

## Solid-State Maser Amplifier\*

ALAN L. McWHORTER AND JAMES W. MEYER

Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts

(Received August 14, 1957)

The operation of a solid-state maser amplifier at 2800 Mc/sec is described. A dual-frequency cavity containing paramagnetic potassium chromicyanide in an isomorphous cobalt diluent is used at 1.25°K. The experimental observations of the maser both as an amplifier and as an oscillator are compared with theory.

### I. INTRODUCTION

A SOLID-STATE maser of the type proposed by Bloembergen<sup>1</sup> has been operated both as an amplifier and as an oscillator at 2800 Mc/sec, using  $K_3Co(CN)_6$  containing 0.5% Cr as the paramagnetic salt. This material is particularly suited for maser application by virtue of its unusually long spin-lattice relaxation time, which was found to be 0.2 sec at 1.25°K by resonance saturation techniques.

The upper three of the four energy levels of the  $Cr^{+++}$  ion were used, with the energy level spacing suitably adjusted by means of the magnitude of the dc magnetic field and its orientation with respect to the crystalline electric field of the salt. Spin state populations were inverted by saturating the resonance absorption at 9400 Mc/sec.

The amplifier, regenerative in nature, has as a result much narrower band width than one would expect from a casual consideration of the circuit  $Q$ 's involved. For simplicity the measurements reported below were made

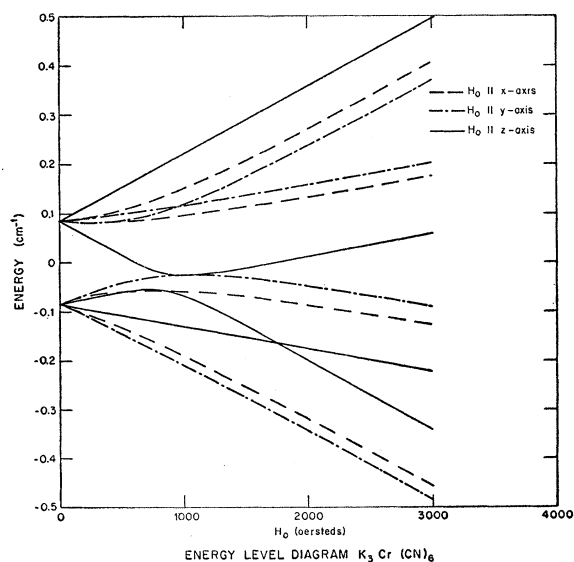


FIG. 1. Energy level diagram  $K_3Cr(CN)_6$  with orientation of magnetic axes as parameters.

\* The research reported in this document was supported jointly by the Army, Navy, and Air Force under contract with Massachusetts Institute of Technology.

<sup>1</sup> N. Bloembergen, Phys. Rev. **104**, 324 (1956).

with a relatively high  $Q$  reflection cavity. To achieve the large band widths inherent in the width of the paramagnetic resonance line without sacrificing gain, a very low  $Q$  structure, e.g., a slow wave structure, containing a much larger volume of the paramagnetic salt would have been necessary. In spite of its band-width limitation, however, the present circuit shows that a solid-state maser can be made to operate in reasonable agreement with the theoretical predictions.

### II. ENERGY LEVELS

Baker, Bleaney, and Bowers<sup>2</sup> have interpreted the paramagnetic resonance spectra of  $K_3Cr(CN)_6$  as arising

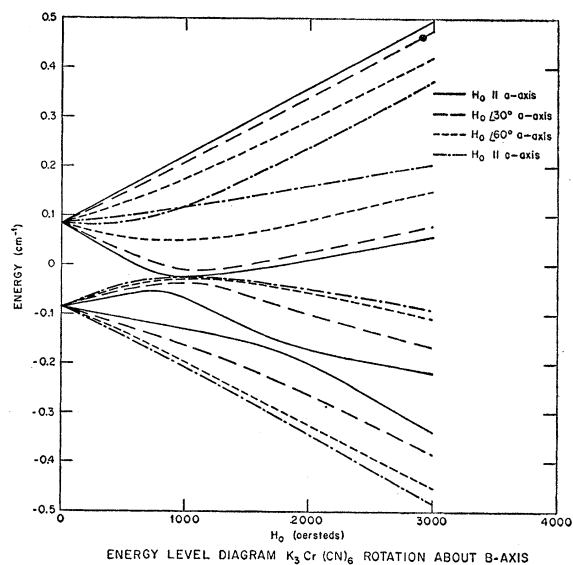


FIG. 2. Energy level diagram  $K_3Cr(CN)_6$  rotation about  $b$  axis.

