Further Aspects of the Theory of the Maser*

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(Received November 28, 1955)

The theory of the molecular transitions which are induced by the microwave field in a maser and the effects of various design parameters are examined in detail. It is shown that the theoretical minimum detectable beam intensity when the maser is used as a spectrometer for the 3–3 line of ammonia is about 10^9 molecules/sec under typical experimental conditions. Various systematic frequency shifts and random frequency fluctuations of the maser oscillator are discussed and evaluated. The most prominent of the former are the "frequency-pulling" effect, which arises from detuning of the cavity, and the Doppler shift due to the asymmetrical coupling of the beam with the two traveling wave components of the standing waves which are set up in the cavity. These two effects may produce fractional shifts as large as one part in 10^9. If adequate precautions are taken, however, they can be reduced to one part in 10^{10} or possibly less. The random fluctuations are shown to be of the order of one part in 10^{13} under typical operating conditions. For molecular beams in which the electric-dipole transition is used, the TM_{010} mode is usually the most suitable for the maser; while in atomic beams in which magnetic transitions are utilized, the TE_{011} mode is to be preferred.

I. INTRODUCTION

A DEVICE, which has been called a maser, involving a beam of molecules which give "Microwave Amplification by Stimulated Emission of Radiation," has already been described and much of the basic theory stated.[1,2] In the following discussion, we have examined in more detail certain aspects of the theory involved and explored some effects or conditions which were previously ignored or mentioned only briefly. In particular, the effects of saturation and of resonant cavity design are considered, and various types of noise and frequency shifts of the oscillator are treated.

II. ANALYSES OF THE MASER SPECTROMETER

A. Induced Emission and Saturation Effect

A beam of molecules (or atoms) in a certain quantum state passes through a resonant cavity tuned approximately to the frequency of transition of the molecules. The cavity is excited by a microwave generator through a coupled wave guide, which stimulates the transition of molecules and results in emission or absorption of microwaves. Assuming a low density of molecules in the beam, any direct interaction between the molecules, such as collision, can be neglected. The velocity of the molecules is not uniform, but all the molecules may be considered to be in the common microwave field and the emission or absorption waves from each molecule are superposed to produce the total emission or absorption.

Consider a molecule which is initially in state ψ_2 with energy W_2, and which is stimulated by a perturbation to emit or absorb microwave energy by transition to state ψ_1 with energy W_1. The wave function can at any time

be expressed by

$$\psi = a_1\psi_1 + a_2\psi_2. \tag{1}$$

As an initial condition at $t = t_0$, $|a_2| = 1$ and $a_1 = 0$. The resonant frequency of the molecule is

$$\omega_0 = (W_2 - W_1)/\hbar. \tag{2}$$

For simplicity of calculation, let us assume that the periodic perturbation

$$H' = -\mathbf{E} \cdot \mathbf{\mu} \cos\omega t \tag{3}$$

is given to the molecule from $t = t_0$ to t. Here $\mathbf{E}$ is the electric field strength and $\mathbf{\mu}$ the dipole moment. The effect of field inhomogeneity in the cavity and of the thermal noise field will be discussed later.

The coefficients a_1 and a_2 as functions of t, ω, and E can then be obtained by perturbation theory as

$$a_1(t) = -e^{\frac{1}{2}i(\omega-\omega_0)t}\frac{x}{[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}}$$
$$\times \sin\left\{[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}\left(\frac{t-t_0}{2}\right)\right\} \tag{4}$$

and

$$a_2(t) = e^{-\frac{1}{2}i(\omega-\omega_0)t}\left\{-\frac{\omega-\omega_0}{[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}}\right.$$
$$\times \sin\left[[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}\left(\frac{t-t_0}{2}\right)\right]$$
$$\left. +i\cos\left[[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}\left(\frac{t-t_0}{2}\right)\right]\right\}, \tag{5}$$

where

$$x = E\bar{\mu}/\hbar$$

and $\bar{\mu}$ is the matrix element between the two states for the component of the dipole moment along the direction of E.

The microwave power emitted from the beam of n

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[1] J. P. Gordon, Phys. Rev. **99**, 1253 (1955).
[2] Gordon, Zeiger, and Townes, Phys. Rev. **99**, 1264 (1955).

molecules per second is given by

$$\Delta P = nh\nu_0 |a_1(L/v)|^2, \tag{6}$$

where L/v is the transit time of molecules with velocity v through the cavity of length L. If the nonuniform velocity distribution is taken into consideration, Eq. (6) should be replaced by

$$\Delta P = h\nu_0 \int_0^\infty n(v) \left| a_1\left(\frac{L}{v}\right) \right|^2 dv. \tag{7}$$

ΔP depends on E^2, the square of the electric field.

Assuming a uniform field distribution within the cross section of the cavity resonator, one obtains

$$E^2 = 8\pi W/AL, \tag{8}$$

where A is the area of the cross section, and W the stored microwave energy. To allow for the nonuniformity of the field distribution, this formula can be corrected to the form

$$E_e^2 = 8\pi W/A_e L,$$

where

$$A_e = \frac{\int\int rn(r)d\theta dr \int\int r|E(r,\theta)|^2 d\theta dr}{\int\int r|E(r,\theta)|^2 n(r)d\theta dr}, \tag{9}$$

$$E_e^2 = \frac{\int\int r|E(r,\theta)|^2 n(r)d\theta dr}{\int\int rn(r)d\theta dr},$$

and $n(r)$ represents the density of the molecular beam. This assumes cylindrical symmetry, and also assumes that each molecule travels parallel to the axis so that it remains in a constant field.

Assuming a uniform molecular velocity as in (6), the emitted power may be written

$$\Delta P = nh\nu_0 \left(\frac{\theta^2}{\delta^2 + \theta^2}\right) \sin^2(\delta^2 + \theta^2)^{\frac{1}{2}}, \tag{10}$$

where

$$\theta^2 = W/W_c, \quad W_c = h^2 A v^2/8\pi^3 \bar{\mu}^2 L, \tag{11}$$

and

$$\delta = (\omega - \omega_0)(L/2v) = \pi(\nu - \nu_0)L/v. \tag{12}$$

Equation (10) shows that saturation of the spectral line becomes appreciable when

$$\theta \gtreqless 1, \quad \text{or} \quad W \geq W_c.$$

When the input power to the cavity is so small that there is no appreciable saturation, the emitted power is, from (10),

$$\Delta P = nh\nu_0 \theta^2 (\sin^2\delta/\delta^2). \tag{13}$$

This expression applies accurately only when $\delta \gg \theta \ll 1$ and for a uniform field distribution along the axis of the cavity. It gives a width to the line (frequency difference between half power points) of $2\Delta\nu = 0.89v/L$. If the field varies along the axis as $\sin(\pi z/2L)$, with z extending from 0 to L, as in the case of a TE mode, the line shape is given by

$$\Delta P \propto \left[\frac{\cos\delta}{1 - (2\delta/\pi)^2}\right]^2, \tag{14}$$

which gives the line width $2\Delta\nu = 1.19v/L$. Equation (10) is not exact in this case, but it holds to a fairly good approximation if one takes, instead of (11),

$$W_c = h^2 A v^2/64\pi\bar{\mu}^2 L. \tag{15}$$

B. Detection of the Emitted Power

Consider a high-Q cavity resonator which has an output waveguide with coupling represented by Q_1, and an input waveguide of coupling Q_2. The beam of molecules is admitted into the cavity through another hole parallel to the axis of the cylinder. With the available power, P_a, in the input wave guide, the stored energy at the resonant frequency in the cavity is

$$W = (2/\pi\nu)(Q^2/Q_2)P_a, \tag{16}$$

where Q is the loaded Q of the cavity. The output power in the absence of the molecular beam is given by

$$P_0 = (2\pi\nu W/Q_1) = (4Q^2/Q_1Q_2)P_a. \tag{17}$$

In the presence of the beam, the increase of power in the output wave guide due to induced emission can be calculated as

$$\Delta P_0 = (2Q/Q_1)\Delta P, \tag{18}$$

assuming that the emitted power is small compared to the loss in the cavity.

Since the signal power to be compared to the noise power is[3]

$$\Delta P_s = (\Delta P_0)^2/4P_0,$$

the minimum detectable number of molecules per unit time, n_{min}, can be calculated from the following condition:

$$\frac{Q^2}{Q_1^2}\left(\frac{(\Delta P_{min})^2}{P_0}\right) = FkT\Delta f. \tag{19}$$

Here F is the over-all noise figure of the detector and amplifier, and Δf their effective band width. From Eqs. (10) and (19) with $\omega = \omega_0$, one obtains

$$n_{min} = \frac{Q_1(P_0 FkT\Delta f)^{\frac{1}{2}}}{Qh\nu \sin^2\theta}$$

$$= \frac{\theta}{\sin^2\theta}\frac{(Q_1)^{\frac{1}{2}}}{Q}\frac{(2\pi\nu W_c \cdot FkT\Delta f)^{\frac{1}{2}}}{h\nu}. \tag{20}$$

[3] C. H. Townes and S. Geschwind, J. Appl. Phys. 19, 795 (1948).

