \right) - i e_k \frac{\omega_k}{\omega} \frac{1}{Q_k} + i \frac{\omega_k}{\omega} \sum_{\substack{l=1 \\ l \neq k}}^n e_l M_{kl} = 0, \quad k = 2, \dots, n. \quad (4.12)$$

Terms with  $(+1)$  as a coefficient correspond to gains, terms with  $(-1)$  to losses for the cavity  $k$ .

These equations represent a set of simultaneous equations for the determination of the electric field amplitudes  $e_k$  and their respective phases (since the  $e_k$  are complex). To simplify the calculations, we restrict ourselves to the case of identical cavities. This means

$$\left. \begin{aligned} \omega_k &= \omega_c, \\ Q_k &= Q, \end{aligned} \right\} \quad k = 1, \dots, n, \quad (4.13)$$

where  $\omega_c$  and  $Q$  are their common frequency and figure of merit, respectively.

Let us introduce a quantity  $\alpha$ , proportional to the detuning of the cavities, i.e. proportional to the deviation of the cavity frequency  $\omega_c$  from the molecular frequency  $\omega_0$ :

$$\alpha = 2 Q \frac{\omega_c - \omega_0}{\omega_0}. \quad (4.14)$$

Assuming good tuning, i.e.  $\omega_c - \omega_0 \ll \omega_0$ , we have

$$\alpha = Q \left( \frac{\omega_c}{\omega} - \frac{\omega}{\omega_c} \right). \quad (4.15)$$

Also, let us define a quantity  $\eta$  representing the pulling of the oscillator, i.e. a quantity proportional to the deviation of the actual frequency of oscillation  $\omega$  of the electric field from the molecular frequency  $\omega_0$

$$\eta = \frac{Q_B \Theta}{2 \pi \Phi} \frac{\omega - \omega_0}{\omega_0}. \quad (4.16)$$

It is convenient to set

$$K_{kl} = Q M_{kl} \quad (4.17)$$

and refer to the  $K_{kl}$  as the coupling parameter<sup>7)</sup>. The  $K_{kl} = -K_{lk}$ .

To solve the set of Equations (4.11) and (4.12), we make use of the fact that the choice of the phase of the electric field is arbitrary in one of the cavities. We choose

$$\left. \begin{aligned} e_1 &= \text{real}, \\ e_k &= e'_k + i e''_k, \quad e'_k, e''_k = \text{real}; \quad k = 2, \dots, n. \end{aligned} \right\} \quad (4.18)$$

Expressing  $n/n_{th}$  from the real part of (4.11) and inserting it into the imaginary part of (4.12), and separating real and imaginary parts of the remaining  $n - 1$  equations, we arrive to  $2(n - 1) + 1$  equations, which, written in matrix form read

$$\left\| \begin{array}{cccccc} (\alpha - \eta) & -\eta K_{12} & K_{12} & -\eta K_{13} & K_{13} & \cdots & K_{1n} \\ -K_{21} & 1 & -\alpha & K_{23} & 0 & \cdots & 0 \\ 0 & \alpha & 1 & 0 & K_{23} & \cdots & K_{2n} \\ -K_{31} & -K_{32} & 0 & 1 & -\alpha & \cdots & 0 \\ 0 & 0 & -K_{32} & \alpha & 1 & \cdots & K_{3n} \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdots & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdots & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdots & \cdot \\ 0 & 0 & -K_{n2} & 0 & K_{n3} & \cdots & 1 \end{array} \right\| \cdot \left\| \begin{array}{c} e_1 \\ e'_2 \\ e''_2 \\ e'_3 \\ e''_3 \\ \vdots \\ e''_n \end{array} \right\| = 0. \quad (4.19)$$

It is necessary that the determinant of the matrix vanishes for nontrivial solutions  $e_k$ . This condition will be the required equation determining the pulling  $\eta$  in function of the coupling parameters  $K_{kl}$  and the detuning  $\alpha$ . Setting the determinant of (4.19) equal to zero, and expanding it, follows

$$\eta = \frac{\alpha \Delta_1 + K_{ij} \Delta_j}{\Delta_1 - \sum_{j=2} K_{ij} \bar{\Delta}_j}, \quad (4.20)$$

where

$\Delta_j$  = minor<sup>8)</sup> of column  $K_{ij}$ ,

$\bar{\Delta}_j$  = minor of column  $\eta K_{ij}$ ,

$\Delta_1$  = minor of column  $(\alpha - \eta)$ .