from two magnetically similar but differently oriented complexes per unit cell, with the spin Hamiltonian

$$H = \beta \mathbf{H} \cdot g \cdot \mathbf{S} + D[S_z^2 - \frac{1}{3}S(S+1)] + E(S_x^2 - S_y^2),$$

where for cobalt as the diluent,  $D = 0.083 \text{ cm}^{-1}$ ,  $E = 0.011 \text{ cm}^{-1}$ , and  $g$  is approximately isotropic and equal to 1.99. The direction cosines between the magnetic axes ( $x$ ,  $y$ , and  $z$ ) and the pseudo-orthorhombic crystalline axes

<sup>2</sup> Baker, Bleaney, and Bowers, Proc. Phys. Soc. (London) **B69**, 1205 (1956).

( $a$ ,  $b$ , and  $c$ ) are given as

	$x$	$y$	$z$
$a$	0.104	0	0.994
$b$	$\pm 0.994$	0	$\mp 0.104$
$c$	0	1	0

Several energy level diagrams computed from this spin Hamiltonian are shown in Figs. 1 and 2. Within experimental accuracy, our preliminary measurements agree with these curves both in the 3 cm and 10 cm wavelength regions.

As these data indicate, several combinations of magnetic field and crystal orientation can be chosen which will permit operation at the selected frequencies, 2800 Mc/sec and 9400 Mc/sec. The combination which gave the best results in the initial investigation, and which was used for the measurements reported below, is shown in Fig. 3. The desired splitting was achieved

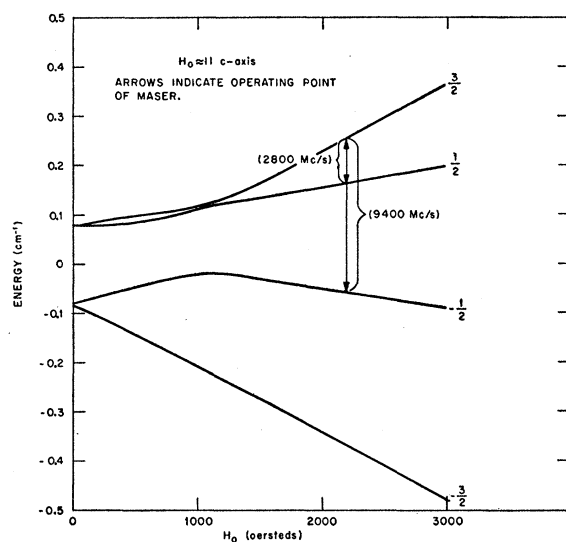


FIG. 3. Energy level diagram  $K_3Cr(CN)_6$  showing point of maser operation.

by means of a slight rotation, about the  $a$  axis, of the  $c$  axis from the dc magnetic field.

### III. APPARATUS AND METHOD

#### A. Microwave Apparatus

Because of its essential simplicity, a fixed-tuned, dual-mode coaxial microwave cavity was employed. Minor adjustment of the splitting of the energy levels to correspond to the resonant frequencies of the cavity was done by small shifts in crystal orientation. The cavity is one-half wavelength long at 2800 Mc/sec when operated in the  $TEM$  mode with the sample in place and with the remainder of the cavity volume filled with liquid helium. At 9400 Mc/sec, the cavity operated in the  $TE_{113}$  mode, which was especially chosen to reduce cross coupling of 9400-Mc/sec power into the coaxial