The optimum condition of the operating power level and couplings of wave guides can be evaluated by differentiating Eq. (20). This gives

$$\tan\theta = 2\theta,$$

which shows that for optimum sensitivity

$$\theta = 1.16, \quad W = 1.35 W_c, \tag{21}$$

and

$$\theta/\sin^2\theta = 1.38. \tag{22}$$

The highest sensitivity can be obtained when the cavity is designed for minimum value of Q_1/Q^2. Assuming Q_0, the unloaded quality factor of the cavity, to be constant, the minimum value of Q_1/Q^2 is

$$\frac{Q_1}{Q^2} = \frac{2}{Q} = 4\left(\frac{1}{Q_0} + \frac{1}{Q_2}\right). \tag{23}$$

As will be shown later, the available input power of the order of milliwatt is much larger than the power in the cavity in most cases, so that $Q_2 \gg Q_0$. Hence

$$Q_1/Q^2 \simeq 4/Q_0, \quad Q_1 \simeq Q_0 \simeq 2Q, \tag{24}$$

for the optimum coupling. Then the minimum flow of molecules per unit time which can be detected is given by

$$n_{\min} = \frac{6.92}{h}\left(\frac{W_c F k T \Delta f}{Q_0 \nu}\right)^{\frac{1}{2}}$$

$$= 0.44 \frac{\nu}{\bar{\mu}}\left(\frac{A}{Q_0 L \nu}\right)^{\frac{1}{2}} (F k T \Delta f)^{\frac{1}{2}}, \tag{25}$$

using Eqs. (11), (20), (22), and (24).

If a cavity resonator is coupled by only one wave guide and one observes the change of power reflected from the cavity, the condition for the optimum coupling should have the same form as Eq. (24), resulting in the same equation for the optimum sensitivity.

As a specific example, consider a spectrometer for ammonia. The average velocity of molecules at a temperature T is given by kinetic theory as

$$v = (2/\pi^{\frac{1}{2}})(2RT/M)^{\frac{1}{2}} = 14\,551\,(T/M)^{\frac{1}{2}} \text{ cm/sec},$$

where M is the molecular weight, and R the gas constant. Letting $M = 17$ and $T = 290°$K, this expression gives $v = 6.0 \times 10^4$ cm/sec. For the 3–3 line of NH_3, $\nu = 2.4 \times 10^{10}$ cps and $\bar{\mu} = 1 \times 10^{-18}$ cgs unit. The design of the cavity will be discussed in the following section, but the following values are taken as typical. Putting $A = 1$ cm^2, $L = 10$, one obtains for the 3–3 line of ammonia

$$W_c = 6.4 \times 10^{-11} \text{ erg}.$$

The optimum energy in the cavity is then found from Eq. (21). Using values $Q_1 = Q_0 = 10^4$, and $Q = 5 \times 10^3$, the net power flow from the cavity is

$$P_0 = 2\pi\nu W/Q_1 = 0.97 \times 10^{-10} \text{ watt}.$$

Thus, optimum sensitivity is reached with a very small flow of power.

To evaluate the spectrometer sensitivity, let us take, as an example, $F = 100$, $\Delta f = 10$ cps and $kT = 4 \times 10^{-14}$ erg. Then Eq. (25) gives

$$n_{\min} = 3.4 \times 10^9 \text{ sec}^{-1}.$$

Since the number of molecules focused into the cavity may be as great as 10^{13} to 10^{14} sec^{-1}, a signal-to-noise ratio of 10^4 to 10^5 can be expected for the 3–3 line of ammonia. This theoretically expected sensitivity of the maser spectrometer has been demonstrated experimentally by observation of the magnetic hyperfine components of the quadrupole satellites with a signal-to-noise ratio of 10 to 100, using an amplifier band width of 40 cps.[4] A further estimate of the practical limit of sensitivity will be given below, after a discussion of cavity design.

C. Figure of Merit for Cavity Resonators

Consider now the effect of cavity design on the strength of coupling between molecules and the electromagnetic field, and hence on the spectrometer sensitivity. Equation (25) shows that it is not simply Q_0 which determines the sensitivity, but the quantity $Q_0 L/A$. The threshold condition for an oscillation to occur is also determined by the same factors. Furthermore, if the mode number n along the axis is not zero, but unity, the resonance line is broader by a factor $\pi^2/8$, and the coupling between molecules and electromagnetic field is correspondingly decreased. Hence a figure of merit M of the cavity resonator for producing induced transitions may be defined as

$$M = (LQ_0/A)(8/\pi^2)^n, \tag{26}$$

for $n = 0$ or 1.

Because Q_0 is roughly proportional to the radius, a, of the cylindrical cavity resonator near cutoff, while A is roughly proportional to a^2, the largest values of M are expected in the lower modes of resonance. Therefore, values of M are computed and compared for some of the lower modes. The results for a cylindrical cavity with $L = 12$ cm and a wavelength of 1.25 cm are shown in Table I. In this table ϵ is the skin depth multiplied by the specific permeability μ' of the wall material,

$$\epsilon = (c/2\pi)(\mu' \tau/\nu)^{\frac{1}{2}}. \tag{27}$$

TABLE I. Calculated values of parameters for cylindrical cavity resonator. $L = 12$ cm, $\lambda = 1.25$ cm.

Mode	Radius, a in cm	Narrow beam $A\epsilon/\pi a^2$	$M\epsilon$	Broad beam $A\epsilon/\pi a^2$	$M\epsilon$	Q_0
TE_{111}	0.37	0.48	12.2	1.00	5.9	6100
TM_{010}	0.48	0.27	28.4	1.00	7.7	10 800
TM_{011}	0.48	0.27	22.2	1.00	6.0	10 400
TE_{211}	0.61	∞	0	1.00	2.9	8100
TE_{011}	0.76	∞	0	1.00	4.1	17 800

[4] K. Shimoda and T. C. Wang, Rev. Sci. Instr. **26**, 1148 (1955).

Here τ is the electric resistivity. For copper at a frequency of 2.4×10^{10},

$$\epsilon = 4.27 \times 10^{-5} \text{ cm.} \tag{28}$$

The third and fourth columns in Table I assume a sharp narrow beam entering the cavity along its axis. The fifth and sixth columns are for a uniform intensity of beam throughout the cross section of the cavity. The actual values lie somewhere between these two extreme cases. The table indicates that the TM_{010} mode of the cylindrical cavity is the best with a figure of merit about three times that for the TE_{011} mode, which was used in most of the experiments reported earlier.[1,2]

Similar results for rectangular cavities with cross section $a \times b$ are shown in Table II. Although the TE_{011} mode in the rectangular cavity has a fairly large value of M, it should be noted that it is obtained with a small cross section. When the focused beam of molecules is not rather sharp, the value of M is counterbalanced by the loss of the beam. For a uniform beam of very large cross section, M times the cross-sectional area of the cavity is, in fact, a more appropriate figure of merit than M itself. Hence for a rectangular guide, the TM_{110} mode is probably preferable in most cases. This cavity is also convenient since the resonant frequency may be tuned by changing the width, a or b.

A cylindrical TM_{010} cavity for the 3–3 line of ammonia and a tunable TM_{110} mode rectangular cavity were constructed and tested. The holes to admit the beam of molecules were about 8 mm in diameter in both cavities, but the leakage of microwave energy from them was practically negligible compared with the losses through the wave guide and in the cavity walls. The cylindrical TM cavity was constructed for a maser oscillator and found, as may be expected from Table I, to produce oscillations with a flow of molecules about three times smaller than the minimum required for a TE_{011} mode with the same Q. The rectangular TM cavity was constructed to be tunable in the range from 22 500 Mc/sec to 26 400 Mc/sec. It had one wave guide with nearly optimum coupling, and the loaded Q was measured to be near 4000. This value is only a little lower than the expected value of loaded Q ($\frac{1}{2}Q_0$). It was found that a rather precise parallelism of walls is required for a large value of Q.

Increasing the length of the cavity will decrease the line width and increase M and sensitivity. However, the longer the cavity, the closer the resonant frequencies of
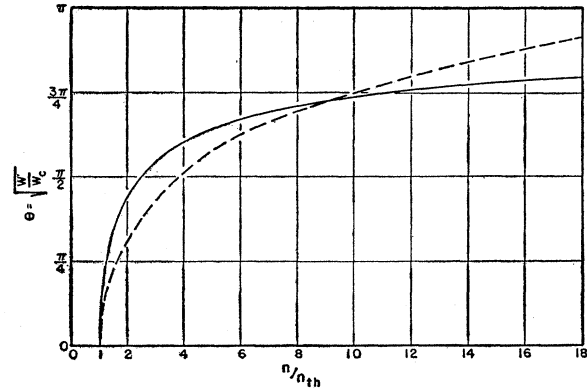

FIG. 1. The amplitude of oscillation, $\theta = (W/W_c)^{\frac{1}{2}}$ versus n/n_{th} as given by Eq. (31). The dashed line shows a crude estimate for nonuniform velocities.

the different axial modes and the more parallel stream of molecules is necessary. By the proper choice of coupling or by other devices, some modes can of course be suppressed.

An estimate of the practical limit of sensitivity of a maser spectrometer can be made with the following conditions. Using the TM_{010} mode of a cylindrical cavity resonator made of copper, one obtains $A = 0.5$ cm^2 and $Q_0 = 10^4$ with $L = 20$ cm and $\nu = 2.4 \times 10^{10}$ cps. Equation (25) then gives

$$n_{\text{min}} = 5.4 \times 10^7 \text{ sec}^{-1}$$

for the 3–3 line of ammonia, if an amplifier with $F = 10$ and $\Delta f = 0.1$ cps is used. Decreasing the velocity of molecules by reducing the temperature T_s of the source can increase the sensitivity slightly since, from (25), n_{min} is proportional to v and therefore to $T_s^{\frac{1}{2}}$. Also, cooling the cavity will increase the sensitivity by increasing Q_0.