Expanding further, one may write  $\eta$  as the ratio of two polynomials in  $\alpha$ :

$$\eta(\alpha) = \frac{\alpha^{2n-1} + \sum_{m=1}^{n-1} \frac{1}{(2m-1)!} \left[ (2m-1) \Delta_1^{2m-2}(0) + \sum_{j=2}^n K_{ij} \Delta_j^{2m-1}(0) \right] \alpha^{2m-1}}{\alpha^{2n-2} + \sum_{m=1}^{n-1} \frac{1}{(2m-2)!} \left[ \Delta_1^{2m-2}(0) - \sum_{j=2}^n K_{ij} \Delta_j^{2m-2}(0) \right] \alpha^{2m-2}}, \quad (4.21)$$

where

$$\Delta_j^m(0) = \frac{d^m}{d\alpha^m} (\Delta_j) |_{\alpha=0}. \quad (4.22)$$

It is our aim to give the coupling parameters such values as to make the pulling  $\eta$  as small as possible for a given detuning  $\alpha$ . Since  $\alpha < 1$ ,  $\eta$  will be minimal if in the numerator of (4.21) the coefficients of all powers of  $\alpha$ —except that of  $\alpha^{2n-1}$ —vanish:

$$(2m-1) \Delta_1^{2m-2}(0) + \sum_{j=2}^n K_{ij} \Delta_j^{2m-1}(0) = 0, \quad m = 1, \dots, n-1. \quad (4.23)$$

If the  $n(n-1)/2$  coupling parameters  $K_{ij}$  are so chosen that they fulfill these  $n-1$  equations, the pulling will be minimal, and equal to

$$\eta(\alpha) = \frac{\alpha^{2n-1}}{\Delta_1(0) - \sum_{j=2}^n K_{ij} \bar{\Delta}_j(0)}. \quad (4.24)$$

It is possible to fulfill equations (4.23) by setting all coupling constants but  $K_{j,j+1}$  ( $j = 1, \dots, n-1$ ) equal to zero. This means that it is not necessary to cross-couple the  $n$  cavities (i.e. to couple each one with every other one) but to couple them to form a linear chain. Hence for minimum pulling it is sufficient to demand

$$(2m-1) \Delta_1^{2m-2}(0) + K_{12} \Delta_2^{2m-1}(0) = 0, \quad m = 1, \dots, n-1. \quad (4.25)$$

The pulling then becomes

$$\eta(\alpha) = \frac{\alpha^{2n-1}}{\Delta_1(0) - K_{12} \bar{\Delta}_2(0)}, \quad (4.26)$$

or, using (4.14) and (4.15)

$$\frac{\omega - \omega_0}{\omega_0} = \frac{2\pi(2Q)^{2n-1}}{Q_B \Theta [\Delta_1(0) - K_{12} \bar{\Delta}_2(0)]} \left( \frac{\omega_c - \omega_0}{\omega_0} \right)^{2n-1}. \quad (4.27)$$

## 5. Coupling of the Cavities and Power Dissipation

Once the coupling parameters are determined from (4.25) the electric field strengths  $e_i$  can be found in each cavity by solving the equations (4.19). The quantities  $\alpha$  and  $\eta$  can be set equal to zero in these equations, since their influence on the field strengths are negligible. The solution of the equations is then simplified by noticing that all field strengths are real, i.e.  $e_k'' = 0$ ,  $k = 1, \dots, n$ .

The power dissipated in a slightly overcritical single cavity maser has been calculated in references 3) and 4). It is of interest to compare it with the power dissipation in the multiple-cavity system. The ratio  $R$  of the latter to the former, expressed in terms of the fields strengths, is

$$R = \sum_{i=1}^n e_i^2 / e_1^2. \quad (5.1)$$

The power dissipation can also be obtained without evaluating the field strengths from the relation

$$R = \frac{n_{th}}{n_{th}(1)}, \quad (5.2)$$

where  $n_{th}(1)$  is the threshold number of molecules for a simple cavity maser.

## 6. Reduction of the Pulling by Multiple Cavity Systems

Formula (4.27) shows, that if  $n$  cavities are used instead of one and they are coupled so that the coupling parameters fulfill the conditions (4.25), the pulling will be reduced by a factor of the order of

$$\left(2Q \frac{\omega_c - \omega_0}{\omega_0}\right)^{2n-2}.$$

On the one hand, to insure low power dissipation it is advantageous to use cavities with high figures of merit; on the other hand, since the reduction of pulling requires

$$2Q \frac{\omega_c - \omega_0}{\omega_0} < 1,$$

$Q$  should not be chosen excessively large.