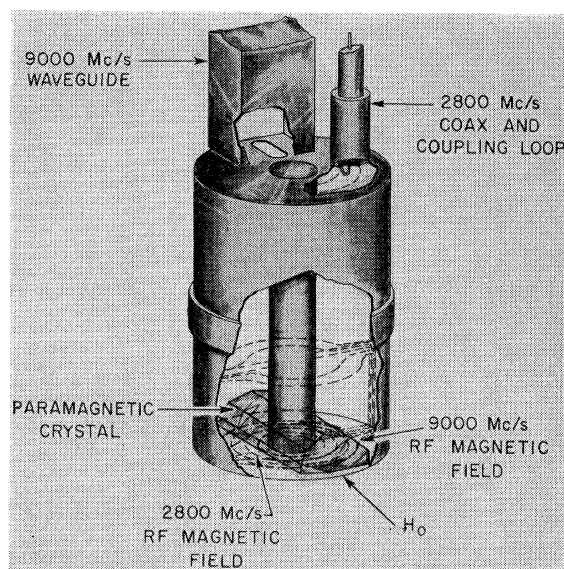


FIG. 4. Dual-frequency maser cavity.

line employed for 2800-Mc/sec operation. The cross coupling was further reduced by means of low-pass filters in the external coaxial line. The resulting magnetic field configurations and cavity features are shown in Fig. 4. Saturating power was coupled to the cavity by means of a section of silver-plated stainless steel wave guide terminated by a magnetic coupling hole. The size of this hole was adjusted to give approximately critical coupling when the paramagnetic resonance was saturated. The 2800-Mc/sec coaxial line was coupled to the cavity by means of a loop. The extent of coupling could be adjusted both by rotation and depth of immersion of the loop in the cavity.

The low-temperature head, Fig. 5, was installed in a suitable double Dewar system. Thermal isolation of the cavity provided by the stainless-steel wave guide and coaxial line was sufficiently good to permit several hours of operation at 1.25°K with approximately one liter of liquid helium.

#### B. Sample Preparation

Single crystals were grown from an aqueous solution of amounts of cobalt and chromium potassium cyanide appropriate to the chromium concentration desired. Crystals were prepared with chromium concentrations of from 0.1% to 2%. Standard crystal-growing techniques were employed to produce crystals of more than one square centimeter in cross section by three to five centimeters long.

#### C. Crystallography

The single crystals had two principle growth habits, as indicated in Fig. 6, one which produced a crystal form elongated in the  $c$ -axis direction with prominent  $m$

faces  $\{110\}$ ,<sup>3</sup> the other which produced flat hexagonal plates with prominent  $a$  faces  $\{100\}$ . The correct orientation was determined from goniometric measurements on the well-formed faces.

#### IV. EXPERIMENTAL RESULTS

##### A. Amplifier Characteristics

The operation of the maser as an amplifier was investigated by applying the input power to the cavity through a directional coupler as illustrated in Fig. 7. This arrangement permitted gain-band-width measurements on the reflection cavity type amplifier through its single coaxial coupling line without the use of a circulator. The gain was determined by the amount of attenuation needed in the maser output line to maintain a constant signal amplitude at the spectrum analyzer. This additional attenuation in the output line, together with the ferrite isolator, served the additional purpose of keeping any power reflected from the spectrum analyzer from reaching the maser and being reamplified.

The band width was taken as the total frequency

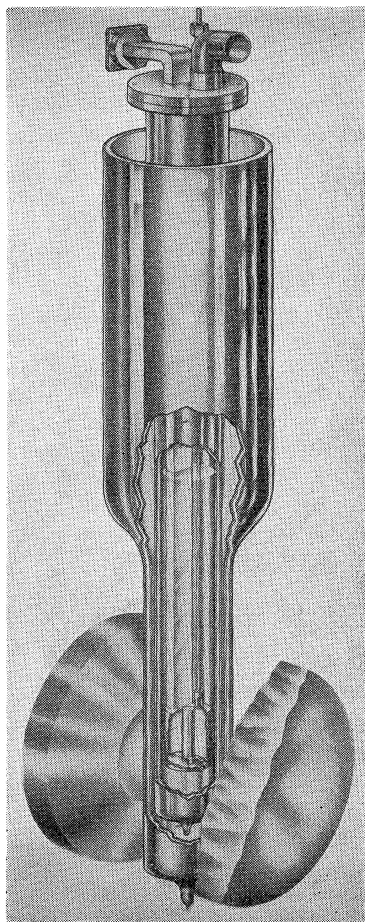


FIG. 5. Low-temperature head and double Dewar system.