III. THEORY OF THE MOLECULAR OSCILLATOR

A. Simple Theory of the Maser Oscillator

If a large number of molecules in the upper energy level is focused into the cavity resonator, a self-sustained oscillation can be obtained. The power loss from the microwave oscillation in the cavity is compensated by the emitted power from the molecular beam. This condition is given by the following formula, using Eq. (10) at $\omega = \omega_0$:

$$\frac{1}{Q} = \frac{\Delta P}{\omega W} = \frac{nh}{2\pi W_c} \frac{\sin^2\theta}{\theta^2}. \tag{29}$$

Because $\sin^2\theta/\theta^2 \simeq 1$ for small values of θ, the threshold rate of flow of molecules, n_{th}, required to build up oscillation in the cavity is

$$n_{\text{th}} = 2\pi W_c/Qh = hv^2 A/4\pi^2\bar{\mu}^2 LQ. \tag{30}$$

If n molecules enter the cavity per unit time, Eq. (29) may be written

$$n/n_{\text{th}} = \theta^2/\sin^2\theta. \tag{31}$$

TABLE II. Calculated values of parameters for rectangular cavity resonators. $L = 12$ cm, $= 1.25$ cm.

| Mode | Dimensions | | Narrow beam | | Broad beam | |
	a (cm)	b (cm)	A_e/ab	$M\epsilon$	$M\epsilon$	Q_0
TE_{011}	0.63	$\ll 0.63$	0.50	15.4	7.7	$\ll 5000$
TE_{011}	0.63	0.31	0.50	15.5	7.8	3700
TE_{011}	0.63	0.63	0.50	10.3	5.2	4900
TM_{110}	0.89	0.89	0.25	26.0	6.5	10 100

Figure 1 shows how the oscillation amplitude or the square root of W varies with n from Eq. (31). The output power P_0 is of course proportional to W, being given by

$$P_0 = \omega W/Q_1. \qquad (32)$$

The electromagnetic energy W in the cavity approaches a saturation value W_{sat}, when the intensity of the molecular beam is increased. From Eq. (31), this occurs when $\theta = \pi$, or

$$W_{sat} = \pi^2 W_c. \qquad (33)$$

The output power saturates at the same time. For typical conditions such as those described in Sec. II.C, the output power at saturation is about 10^{-9} watt. However, more power can be obtained, even if such saturation occurs, by increasing the output coupling (decreasing Q_1).

For a certain intensity of the molecular beam, the output coupling to get maximum output of molecular oscillation may be calculated by eliminating θ in the following two equations:

$$\tan\theta/\theta = 1 + (Q_0/Q_1),$$
$$\sin2\theta/2\theta = (2\pi W_c/nQ_0 h). \qquad (34)$$

These expressions come from using Eqs. (29) and (32) and optimizing P_0 with respect to Q_1. Using optimum coupling given by Eq. (34), the maximum output power can be calculated as

$$P_{0,max} = \omega W_c/Q_1. \qquad (35)$$

Although expressions (34) and (35) apply accurately only for the case of uniform velocity, they can serve as qualitative guides for the case of a distribution of molecular velocities.

If the velocity distribution is taken into account, the stored energy W and the output power would continue to increase with increasing the intensity of the molecular beam instead of reaching a saturation value. When molecules with different velocities are put in a common electric field, the induced emission from all molecules is superimposed. An estimated curve for the beam of molecules with nonuniform velocities is shown by the dashed curve in Fig. 1.

Experimental tests of a maser oscillator were made using ammonia molecules in the 3-3 state and a cavity of $Q=12\,000$ operating in the TE_{011} mode. The minimum focuser voltage required to start oscillation with a source pressure of 6 mm Hg was found to be 11 kv. For a TM_{010} mode cavity with $Q=10\,000$, the minimum focuser voltage for oscillation was 6.9 kv at the same source pressure. At a source pressure of 1.2 mm Hg, the critical focuser voltage was 15 kv for the same TM_{010} cavity. Since for a fixed source pressure, the number of molecules entering the cavity is roughly proportional to the square of the focuser voltage,[1,2] the above results show clearly that for the TM_{010} cavity, the threshold

number of molecules, n_{th}, was about three times smaller than that for the TE_{011} cavity.

The threshold intensities of the molecular beam calculated from Eq. (30) are $n_{th} = 4 \times 10^{13}$ sec^{-1} and 1×10^{13} sec^{-1} for the TE_{011} and TM_{010} cavities, respectively. The ratio of these two numbers agrees reasonably well with the above results. The actual number of effective molecules per unit time in the experimental apparatus at a source pressure of 6 mm Hg and focuser voltage of 15 kv is hence probably close to

$$n = 5 \times 10^{13}\ \text{sec}^{-1}.$$

This number is also consistent with estimates from the amount of total flow and directivity of the beam from the source.

B. Frequency Deviation of the Molecular Oscillator

Frequency shifts and noise in the molecular oscillation can be analyzed in detail by the method shown below. First the oscillating induced dipole moment of the molecular beam is calculated, assuming the electromagnetic field in the cavity. Secondly the electromagnetic field generated by the oscillating polarization of molecules is calculated and finally the assumed field and that generated by the molecules are made consistent, or equated in case there is no noise. In this section the noiseless case is treated.

The dipole moment of a molecule in the beam at anytime is

$$p = \int \psi^* \mathbf{u} \psi d\tau = a_2^* a_1 \mathbf{u}_{21} e^{i\omega_0 t} + a_1^* a_2 \mathbf{u}_{12} e^{-i\omega_0 t}, \qquad (36)$$

where it has been assumed that ψ has the form (1). With the perturbing field given by (3), the dipole moment of a molecule at $z = v(t - t_0)$ is, when one uses (4) and (5),

$$p(t) = \bar{\mu} e^{i\omega t} \frac{x}{[(\omega-\omega_0)^2 + x^2]^{\frac{1}{2}}}$$

$$\times \left(\frac{i}{2} \sin\{ [(\omega-\omega_0)^2 + x^2]^{\frac{1}{2}} (t-t_0) \} \right.$$

$$+ \frac{\omega-\omega_0}{[(\omega-\omega_0)^2 + x^2]^{\frac{1}{2}}} \sin^2\{ [(\omega-\omega_0)^2 + x^2]^{\frac{1}{2}} $$

$$\left. \times (t-t_0)/2 \} \right) + \text{complex conj.}$$

$$= p^\dagger e^{i\omega t} + p^{\dagger*} e^{-i\omega t}. \qquad (37)$$

The oscillating polarization density, $P(z)$, produced by molecules in the beam is given by

$$P(z) = (n/vA)p(z). \qquad (38)$$

The electric field E generated by the oscillating polarization may now be calculated. The polarization is

small enough that it may be considered as a small perturbation of the normal mode of oscillation of the cavity, or of the distribution of electric field, E_n. In this case, the frequency and the quality factor Q_m for the cavity with the molecular beam can be calculated from a result given by Slater[5] for a cavity containing a microwave current:

$$\frac{1}{Q_m}-2i\frac{\omega-\omega_c}{\omega_c}=\frac{1}{Q}+4\pi i\frac{\int P^\dagger\cdot E_n^{\dagger*}dV}{\int E^\dagger\cdot E_n^{\dagger*}dV}. \quad (39)$$

Here $\omega_c/2\pi$ is the resonant frequency and Q the loaded quality factor of the cavity without the beam. The electric field and polarization are expressed by the complex vector quantities: $E\cos\omega t=\frac{1}{2}Ee^{i\omega t}+\frac{1}{2}Ee^{-i\omega t}=E^\dagger e^{i\omega t}+E^{\dagger*}e^{-i\omega t}$ and $P=P^\dagger e^{i\omega t}+P^{\dagger*}e^{-i\omega t}$. Actually, E and P are parallel in the case considered, so that vector rotation is not important.

For a stationary state of oscillation, the damping should be zero and Q_m should be infinity, so that (39) becomes

$$\frac{1}{Q}+2i\frac{\omega-\omega_c}{\omega_c}=-4\pi i\int P^\dagger\cdot E_n^{\dagger*}dV\Big/\int E^\dagger\cdot E_n^{\dagger*}dV. \quad (40)$$

If the distribution of the polarization in the cross section of the cavity is proportional to the field distribution of the resonant mode, the following equation is obtained from the TM_{mn0} mode:

$$\int P^\dagger\cdot E_n^{\dagger*}dV\Big/\int E^\dagger\cdot E_n^{\dagger*}dV=\frac{2}{L}\int_0^L P^\dagger(z)\frac{dz}{E}. \quad (41)$$

If the distributions of P and E in the cross section are different, the above equation does not hold exactly, but the discrepancy of the distributions usually causes only a slight deviation from Eq. (41). Therefore the following result is obtained, assuming $Q[(\omega-\omega_c)/\omega_c]\ll1$:

$$\frac{1}{2}E=-i\frac{4\pi Q}{L}\left(1-2iQ\frac{\omega-\omega_c}{\omega_c}\right)\int_0^L P^\dagger(z)dz. \quad (42)$$

By using Eq. (38), Eq. (42) may be written

$$\frac{1}{2}E=-i\frac{4\pi nQ}{vAL}\left(1-2iQ\frac{\omega-\omega_c}{\omega_c}\right)\int_0^L p^\dagger(z)dz. \quad (43)$$

Since $p(z)$ has been given as a function of $x=E\bar\mu/\hbar$, the condition of stationary oscillation is obtained from Eqs. (43) and (37) as

[5] J. C. Slater, Revs. Modern Phys. 18, 441 (1946).

$$\frac{x}{2}=\frac{4\pi^2 Qn\bar\mu^2}{vhAL}\left(1-2iQ\frac{\omega-\omega_c}{\omega_c}\right)$$

$$\times\int_0^L\left\{\sin[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}\frac{z}{v}-i\frac{\omega-\omega_0}{[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}}\right.$$

$$\left.\times\left[1-\cos[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}\frac{z}{v}\right]\right\}dz, \quad \text{for } \omega-\omega_0\ll x. \quad (44)$$

From the real part of Eq. (44), the amplitude of oscillation $E=x\hbar/\bar\mu$ can be obtained. Remembering that $\theta^2=W/W_c$, or $\theta=Lx/2v$ from Eqs. (8) and (11), one can thus obtain Eq. (31) when $\omega=\omega_0$.