In particular, we obtain for a single cavity

$$\frac{\omega - \omega_0}{\omega_0} = \frac{2\pi\Phi}{Q_B\Theta} 2Q \frac{\omega_c - \omega_0}{\omega_0}, \quad (6.1)$$

for two cavities

$$K_{12}^2 = 1, \quad (6.2)$$

$$\frac{\omega - \omega_0}{\omega_0} = \frac{2\pi\Phi}{Q_B\Theta} 1/2 \left(2Q \frac{\omega_c - \omega_0}{\omega_0}\right)^3, \quad (6.3)$$

for three cavities

$$K_{12}^2 = 4(\sqrt{2} - 1) = 1.657, \quad K_{23}^2 = 3 - 2\sqrt{2} = 0.172, \quad (6.4)$$

$$\frac{\omega - \omega_0}{\omega_0} = \frac{2\pi\Phi}{Q_B H} \frac{1}{8\sqrt{2}} \left(2Q \frac{\omega_c - \omega_0}{\omega_0}\right)^5. \quad (6.5)$$

The calculation shows that the two cavity system dissipates  $R = 2$  times, the three cavity system dissipates  $R = 3.67$  times the power of a single cavity.

### Appendix A

As was pointed out by RABI, RAMSEY and SCHWINGER<sup>6)</sup> the calculation with exponential functions of Pauli matrices presents some difficulties because of the non-commutativity of these matrices. Indeed, errors may arise from the use of equalities familiar from the theory of functions of complex variables, which are invalid for functions of Pauli matrices. For example, it is

$$e^{i\sigma_x} e^{i\sigma_y} \neq e^{i(\sigma_x + \sigma_y)}.$$

Hence, for reference purposes a few relations are reproduced below, and their proof indicated.

Let  $\sigma_k$  be a Pauli matrix normalized to  $\sigma_k^2 = 1$  (i.e. without the factor  $1/2$ ), and let us denote by  $\sigma$  the vector with components  $\sigma_x, \sigma_y$  and  $\sigma_z$ . (The exponential functions of Pauli matrices be defined by their respective power series.) Then, if  $t$  is a real parameter

$$e^{it\sigma_k} = \cos t + i \sigma_k \sin t, \quad k = x, y, z, \quad (\text{A.1})$$

and if  $\Omega$  is a real vector with components  $\Omega_x, \Omega_y$  and  $\Omega_z$  (magnitude  $\Omega$ ), it is

$$e^{it\Omega \cdot \sigma} = \cos \Omega t + i \frac{\Omega \cdot \sigma}{\Omega} \sin \Omega t. \quad (\text{A.2})$$

(A.2) can be most easily proved by performing an orthogonal basis transformation in the vector space of Pauli matrices. Defining  $\sigma_1$  as a new basis vector through

$$\sigma_1 = \frac{1}{\Omega} \Omega \cdot \sigma, \quad (\text{A.3})$$

application of (A.1) yields

$$e^{it\Omega \cdot \sigma} = e^{it\Omega \cdot \sigma_1} = \cos \Omega t + i \sigma_1 \sin \Omega t. \quad (\text{A.4})$$

Inserting (A.3) into (A.4) the required relation (A.2) follows.

If the product of two exponentials is encountered such as, e.g.

$$e^{it_1\Omega_1 \cdot \sigma} e^{it_2\Omega_2 \cdot \sigma},$$

expression (A.2) is inserted for both of them, and the product of the right hand side terms evaluated.

The following relations (and those obtained from them by cyclic permutation of the indices) are also used in the text:

$$e^{it\sigma_x} \sigma_x = \sigma_x e^{it\sigma_x}, \quad (\text{A.5})$$

$$e^{it\sigma_k} \sigma_x = \sigma_x e^{-it\sigma_k}, \quad k = y, z. \quad (\text{A.6})$$

### Appendix B

It is of interest to establish the connection between the method used in the text and the geometrical method described in <sup>5</sup>).