deviation required to reduce the amplifier power output to one-half its midband value. Band widths were measured on the spectrum analyzer after calibrating its frequency axis with the modulating scheme shown in the block diagram.

The results of the gain-band-width measurements are shown in Fig. 8. Parametric curves of both gain and band width are plotted as a function of 9400-Mc/sec power for two different values of 2800-Mc/sec external  $Q$ , which was adjusted by means of the degree of coupling. With still higher external  $Q$  it was possible to achieve gains of 30 db or more with only 1 mw of saturating power, although the maser would then oscillate at the larger saturating powers. Stable gains of 37 db with 25 kc/sec band width were also possible. In all of these cases the band widths were limited by the  $Q$  of the associated circuitry and not by the intrinsic band width of the paramagnetic resonance, which here was in the 30–50 Mc/sec region.

Observations of the gain as a function of input 2800-Mc/sec power revealed the expected decreased gain as the difference in populations established by the saturating power is affected by the signal power. There was no change in gain when the signal power was increased from  $10^{-11}$  to  $10^{-10}$  watt, but thereafter the gain diminished and the band width increased.

##### B. Oscillator Characteristics

The initial investigation of the maser as an oscillator was made by using a frequency-modulated probing signal applied to the coaxial coupling line. The frequency of the probing oscillator is swept by the time base of the oscilloscope and the power reflected from

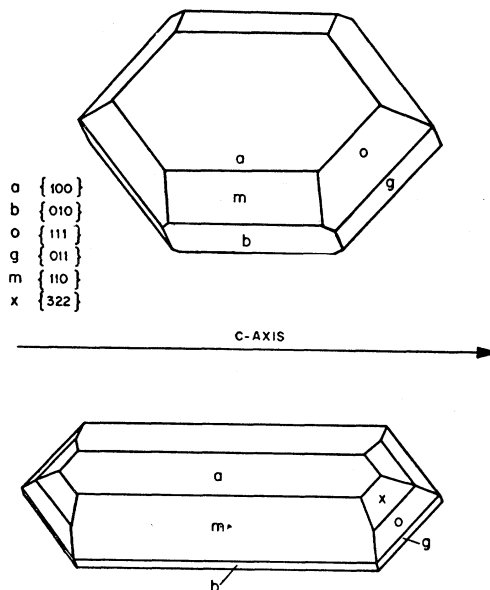
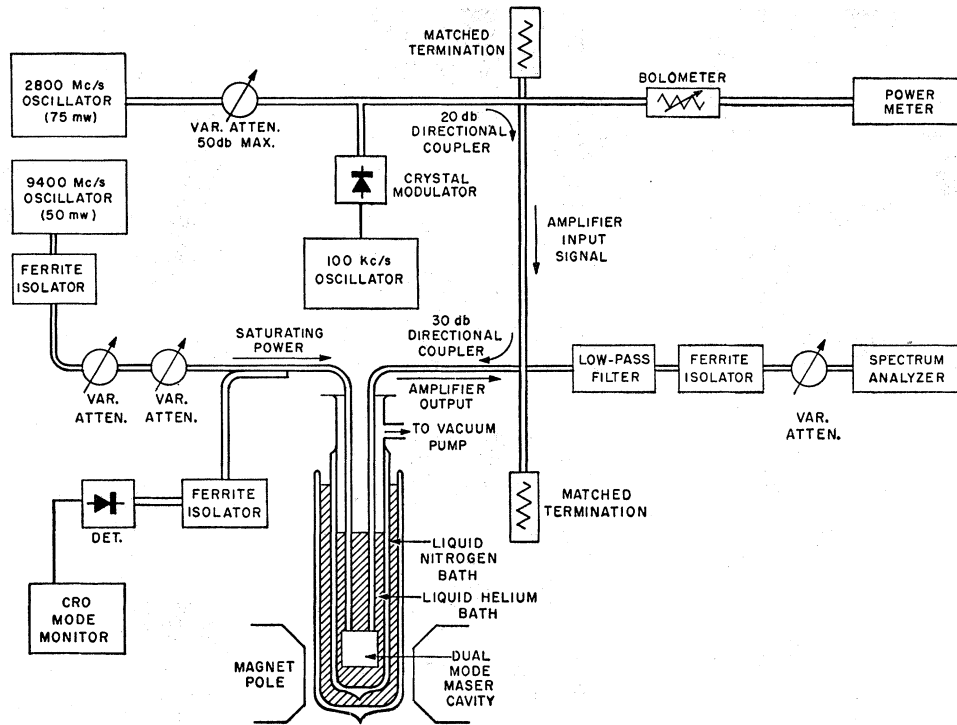