If $\omega-\omega_0$ is considerably smaller than the line width $(\omega-\omega_0\ll v/L$, or $\omega-\omega_0\ll x$, since x is comparable with $v/L)$, then $[(\omega-\omega_0)^2+x^2]^{\frac{1}{2}}$ can be approximated as x and the imaginary part of Eq. (44) gives

$$2Q\frac{\omega-\omega_c}{\omega_c}\int_0^L\frac{x}{v}\sin\frac{z}{v}dz$$

$$+\frac{\omega-\omega_0}{x}\int_0^L\left(1-\cos\frac{x}{v}z\right)dz=0. \quad (45)$$

Integration of Eq. (45) gives

$$\frac{\omega-\omega_0}{\omega_0}=2Q\frac{(v/L)}{\omega_0}\frac{1-\cos2\theta}{1-\sin2\theta/2\theta}\frac{\omega_c-\omega}{\omega_c}. \quad (46)$$

If the Q of the line is defined by

$$Q_l=\frac{\nu_0}{2\Delta\nu}=\frac{\nu_0}{(0.89v/L)}, \quad (47)$$

then Eq. (46) shows that the fractional deviation of the frequency of the molecular oscillation is

$$\frac{\omega-\omega_0}{\omega_0}=\frac{Q}{Q_l}\frac{1}{2.8}\frac{1-\cos2\theta}{(1-\sin2\theta/2\theta)}\frac{\omega_c-\omega_0}{\omega_0}. \quad (48)$$

This is similar to the expression previously given for "pulling" of the oscillation by the cavity.[2] However, it is more complete in allowing for saturation of the molecular response. If the oscillation is weak so that saturation does not occur, $\theta\ll1$ and Eq. (48) reduces to

$$\frac{\omega-\omega_0}{\omega_0}=1.07\frac{Q}{Q_l}\frac{\omega_c-\omega_0}{\omega_0}. \quad (49a)$$

Or, under more normal conditions when $\theta\simeq\frac{1}{2}\pi$, Eq. (48) is

$$\frac{\omega-\omega_0}{\omega_0}=0.72\frac{Q}{Q_l}\frac{\omega_c-\omega_0}{\omega_0}. \quad (49b)$$

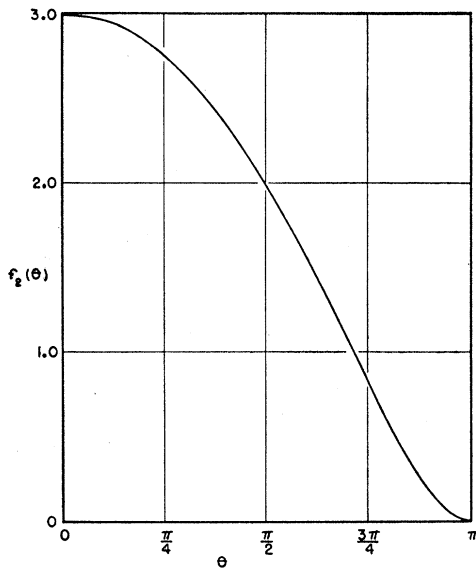The effect of saturation on frequency deviations of the above type is shown in Fig. 2. Since the factor

FIG. 2. A curve for $f_2 = (1-\cos2\theta)/(1-\sin2\theta/2\theta)$, giving the dependence of frequency pulling by cavity tuning on the amplitude of oscillation θ. See Eq. (48).

$(1-\cos2\theta)(1-\sin2\theta/2\theta)^{-1}$ decreases with increasing θ, the frequency deviation will decrease with increasing beam intensity. This may well be the origin of frequency variations with source pressure and with focusing field which have been observed. Both of these vary the number of molecules entering the cavity, and hence vary θ. Qualitative features of the observed variations agree with what may be expected from Eq. (48). However, it must be remembered that in deriving the precise form of Eq. (48), it was assumed that molecules of uniform velocity flow through the cavity and that the saturation does not depend on the distance of the molecules from the axis. Neither of these assumptions is strictly correct.

Bassov and Prokhorov have indicated[6] that if the "natural frequency" of the resonator is tuned to the center frequency of the spectral line, the frequency of molecular oscillation will be shifted by

$$(\omega - \omega_0)/\omega_0 \simeq -1/(2QQ_l).$$

It may be worthwhile to note that this type of frequency shift is, however, just due to the change of resonant frequency of the cavity by damping, since

$$\omega_c = \omega_{\text{nat}}(1 - 1/2Q^2),$$

where ω_{nat} is the "natural frequency" used by Bassov and Prokhorov, and ω_c is the actual resonant frequency as ordinarily measured. A frequency shift of $\omega/2Q^2$ in the cavity pulls the frequency by the amount $\omega/2QQ_l$. Hence if the actual resonant frequency is tuned to the frequency of the spectral line, there is no shift of the frequency of oscillation as indicated by expression (48).

[6] N. G. Bassov and A. M. Prokhorov, Discussions Faraday Soc. 19, 96 (1955).

There are several other types of systematic shifts in the frequency of oscillation. They include shifts due to unbalanced traveling waves in the cavity, changes in the resonant frequency of the cavity due to the polarization or dielectric constant of molecules in states other than those of interest, the effect of molecular collisions within the cavity, and also shifts due to Stark and Zeeman effects. Some of these will not be examined in much detail, since they are usually considerably less important than the "pulling" due to the cavity which was discussed above.

The frequency shift caused by the presence of traveling waves in the cavity which produce a net flow of power in one direction may be regarded as a type of Doppler shift. Thus if there is net power flow along the length of the cavity in the direction of the molecular velocity, the oscillation frequency may be expected to increase, since the frequency experienced by the moving molecules tends to correspond to the fixed molecular resonance frequency. Conversely, if there is a net flow of power in a direction opposite that of the molecular motion, the oscillation frequency should be somewhat decreased. The amount of frequency change due to traveling waves can in fact be approximately calculated by making a Fourier analysis of the apparent frequencies seen by the moving molecules. However, in order to be consistent with the formulation developed here and to show roughly the effect of saturation, this frequency shift is calculated in another, perhaps less transparent, manner.

When a small fraction of unbalanced traveling waves are present in a TM_{010} cavity, the electric field may be written

$$E = E_0 \cos\omega t + E_1 \cos(\omega t - \beta z)$$
$$\simeq (E_0 + E_1) \cos\omega t + E_1\beta z \sin\omega t, \quad (50)$$

where $E_1 \ll E_0$, $\beta L \ll 1$, and $E_1 \cos(\omega t - \beta z)$ represents a flow of microwave power along the cavity in the direction of the molecular velocity, i.e. the positive z direction if $E_1\beta$ and E_0 have the same sign. The oscillating dipole moment of the molecules in the cavity due to the stimulating field given by (50) may be expressed as

$$p_0^\dagger e^{i\omega t} + p_1^\dagger e^{i\omega t} + \text{complex conj.}$$

Here p_0 is the dipole moment produced by the field E_0 and p_1 the small change in dipole moment due to E_1. Allowing the electric field produced by the polarization of molecules in the cavity to equal the presupposed field (50), one obtains in analogy with Eq. (43)

$$\frac{1}{2}\left[x + x_1 - \frac{ix_1\beta}{L}\int_0^L z\,dz\right]$$
$$= \frac{-2in v}{\bar{\mu}n_{\text{th}}L^2}\left[1 - 2iQ\frac{\omega - \omega_c}{\omega_c}\right]\int_0^L (p_0^\dagger + p_1^\dagger)dz, \quad (51)$$

where $x = E_0\bar{\mu}/\hbar$ and $x_1 = E_1\bar{\mu}/\hbar$.