To do so, we turn to the three-dimensional representation of the spinor  $\mathbf{a}$  and of the Pauli matrices  $\sigma_x$ ,  $\sigma_y$  and  $\sigma_z$ . It can be shown<sup>9</sup>) that the spinor  $\mathbf{a}$  may be considered as the stereographic projection of a three-dimensional vector  $\mathbf{r}$ . The standard formulae connecting the components of  $\mathbf{a}$  and  $\mathbf{r}$  can be found in <sup>9</sup>).

Let us refer the vectors in the three-dimensional space to the unit vectors  $\mathbf{e}_j$ ,  $j = 1, 2, 3$ . An infinitesimal rotation of the vector  $\mathbf{r}$  through an angle  $dt \Omega$  about an axis denoted by a vector  $\Omega$

$$\Omega = \sum_{j=1}^3 \Omega_j \mathbf{e}_j \quad (\text{B.1})$$

will produce a change  $d\mathbf{r}$  in  $\mathbf{r}$ , which can be expressed as:

$$d\mathbf{r} = dt \Omega \times \mathbf{r} = \sum_{j=1}^3 dt \Omega_j \mathbf{e}_j \times \mathbf{r}. \quad (\text{B.2})$$

The operators  $dt \Omega_j \mathbf{e}_j \times$  represent infinitesimal rotations about the  $j$ -coordinate axis by the angle  $dt \Omega_j$ . They can be expressed in terms of the spin matrices as  $1/2 i dt \Omega_j \sigma_j$ , when operating on some  $\mathbf{a}^{10}$ ). Conversely, an arbitrary linear combination of the spin matrices  $dt \Omega_j \sigma_j$  which operates on  $\mathbf{a}$  will be represented by the operator  $-2 i dt \Omega \times$ , acting on  $\mathbf{r}$ . Hence, the following correspondence is established

$$1/2 i dt \Omega \cdot \sigma = 1/2 \sum_{j=1}^3 i dt \Omega_j \sigma_j \rightarrow dt \Omega \times. \quad (\text{B.3})$$

In the three-dimensional representation Equation (3.15) can be written as

$$dt \Omega \times \mathbf{r} = d\mathbf{r} \quad (\text{B.4})$$

and comparison of (3.14) and (B.3) yields

$$\Omega = \left( -V_x, -V_y, -V_z + \frac{\omega_0}{2} \right). \quad (\text{B.5})$$

In the particular example (3.17) one has

$$\Omega = \left( \mu E(t), 0, \frac{\omega_0}{2} \right). \quad (\text{B.6})$$

Since Equation (B.4) is the familiar equation of the precession of a top, then if  $\mathbf{r}$  is considered as the angular momentum of the top and  $\Omega$  is the driving torque, the behavior of the system can be visualized by studying the motion of the top itself.



Correspondingly, the polarization can be expressed as  $p = \mathbf{a}^\dagger \cdot \sigma_x \mathbf{a} = r_x$ , and the problem is reduced to the determination of the projection of the vector  $\mathbf{r}$  upon the  $x$ -axis at time  $t$ .

It is seen from (B.3) or (B.5), that if the potential  $V$  contains a term  $V_0 \sigma_0$ , the three-dimensional representation of the Schrödinger equation is just as impossible as the spinor representation. This is due to the fact that the two states  $\psi_1$  and  $\psi_2$  do not in this case form a complete set of eigenstates of the system; to the contrary, the potential  $V$  will induce transitions to other states not taken into account in the formalism.

### Acknowledgement

The Author's sincere thanks are due to Dr. R. C. HERNDON for his valuable cooperation.

### References

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- <sup>4)</sup> SHIMODA, WANG and TOWNES, Phys. Rev. 102, 1308 (1956).
- <sup>5)</sup> FEYNMAN, VERNON and HELLWARTH, J. Appl. Phys. 28, 49 (1957).
- <sup>6)</sup> RABI, RAMSEY and SCHWINGER, Revs. Modern Phys. 26, 167 (1954).
- <sup>7)</sup> In the literature often the square of  $K_{kl}$  is referred to as the coupling constant.
- <sup>8)</sup> In calculating the minor of the element with row and column indices  $i$  and  $j$ , respectively, no sign  $(-1)^{i+j}$  is to be included.
- <sup>9)</sup> H. WEYL, *The Theory of Groups and Quantum Mechanics* (Dover Publ. 1931), p. 144.
- <sup>10)</sup> See for example LANDAU and LIFSHITZ, *Quantum Mechanics* (Pergamon Press, London 1958), p. 189.