FIG. 6. Typical growth habit and crystal forms,  $K_3Co(CN)_6-K_3Cr(CN)_6$ .

<sup>3</sup> P. Groth, *Chemische Kristallographie* (W. Engelmann, Leipzig, 1906), Vol. 1, p. 422.

FIG. 7. Block diagram of microwave instrumentation.



the cavity is displayed on the  $y$  axis as a function of frequency. In Fig. 9(a) we see the absorption resulting from the 2800-Mc/sec microwave resonance centered in the klystron mode pattern. With the magnetic field adjusted for paramagnetic resonance, the power reflected from the undercoupled cavity increases, [Fig. 9(b)]. The application of 9400-Mc/sec power [Fig. 9(c)] shows how the negative resistance produced by maser action improves the  $Q$  of the cavity, which in turn improves the coupling although no changes were made in the coupling loop adjustments. Further increase of saturating power enhances this effect [Fig. 9(d)], and in Fig. 9(e) the maser is beginning to produce power at 2800 Mc/sec. In Fig. 9(f) the beat signal between the output of the oscillating maser and the frequency-modulated probe signal is clearly seen with the video detector system.

The output of the oscillating maser was also observed on a spectrum analyzer in the absence of an input 2800-Mc/sec signal. Maser power out as a function of saturating input power is shown in Fig. 10. The efficiency,  $P_o$  (2800 Mc/sec)/ $P_i$  (9400 Mc/sec) is also given. The maximum efficiency obtained as operated here was  $-28.5$  db, or 0.14%.

The realization of an oscillator with such small amounts of saturating power demonstrates the usefulness of chromium cobalticyanide as a maser material by reason of its advantageously long relaxation time. The relaxation time is not so long, on the other hand, that the maser will not handle reasonable signal input powers as an amplifier.

C. Relaxation Times

The product of the phenomenological relaxation times, spin-lattice  $T_1$  and spin-spin  $T_2$ , were measured using the power saturation of paramagnetic resonance technique.<sup>4</sup> Values for  $T_2$  were then obtained from line

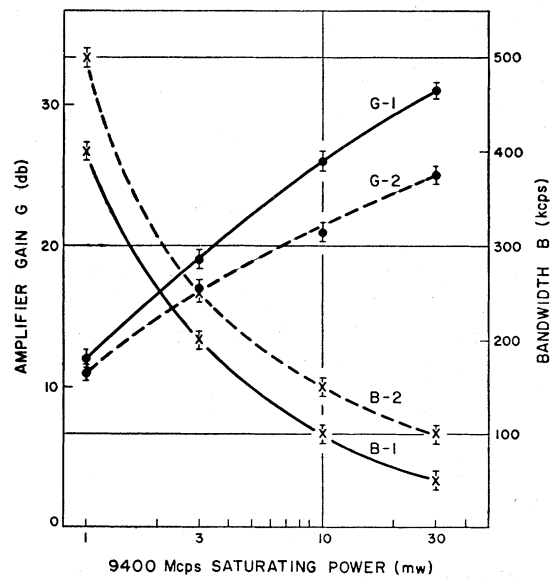


FIG. 8. Gain and band-width curves of maser amplifier with degree of coupling as a parameter.