The frequency of oscillation may be obtained from the imaginary part of Eq. (51). If the cavity frequency ω_c is tuned very close to the oscillating frequency ω, the terms in $\omega - \omega_c$ may be neglected and the imaginary part of (51) is, to a good approximation,

$$-\frac{x_1 \beta}{2} \int_0^L z \, dz = -\frac{2n}{\bar{\mu} n_{\text{th}}} \frac{v}{L} \int_0^L \text{Re}(p_0^\dagger + p_1^\dagger) dz. \quad (52)$$

Now p_0 is obtained from (37) as

$$\text{Re}(p_0) = \bar{\mu} \frac{\omega - \omega_0}{x} \sin^2\left[\frac{x}{2}(t - t_0)\right], \quad (53)$$

and p_1 can be calculated by a perturbation calculation similar to that given in Sec. III.C. For this purpose, Eq. (67) has to be replaced by (50); then p_1 can be obtained from (78) by substituting x_1 and $-x_1 \beta v t$ for x_n' and x_n'', respectively, in that equation. One obtains,

$$\text{Re}(p_1) = \frac{\bar{\mu}}{2} \frac{x_1}{x} \beta v \left\{ (t - t_0) \sin[x(t - t_0)] \right.$$
$$\left. - \frac{2}{x} \sin^2\left[\frac{x}{2}(t - t_0)\right] \right\}. \quad (54)$$

Using (53) and (54), Eq. (52) can be written as

$$\frac{x_1 \beta L}{4} = \frac{n}{n_{\text{th}}} \frac{v^2}{L^2} \int_{t-L/v}^{t} \left\{ \frac{2}{x}\left(\omega - \omega_0 - \frac{x_1}{x}\beta v\right) \sin^2\left(\frac{x}{2}(t - t_0)\right) \right.$$
$$\left. + \frac{x_1}{x}\beta v (t - t_0) \sin[x(t - t_0)] \right\} dt_0.$$

After integration the following equation is obtained, using $\theta = Lx/2v$:

$$\frac{x_1}{x}\beta v \theta^2 = \frac{n}{n_{\text{th}}}\left[(\omega - \omega_0)\left(1 - \frac{\sin 2\theta}{2\theta}\right) \right.$$
$$\left. - \frac{x_1}{x}\beta v\left(\cos 2\theta + 1 - \frac{\sin 2\theta}{\theta}\right)\right].$$

Using n/n_{th} as given by Eq. (31), the fractional deviation of the frequency of oscillation is

$$\frac{\omega - \omega_0}{\omega_0} = \frac{x_1}{x} \frac{2\beta v}{\omega_0}\left[1 - \frac{\theta \sin^2\theta}{2\theta - \sin 2\theta}\right]. \quad (55)$$

The traveling waves in the cavity may be considered to be due to the loss through an output coupling hole at one end of the cavity with a value of Q_t, which can be shown to be

$$Q_t = \frac{\omega_0 W}{S} = \frac{2\pi^2 L}{\lambda^2 \beta} \frac{x}{x_1},$$

because the stored energy is $W = KLx^2$, and the power

flow S is given by $S = 2K(\beta c^2/\omega_0)x x_1$. When one uses this Q_t and the Q_l given in (47), Eq. (55) becomes

$$\frac{\omega - \omega_0}{\omega_0} = \frac{2\pi}{0.89} \frac{1}{Q_t Q_l} \frac{L^2}{\lambda^2}\left[1 - \frac{\theta \sin^2\theta}{2\theta - \sin 2\theta}\right]. \quad (56)$$

It should be noted from Eq. (55) that the sign of the shift reverses, if the velocity v of the beam changes sign, or hence if the direction of power flow due to the traveling waves reverses with respect to the molecular velocity. For small saturations, $\theta \ll 1$ and the bracketed factor in (55) or (56) is 0.25. For $\theta = \frac{1}{2}\pi$ it is 0.5. In a real oscillator, there is of course a distribution of molecular velocities and of effective values of θ. However, as an approximation, the bracketed factor may be assumed to equal 0.5, and this assumption gives

$$\frac{\omega - \omega_0}{\omega_0} \simeq \frac{3.5}{Q_t Q_l} \frac{L^2}{\lambda^2}. \quad (57)$$

This equation shows that, the closer the output coupling, the larger will be the frequency shift. The shift given by (57) may be seen to have roughly the same form as the shift mentioned by Bassov and Prokhorov[6] and discussed above. However, it has a quite different origin.

Assuming the values, $L = 10$ cm, $\lambda = 1.25$ cm, and $Q_l = 4 \times 10^6$, the fractional frequency shift produced by the traveling waves is about 2×10^{-9} for $Q_t = 3 \times 10^4$. If the load impedance changes by 10 percent, a change in frequency of about two parts in 10^{10} will be produced. The frequency shift caused by the output coupling can be very much reduced by placing the coupling hole just halfway between the two ends of the cavity, so that traveling waves progress from both ends of the cavity symmetrically.

In addition to the effect of the asymmetrical location of the output coupling holes described above, the variation of the molecular emission along the length of the beam in the cavity will give rise to corresponding unbalanced traveling waves and, therefore, a corresponding frequency shift. A shift of this kind will be of the same order of magnitude as that due to the asymmetrical coupling of the output hole, since the power emitted from the molecules is of the same order of magnitude as the output power. The molecular transition probability is connected with p_0 and p_1 in Eqs. (53) and (54) which vary along the length of the cavity. However, in obtaining the result given in Eq. (56), the molecular emission is effectively averaged over the molecular path in the cavity. A detailed analysis of the frequency shift due to the variation in beam emission has not been made. However, it should be noted that when little saturation occurs, the molecules deliver most of their energy toward the end of their path in the cavity, whereas for the highly saturated case, most of the energy is delivered near their entrance into the cavity. Thus the power delivered by the molecules flows in one direction for the case of little saturation, and in

the reverse direction for high saturation. In some intermediate condition, the frequency shift due to variation in molecular emission should become zero. A frequency shift due to this type of traveling waves could be reduced considerably by sending two similar molecular beams into opposite ends of the cavity. This method, with the output coupling at the middle of the cavity, would probably reduce frequency shifts due to traveling waves to considerably less than one part in 10^{10}, depending on the accuracy of the symmetry.

Consider now the change in resonant frequency of the cavity due to the presence of ammonia molecules in states other than those between which the desired transition takes place. The change in dielectric constant at frequency ν, due to the presence of other rotational state, is given by

$$\Delta\epsilon = \sum_i \frac{2\pi N_i}{3h} \frac{|\mu_i|^2}{\nu - \nu_i}, \tag{58}$$

where N_i is the density of the molecules in the ith rotational state in the cavity, μ_i the matrix element of the molecular dipole moment, and ν_i the inversion frequency of the ith rotational state. Since the only molecules which are focused are those in the upper inversion state, the contribution of molecules in the lower inversion states is neglected in the above expression. The effect of the (2,2) line on the frequency of the (3,3) line is considered as typical, since it is the nearest strong line. For an intensity of the molecular beam which is about ten times the threshold value for molecular oscillation at the (3,3) line, the change of dielectric constant due to the (2,2) line is estimated as $\Delta\epsilon(2,2) = 8\times10^{-9}$. In this estimate, $\mu_i = 1$ debye, $\nu - \nu_i = 1.5 \times 10^8$ cps and $N_i = n_i/vA = 4\times10^9$ cm^{-3} are used. Then the resonant frequency of the cavity is shifted by $-\frac{1}{2}\Delta\epsilon$, and the frequency of oscillation will be effected by about one part in 10^{11}. A precise calculation of this frequency shift of course requires a summation over all rotational states. However, its approximate magnitude is indicated by considering only the (2,2) state, which produces the largest shift.

It is difficult to isolate the aforementioned frequency shift experimentally, since the simultaneous existence of various states in the cavity cannot be prevented. However, if the relative populations of these states are changed, a small frequency shift might be expected. Since the focusing efficiency is a function of the rotational state as well as the focusing field, fluctuations in focuser voltage of ten percent may give rise to frequency shifts of about one part in 10^{12} due to changes in dielectric constant of gases in the cavity.

When ammonia molecules interact with each other the inversion spectrum is modified, and this effect produces the usual pressure broadening. In addition to a broadening of the inversion transition, there may be a frequency shift of the center of the line, which is proportional to the pressure broadening. Such a shift

has not been observed for any microwave line, and for the inversion spectrum of NH_3 it has been shown to be less than a few percent of the line width.[7] However, in principle, some shift of this type must occur. An upper limit for the resulting effect on the frequency of a maser oscillator can be obtained by assuming that the resonance width of molecules in the cavity is broadened by about ten percent due to collisions with other molecules in the cavity. The pressure broadening is probably less than this in most cases, and the maser would not oscillate very well if it were much larger. Assuming a resonance width $Q_l \simeq 5\times10^6$, the upper limit for the fractional shift is then a few thousandths of $1/(5\times10^6)$, or a few parts in 10^{10}. An additional type of frequency shift by molecular interaction which is proportional to the square of the pressure[8] has been observed, but this is quite negligible for the pressures used in the maser.

Every spectroscopic frequency is affected to some extent by electric or magnetic fields; these effects are referred to as Stark or Zeeman effects, respectively. Stark effects in the inversion spectrum of NH_3 have been discussed in a number of places.[9] They shift the inversion spectrum and disturb the coupling of the various nuclear spins to the molecular rotational motion. The uncoupling of nuclear spins is to a good approximation identical in the upper and lower inversion states, so that it does not shift the frequency of transitions which involve no change in the hyperfine states, and on which the oscillation frequency depends. The fractional change in frequency is hence approximately $(E\bar{\mu}/h\nu_0)^2$, where E is the field strength in esu, $\bar{\mu}$ the dipole matrix element, ν_0 the inversion frequency, and h is Planck's constant. Specifically, the fractional frequency shift for the most sensitive component ($M_J = 3$) of the NH_3 (3,3) line is very close to $10^{-9}E_v{}^2$, where E_v is in volts/cm. Thus if surface charges or varying contact potentials within the cavity produce field strengths as large as $1/30$ volt/cm, the resulting fractional change in frequency is about 10^{-12}.

"First-order" Zeeman effects split each hyperfine component of the inversion spectrum of NH_3 by approximately $g_J\mu_n H$, where g_J is the molecular g-factor, μ_n the nuclear magneton, and H the magnetic field in oersteds. In terms of frequency, this is about 1 kc/sec per oersted. The splitting is symmetric about the undisplaced transition, and hence for small H it corresponds only to some broadening of the line without a shift of its center.

"Second-order" Zeeman effects do shift the line center by a fractional amount which is roughly $(g_J\mu_n H/h\nu_0)^2$, or $2\times10^{-15}H^2$, where H is again in oersteds. Such a shift is usually negligible. It is about 10^6 times smaller than similar effects in atoms such as Cs, since the molecular magnetic moment is of the order of a nuclear magneton

[7] R. R. Howard and W. V. Smith, Phys. Rev. **79**, 128 (1950).
[8] H. Margenau, Phys. Rev. **76**, 1423 (1949).
[9] J. M. Jauch, Phys. Rev. **72**, 715 (1947); Coles, Good, Bragg, and Sharbaugh, Phys. Rev. **82**, 877 (1951).

rather than a Bohr magneton. Partial uncoupling of the nuclear spins from the molecular rotation occurs in molecules subjected to a magnetic field, but as in the case of Stark effect this does not change the frequency of oscillation because the transitions of interest involve no change in hyperfine energy.

First-order Zeeman effects can shift the frequency of oscillation if the oscillation does not occur very near the resonance frequency. For, if the cavity is tuned off resonance to a frequency ν_c, the oscillation frequency is "pulled" by approximately

$$\Delta\nu = (Q/Q_l)(\nu_c - \nu_0)$$

from expression (49). If the magnetic field H changes by a small amount, there is an incipient splitting which changes the line width slightly, and hence changes Q_l and $\Delta\nu$. This effect can be minimized by tuning so that $\nu_c = \nu_0$, and can in fact be useful as a test for the condition $\nu_c = \nu_0$, when oscillation should occur very near resonance.

C. Random Noise in the Maser Oscillator

Two kinds of effects give random fluctuations in the output of a maser oscillator: one comes from fluctuations in the number of molecules in the beam, which may be called "shot" noise, and the other from random fluctuations of the fields inside the cavity due to thermal noise and to "zero-point" fluctuations. We shall consider first the shot noise.

Let the number of molecules which may radiate and which enter the cavity in a time interval t be

$$N = nt.$$

The fluctuation in this number is then $\sqrt{N} = \sqrt{(nt)}$. The output of the maser oscillator is

$$P_0 = Knh\nu, \tag{59}$$

where K depends on θ and Q_l/Q, but is normally of the order of unity. The shot effect may hence be expected to produce an average fluctuation in power during a time t of approximately

$$(\langle \Delta P_0^2 \rangle_{Av})^{\frac{1}{2}} = KN^{\frac{1}{2}}h\nu/t = Kh\nu(n/t)^{\frac{1}{2}}.$$

The fractional fluctuation of amplitude of the electric field E is, from (59) and the above equation

$$\frac{(\langle \Delta E^2 \rangle_{Av})^{\frac{1}{2}}}{E} = \frac{1}{2(nt)^{\frac{1}{2}}}. \tag{60}$$

It must be remembered, however, that the field strength cannot change much more rapidly than the time required for a molecular transition to occur, or than the inverse of the frequency width of the molecular response. This time is approximately

$$\tau = 2Q_l/\omega. \tag{61}$$

In a typical case, it has a value $\tau = 10^{-4}$ sec and $n = 10^{14}$

sec^{-1}, so that the fractional fluctuation is at most $(\langle \Delta E^2 \rangle_{Av})^{\frac{1}{2}}/E \simeq 5 \times 10^{-6}$ which is small enough usually to be negligible. The smallness of this fluctuation results from the large number, i.e., $n\tau = 10^{10}$, of molecules in the cavity at all times.

In addition to an amplitude fluctuation, there is a phase fluctuation during the correlation time τ of the oscillation which is approximately equal to the fractional change in amplitude, or

$$(\langle \Delta\phi^2 \rangle_{Av})^{\frac{1}{2}} = \frac{1}{2(n\tau)^{\frac{1}{2}}}. \tag{62}$$

Such phase fluctuations which occur during each correlation time add together in a random way to produce a root-mean-square phase change of

$$t(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}} = \left(\frac{t}{\tau}\langle \Delta\phi^2 \rangle_{Av}\right)^{\frac{1}{2}} = \frac{1}{2\tau}\left(\frac{t}{n}\right)^{\frac{1}{2}},$$

after a time interval t. Hence the fluctuation of frequency is obtained by using (59) and (61) as

$$\frac{(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}}}{\omega_0} = \frac{1}{4Q_l}\left(\frac{Kh\nu}{P_0 t}\right)^{\frac{1}{2}} \simeq \frac{1}{Q_l}\left(\frac{h\nu}{P_0 t}\right)^{\frac{1}{2}}. \tag{63}$$

The magnitude of frequency fluctuations due to this effect will be shown below to be considerably smaller than that due to thermal noise at normal temperatures.

The molecular oscillator builds up initially from the thermal noise fields present in the cavity. During the stationary state of oscillation, thermal noise in the cavity shifts the frequency and amplitude of oscillation in a random manner. Although the precise behavior of thermal fluctuations in the cavity depends on the presence of molecular beam amplification, a rough estimate which ignores the effect of the saturation of the beam will first be described.

Assuming that the noise power, $kT\Delta f$, is independent of the oscillation in the cavity, the average fractional fluctuation of amplitude and phase of the molecular oscillation is approximately

$$(\langle \Delta E^2 \rangle_{Av})^{\frac{1}{2}}/E \simeq (\langle \Delta\phi^2 \rangle_{Av})^{\frac{1}{2}} \simeq (kT\Delta f/P_0)^{\frac{1}{2}}, \tag{64}$$

where Δf is determined by the length of the time t used for measuring the frequency of the oscillator. When the frequency is observed during a time interval t, the transfer function, that is the frequency characteristics for the frequency modulation noise fluctuating at a frequency f, is proportional to

$$(1/f)\sin(\pi f t).$$

This transfer function gives the effective band width of

$$\Delta f \simeq 1/t. \tag{65}$$

From (64), (65), and (61) the fractional fluctuation of

the frequency of oscillation due to thermal noise is

$$\frac{(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}}}{\omega_0} \simeq \frac{1}{2Q_l}\left(\frac{kT}{P_0 t}\right)^{\frac{1}{2}}. \qquad (66)$$

The maximum frequency fluctuation which can occur is found by setting t equal to the minimum response time τ in (66), which gives

$$\frac{(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}}}{\omega_0} \simeq \frac{1}{2Q_l}\left(\frac{kT}{P_0 \tau}\right)^{\frac{1}{2}}.$$

For typical conditions this quantity is approximately 10^{-10}, which is the fractional frequency change occurring in the time $\tau \simeq 10^{-4}$ sec. For $t=1$ sec, this is reduced to 10^{-12}.

Comparison of Eqs. (63) and (66) shows that

$$\frac{\langle \Delta\omega^2 \rangle_{Av(\text{shot noise})}}{\langle \Delta\omega^2 \rangle_{Av(\text{thermal noise})}} \simeq \frac{h\nu}{kT}.$$

At room temperature and microwave frequencies this ratio is about 3×10^{-3}; hence shot noise can be neglected compared to thermal noise.

It should be noted that spontaneous emission of radiation by the molecules also produces random fluctuations. Their nature is, however, precisely the same as that of fluctuations due to thermal radiation and considerably smaller. For our purposes, spontaneous emission may be considered as an added thermal fluctuation corresponding to a temperature of $h\nu/k$, which for microwave frequencies is only one or two degrees absolute.

The theory of fluctuations of the oscillator, as simplified above, might lead one to suppose that successive changes in both phase and amplitude add in a random manner, so that the amplitude of the oscillating electric field would at times go through zero. This does not actually occur for the same reason that it does not occur in most other oscillators: the nonlinearities in the oscillator's response ensure that the amplitude stays within certain narrow limits rather than increasing or decreasing in a random way. However, as in most other oscillators, the phase variation is quite random and may increase indefinitely as indicated by the above discussion in a way which is proportional to $\sqrt{t}$.

To allow for the effect of molecular amplification and nonlinearities on the thermal noise, a more complete theory is described. In the following discussion the noise field, $E_n(t)$, due to thermal noise is calculated by a perturbation technique, assuming that $E_n(t)$ is small compared to the field E_0 produced by emission of radiation from the molecules.

The electric field in the cavity resonator may be written

$$E = E_0 \cos\omega t + E_n(t), \qquad (67)$$

where ω is the frequency of oscillation, E_0 is assumed to

be constant, and $E_n(t)$ is a small field associated with thermal noise. If one uses

$$x = E_0 \bar{\mu}/\hbar,$$
$$x_n(t) = 2E_n(t)\bar{\mu}/\hbar, \qquad (68)$$

the Schrödinger equation for the wave function $\psi = a_1\psi_1 + a_2\psi_2$ requires

$$\dot{a}_1 = \tfrac{1}{2}i\{x[e^{i(\omega-\omega_0)t} + e^{i(\omega+\omega_0)t}] + x_n e^{-i\omega_0 t}\}a_2,$$
$$\dot{a}_2 = \tfrac{1}{2}i\{x[e^{-i(\omega-\omega_0)t} + e^{-i(\omega+\omega_0)t}] + x_n e^{+i\omega_0 t}\}a_1. \qquad (69)$$

If the oscillation occurs near the molecular resonance, then $(\omega-\omega_0)^2 \ll x^2$, and $(\omega-\omega_0)t \simeq 0$ for the time of passage of the molecules through the cavity. Furthermore $e^{i(\omega+\omega_0)t}$ fluctuates very rapidly during the time of transition when a_1 and a_2 are changing more slowly, so that the terms in (69) including this exponential average to zero and may be neglected to a good approximation. Terms such as $x_n e^{-i\omega_0 t}$ cannot similarly be omitted because x_n varies rapidly with time in such a way that the product $x_n e^{-i\omega_0 t}$ does not necessarily average to zero during the transition.