<sup>4</sup> Bloembergen, Purcell, and Pound, Phys. Rev. **73**, 679 (1948); A. M. Portis, Phys. Rev. **91**, 1071 (1953); A. H. Eschenfelder and R. T. Weidner, Phys. Rev. **92**, 869 (1953).

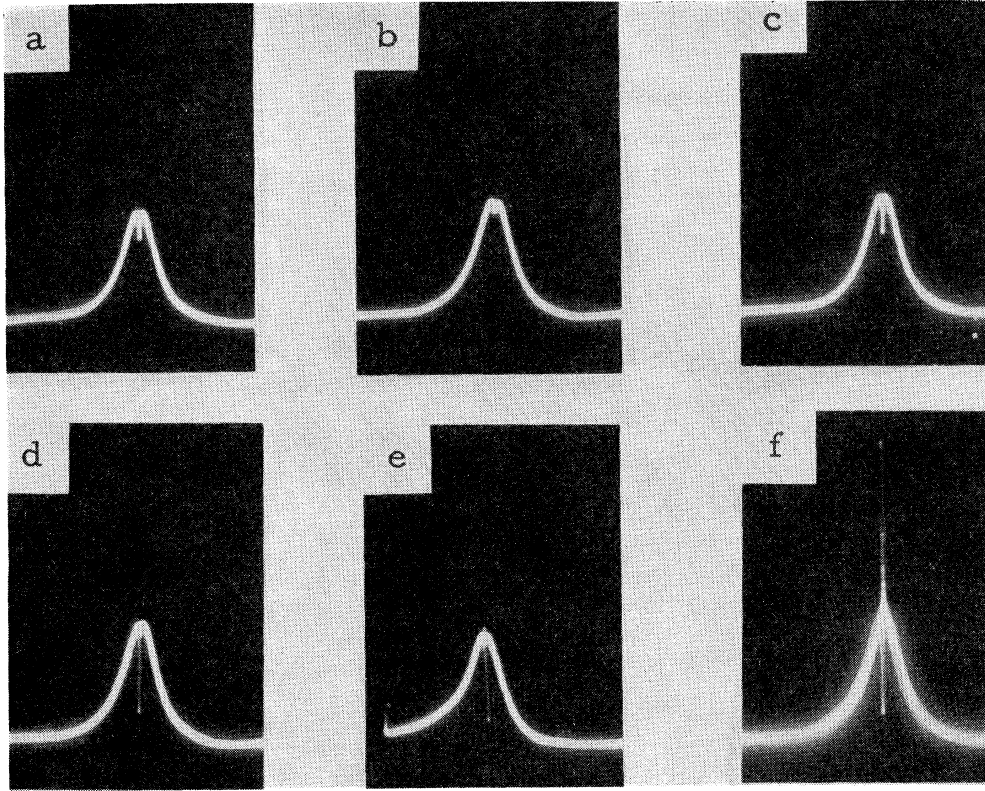


FIG. 9. Oscilloscope display of maser oscillator operation.

width measurements, and values of  $T_1$  subsequently obtained. Data were taken for the three crystalline axes,  $a$ ,  $b$ ,  $c$ , and the three  $\Delta m_s = 1$  transitions at 1.25°K. Although the spin-spin relaxation time varied considerably, the value of  $T_1$  remained 0.2 second to within experimental error for all three transitions along the principal axes.  $T_2$  meanwhile took on values from  $4 \times 10^{-9}$  sec to  $9 \times 10^{-9}$  sec and was largest for the  $-\frac{1}{2} \rightarrow +\frac{1}{2}$  transition at all orientations. The sample contained 0.5% chromium concentration.