Let the solution of (69) for $x_n=0$ be $a_1^{(0)}$ and $a_2^{(0)}$, which are given in expressions (4) and (5) as

$$a_1^{(0)} = -\sin[\tfrac{1}{2}x(t-t_0)],$$
$$a_2^{(0)} = i\cos[\tfrac{1}{2}x(t-t_0)], \qquad (70)$$

when $(\omega-\omega_0)^2 \ll x^2$ as assumed above. Here t_0 is the time at which a molecule enters the cavity. The solution when $x_n \neq 0$ may be written

$$a_1 = a_1^{(0)} + a_1^{(1)},$$
$$a_2 = a_2^{(0)} + a_2^{(1)}, \qquad (71)$$

and then (69) becomes

$$\dot{a}_1^{(1)} = \tfrac{1}{2}i(xa_2^{(1)} + x_n e^{-i\omega_0 t}a_2^{(0)}),$$
$$\dot{a}_2^{(1)} = \tfrac{1}{2}i(xa_1^{(1)} + x_n e^{i\omega_0 t}a_1^{(0)}), \qquad (72)$$

if terms in the product of the two small quantities $a_1^{(1)}$ and x_n or $a_2^{(1)}$ and x_n are neglected.

The quantity x_n in (69) or (72) due to noise may be written

$$x_n = 2x_n' \cos\omega t - 2x_n'' \sin\omega t, \qquad (73)$$

in which case expression (67) for the field becomes

$$E = \frac{\hbar}{\bar{\mu}}(x+x_n')\cos\left(\omega t + \frac{x_n''}{x}\right), \qquad (74)$$

which makes it evident that x_n' gives the amount of amplitude modulation and x_n''/x the phase modulation. Making the substitutions (73) and

$$a_1^{(1)} = Ae^{\frac{1}{2}ix(t-t_0)} - Be^{-\frac{1}{2}ix(t-t_0)},$$
$$a_2^{(1)} = Ae^{\frac{1}{2}ix(t-t_0)} + Be^{-\frac{1}{2}ix(t-t_0)}, \qquad (75)$$

into Eq. (72), one finds

$$\dot{A}=-\tfrac{1}{4}[x_n{}'+ix_n{}''e^{-ix(t-t_0)}],$$
$$\dot{B}=+\tfrac{1}{4}[x_n{}'+ix_n{}''e^{+ix(t-t_0)}]. \qquad (76)$$

Again in obtaining expression (76) it must be remembered that very rapidly varying terms such as $e^{i(\omega+\omega_0)t}$ average to zero and may be omitted to a good approximation.

Now the oscillating dipole moment of the molecule is given by

$$p=\bar{\mu}e^{i\omega_0 t}(a_1{}^{(0)}a_2{}^{(0)*}+a_1{}^{(1)}a_2{}^{(0)*}+a_1{}^{(0)}a_2{}^{(1)*})$$
$$+\text{complex conj.}$$
$$=p_0{}^\dagger e^{i\omega t}+p_n{}^\dagger(t)+\text{complex conj.} \qquad (77)$$

Using Eqs. (70) and (75) together with (76), one obtains

$$p_0{}^\dagger=\tfrac{1}{2}i\bar{\mu}\sin(xt),$$

$$p_n{}^\dagger(t)=-\tfrac{1}{2}\bar{\mu}e^{i\omega_0 t}\left[\int x_n{}''\cos\{x(t-t_0)\}dt\right.$$

$$\left.-i\cos\{x(t-t_0)\}\int x_n{}'dt\right]. \qquad (78)$$

The electric field produced by the polarization of molecules can be calculated in the same way as described in the former section, using (39), (41), and (43). The total field is the sum of fields produced by molecules in the beam and those generated by thermal agitation in the wall of the cavity. It may be written, as $Q[(\omega-\omega_c)/\omega_c]\ll1$,

$$E^\dagger=-i\frac{4\pi Qn}{vAL}\int_0^L p^\dagger(z)dz+e_n{}^\dagger, \qquad (79)$$

where $E^\dagger$ is defined as in Eqs. (39) and (67), and $e_n{}^\dagger e^{i\omega_0 t}+e_n{}^{\dagger*}e^{-i\omega_0 t}$ is the primary thermal noise which would occur without the presence of the molecular beam. After some manipulation, Eq. (79) can be rewritten as

$$\frac{x}{2}+\frac{x_n{}^\dagger}{2}-\frac{\xi_n{}^\dagger}{2}=\frac{4\pi^2 Q_n\bar{\mu}^2}{hAL}\left[\int_{t-L/v}^t\sin(x(t-t_0))dt_0\right.$$

$$+i\int_{t-L/v}^t\left\{\int_{t_0}^t x''\cos(x(t-t_0))dt\right.$$

$$\left.\left.-i\cos(x(t-t_0))\int_{t_0}^t x_n{}'dt\right\}dt_0\right], \qquad (80)$$

where $x_n{}^\dagger$ is defined by $x_n{}^\dagger e^{i\omega t}+x_n{}^{\dagger*}e^{-i\omega t}$, that is, $x_n{}^\dagger=x_n{}'+ix_n{}''$ from (73). In addition,

$$\xi_n{}^\dagger(t)=2\frac{e_n{}^\dagger\bar{\mu}}{\hbar}=\xi_n{}'(t)+i\xi_n{}''(t). \qquad (81)$$

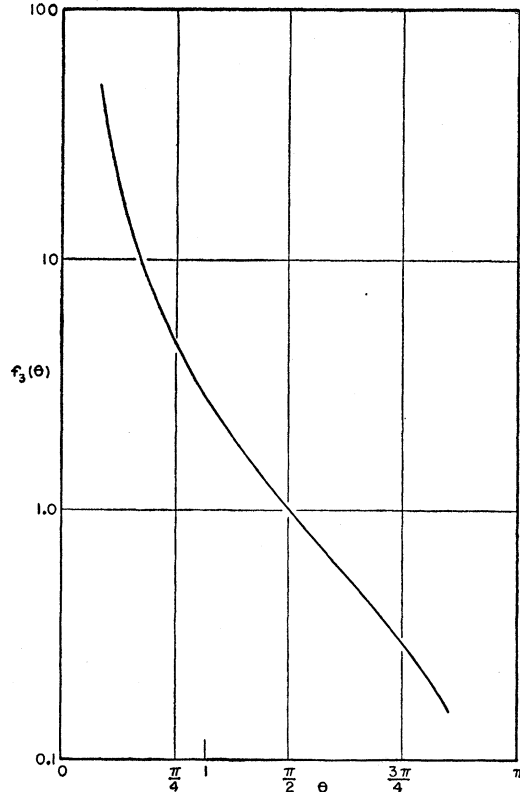The first integral in (80), which gives the field strength of precisely the frequency $\omega_0/2\pi$ and which is

FIG. 3. A curve for $f_3=1/(1-\theta\cot\theta)$, giving the dependence of amplitude fluctuations due to thermal noise on the amplitude of oscillation θ. See Eq. (84).

independent of the noise, yields the same results as were found above in Sec. III.B. The remaining terms represent the noise fluctuations and can be written as

$$x_n{}'=\left[2\frac{n}{n_{\text{th}}}\frac{v^2}{L^2}\int_{t-L/v}^t\cos\{x(t-t_0)\}\int x_n{}'dtdt_0\right]+\xi_n{}', \qquad (82)$$

$$x_n{}''=\left[2\frac{n}{n_{\text{th}}}\frac{v^2}{L^2}\right.$$

$$\left.\times\int_{t-L/v}^t\int_{t_0}^t x_n{}''\cos\{x(t-t_0)\}dtdt_0\right]+\xi_n{}''. \qquad (83)$$

From these equations, amplitude noise $x_n{}'$ and phase noise $x_n{}''/x$ can be computed.

Equation (82) may be simply solved for the important case of noise components which differ from the frequency of oscillation by an amount smaller than the line width. For these components, $x_n{}'$ varies much more slowly than does $\cos(xt)$, so that as a first approximation (82) can be integrated by assuming $x_n{}'$ to be constant. Using expression (31) for n/n_{th} and $\theta=xL/2v$, one obtains the following from this integration:
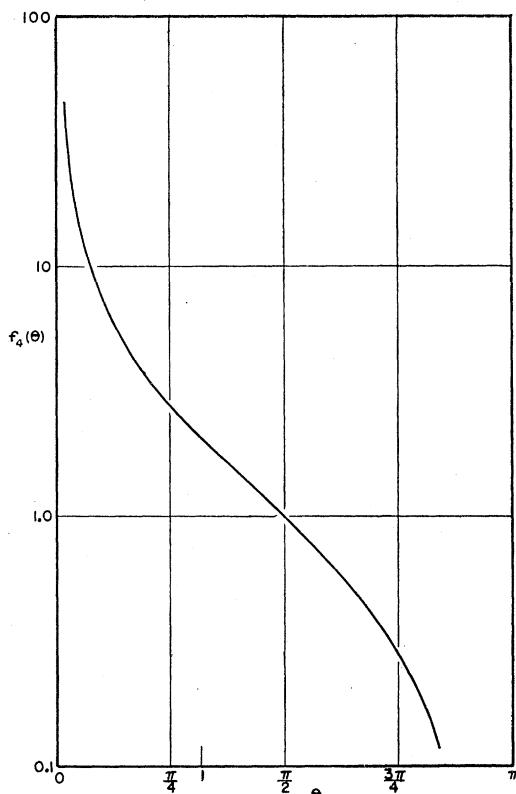
$$2x_n{}'=\frac{\xi_n{}'}{1-\theta\cot\theta}. \qquad (84)$$

FIG. 4. A curve for $f_4 = \pi \sin^2\theta/(2\theta - \sin 2\theta)$, giving the dependence of frequency fluctuations due to thermal noise on the amplitude of oscillation θ. See Eqs. (92).

This expression shows that the magnitude of AM noise depends strongly on the amplitude of oscillation, as can be seen from Fig. 3. Since the primary noise, ξ_n', is independent of frequency, x_n' is also independent of frequency near the center of the molecular transition where the approximations involved in obtaining (84) are valid. The frequency distribution of noise outside this range can also be estimated in a similar way by using $a_1^{(0)}$ and $a_2^{(0)}$ as given by Eqs. (4) and (5) so long as $\omega - \omega_0 \ll \omega_0$, but such a calculation would be very tedious. Presumably the spectrum of amplitude noise is not far different from the line shape.

Evaluation of the fluctuation of phase from Eq. (83) is somewhat more difficult. If the phase fluctuation is computed from (83) by assuming that x_n'' in the integrand is constant, then (83) becomes, after integration, $x_n'' = x_n'' + \xi_n''$. This indicates that x_n'' must approach infinity, corresponding to the indefinite increase in the phase error with increasing time as was found in the simpler discussion above. In order to obtain a finite result for x_n'' and to examine the way in which it approaches large values with increasing time, it is necessary to allow variation of x_n'' in the integrand.

Consider the thermal noise $\xi_n''(t)$ in a small band width df about a frequency which differs from the oscillator frequency by f. Then ξ_n'' which is a function

of time can be written in the form

$$\xi_n'' = \int_0^\infty \xi_n''(f) \cos(2\pi f t + \delta) df, \qquad (85)$$

where δ is a phase angle. x_n'' must have the similar form,

$$x_n'' = \int_0^\infty x_n''(f) \cos(2\pi f t + \delta') df,$$

where $x_n''(f)$ and δ' may be assumed to be constants with respect to time.

Substitution of these expressions into Eq. (83) gives, with the assumption $f \ll v/\pi L$,

$$x_n''(f) = \xi_n''(f) \frac{v}{\pi f L} \frac{\sin^2\theta}{(1 - \sin 2\theta/2\theta)}, \qquad (86)$$

and $\delta - \delta' = \tfrac{1}{2}\pi$.

Because ξ_n'' represents a component in the primary thermal noise given by (79) and (81), $\langle [\xi_n''(f)]^2 \rangle_{Av}$ is actually independent of frequency f. From (85), (81), (68), and (64), one has

$$\frac{\langle [\xi_n''(f)]^2 \rangle_{Av}}{x^2} = \frac{\tfrac{1}{2}\langle \xi_n^2(f) \rangle_{Av}}{x^2} = \frac{2\langle e_n^2(f) \rangle_{Av}}{E_0^2} = \frac{kT df}{\omega_0 W/Q}, \qquad (87)$$

where $\xi_n(f)$ and $e_n(f)$ represents the amplitude of Fourier components at the frequencies $(\omega_0/2\pi) \pm f$. Thus the mean square of x_n'' is obtained as

$$\frac{\langle x_n''^2 \rangle_{Av}}{x^2} = \frac{QkT}{\omega_0 W} \left\{ \frac{v}{\pi L} \frac{\sin^2\theta}{[1 - (\sin 2\theta/2\theta)]} \right\}^2 \int_{f_{min}}^{f_{max}} \frac{df}{f^2}, \qquad (88)$$

where f_{min} and f_{max} are the minimum and maximum frequencies respectively for the range of values of f which are considered. Both f_{max} and f_{min} must be much less than the line width in order that Eq. (86) be approximately correct. Assuming $f_{max} \gg f_{min}$, expression (88) is

$$\frac{\langle x''^2 \rangle_{Av}}{x^2} = \frac{QkT}{\omega_0 W f_{min}} \left[\frac{v}{\pi L} \frac{2\theta \sin^2\theta}{2\theta - \sin 2\theta} \right]^2. \qquad (89)$$

If fluctuations are observed during a time interval t, the minimum frequency of fluctuation which can be observed is approximately

$$f_{min} \simeq 1/2t. \qquad (90)$$

From (74), the root mean square of the fractional fluctuation of frequency is

$$\frac{(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}}}{\omega_0} = \frac{(\langle x''^2 \rangle_{Av})^{\frac{1}{2}}/x}{\omega_0 t}. \qquad (91)$$

Thus one obtains from (89), (90), and (91)

$$\frac{(\langle \Delta\omega^2 \rangle_{Av})^{\frac{1}{2}}}{\omega_0} = \frac{v}{\pi \omega_0 L} \left(\frac{2QkT}{\omega_0 W t} \right)^{\frac{1}{2}} \frac{2\theta \sin^2\theta}{2\theta - \sin 2\theta}.$$

Using $\theta^2 = W/W_c$, and $Q_l = \omega_0 L/(0.89 \times 2\pi v)$, the frequency fluctuations can be written

$$\frac{(\langle\Delta\omega^2\rangle_{Av})^{\frac{1}{2}}}{\omega_0} = \frac{0.16}{Q_l}\left(\frac{QkT}{\omega_0 W_c t}\right)^{\frac{1}{2}}\frac{\sin^2\theta}{2\theta - \sin 2\theta}. \quad (92)$$

This is very similar to the approximate expression (66) obtained earlier, since $\omega_0 W/Q$ is a typical value of emitted power from the molecular beam for $\theta = 1$. Dependence of the frequency fluctuation on power is given by the last factor in (92), which is plotted in Fig. 4. This figure shows that the amount of FM noise increases by a large factor when the oscillation is weak. It must be remembered that Eq. (92) is not exact because of the approximation used in its derivation. In addition, the distribution of molecular velocities and matrix elements over molecules in the beam tends to make expression (92) even less accurate. However, it probably represents the major features of noise fluctuations reasonably well.

As a typical example, let $\theta = \frac{1}{2}\pi$, $W_c = 2\times10^{-11}$ erg, $Q = 5000$, and $Q_l = 5\times10^6$, then

$$\frac{(\langle\Delta\omega^2\rangle_{Av})^{\frac{1}{2}}}{\omega_0} \sim 10^{-13}t^{-\frac{1}{2}}.$$

This calculated fluctuation is somewhat smaller than the upper limit set by experimental observations.[2]

Coherent Neutron Scattering Amplitudes of Krypton and Xenon, and the Electron-Neutron Interaction*

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(Received February 23, 1956)

The coherent scattering amplitudes of krypton and xenon for thermal neutrons have been measured by a comparison method involving reflection of a neutron beam from liquid mirrors. The results were $(7.68\pm0.19)\times10^{-13}$ cm for krypton and $(5.10\pm0.17)\times10^{-13}$ cm for xenon. When these results are used with the scattering measurements of Hamermesh, Ringo, and Wattenberg, one obtains (3900 ± 800) ev for the electron-neutron interaction.

INTRODUCTION

THE electron-neutron interaction[1] is of obvious interest as a test of any complete theory of elementary particles and it has received considerable theoretical[2] and experimental[3-5] attention. At present there is particular interest in the question of whether the interaction shows any structure-dependent effects in addition to a relativistic effect pointed out by Foldy.[6] What is probably the most accurate experimental result[5] is that the interaction is equivalent to a square well of

3860±370 ev depth when the radius, by convention, is taken as $e^2/(mc^2)$. The relativistic effect is approximately 4000 ev, so quite accurate measurements are needed to decide if there is any interaction beyond this. The electron-neutron interaction is so much smaller than the interaction of the neutron with the nucleus or the magnetic dipole interaction that it is very hard to be sure in any measurement that some obscure residual effect of these large interactions is not affecting the experiment. For this reason it seems very desirable to have the measurement made by several methods that are as independent as possible.

The method used by Hamermesh, Ringo, and Wattenberg[4] is considerably different from that of Hughes *et al.* It was devised by Fermi and Marshall[7] and consists of a measurement of the anisotropy of the scattering of a beam of thermal neutrons by gaseous krypton and xenon. The electron-neutron interaction affects this anisotropy through the electronic form factors (more properly, the atomic scattering factors) of the gases. Most of the anisotropy is due to the center-of-mass

* Work supported by the U. S. Atomic Energy Commission.
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[1] In this article we mean by electron-neutron interaction the interaction additional to the large one between the magnetic moments of the electron and neutron. This last interaction is given by the ordinary magnetic dipole interaction expression and accounts for the magnetic scattering of neutrons.
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