## V. DISCUSSION

The gain of a reflection-cavity type maser amplifier, connected with directional couplers and ferrite isolators as was done here for measurement purposes, or with a circulator as would be used for the practical amplifier, is simply the ratio of the reflected to the incident power. In terms of the external and total cavity  $Q$ 's, the gain can therefore be expressed as

$$G = [(1/Q_e - 1/Q_c) / (1/Q_e + 1/Q_c)]^2, \quad (1)$$

where

$$1/Q_c = 1/Q_u + 1/Q_M \quad (2)$$

gives the total cavity  $Q$  in terms of the unloaded  $Q$  and the magnetic  $Q$  resulting from the paramagnetic resonance. The magnetic  $Q$  is positive for resonance absorption and negative for stimulated emission, and the unloaded  $Q$  includes all other losses such as those in

the cavity walls, and the dielectric losses in the salt. At low temperatures,  $Q_u$  will generally be so large that it can be neglected in the gain formula. (Our measured  $Q_u$  was 23 000 at 1.25°K.) When one makes this approximation and uses the absolute value of the magnetic  $Q$ , the gain is

$$G = [(Q_e + |Q_M|) / (Q_e - |Q_M|)]^2. \quad (3)$$

The band width of the amplifier is given by the operating frequency  $f$  divided by the loaded  $Q$ , which with the above approximation gives

$$B \doteq f(1/Q_e - 1/|Q_M|). \quad (4)$$

Consequently,

$$G^{\frac{1}{2}} B = f(Q_e + |Q_M|) / Q_e |Q_M|. \quad (5)$$

For a high-gain amplifier one adjusts the coupling so that  $Q_e$  almost equals  $|Q_M|$ . Under this condition

$$G^{\frac{1}{2}} B \doteq 2f/Q_e \doteq 2f/|Q_M|. \quad (6)$$

Hence, as was pointed out by Strandberg,<sup>5</sup> the square root of the gain times band width should be approximately a constant for a given volume of the salt in a given cavity configuration. Experimentally, this relation is obeyed very closely, the value being about  $1.8 \times 10^6$  sec<sup>-1</sup>. By decreasing the external  $Q$ , gain can be traded for band width, but at a considerable sacrifice of the former. Since the first part of Eq. (6) shows that the

<sup>5</sup> M. W. P. Strandberg, Phys. Rev. **106**, 617 (1957).

band width of the loaded cavity without the magnetic material is reduced by  $G^{\frac{1}{2}}/2$  when it is operating as an amplifier, it is apparent that heavy coupling is required for even modest band width.

Equation (3) shows that the gain of a high-gain amplifier is very sensitive to small relative changes of  $Q_e$  and  $Q_M$ . As a result, slight changes in coupling or degree of saturation can change the gain and band width by large amounts even though  $G^{\frac{1}{2}}B$  remains sensibly constant. This characteristic also indicates that the designer of a pulsed-type two-level maser will have to exercise considerable ingenuity to prevent wide excursions of gain and band width during the amplifying period.

An expected theoretical value for the  $G^{\frac{1}{2}}B$  product can be calculated approximately in the following way: Expressing the magnetic  $Q$  in terms of the cavity volume  $V_c$ , the average rf magnetic field  $\langle H_c^2 \rangle$ , the operating frequency  $\nu_{32}$ , and the magnetically absorbed or emitted power  $P_M$ , we have

$$1/|Q_M| = 4P_M/\nu_{32}\langle H_c^2 \rangle V_c, \quad (7)$$

where, in Bloembergen's<sup>1</sup> notation,

$$P_M \doteq \frac{Nh^2\nu_{32}}{3kT} \left( \frac{w_{21}\nu_{21} - w_{32}\nu_{32}}{w_{32} + w_{21}} \right) W_{32} \quad (8)$$

in the approximation that one has full saturation at frequency  $\nu_{13}$  and small power at the amplifying frequency  $\nu_{32}$ . The formula for  $W_{32}$ , as given by Bloembergen, Purcell, and Pound,<sup>4</sup> is

$$W_{32} = \frac{1}{4}\gamma^2(H_1/2)^2 g(\nu)(S+M)(S-M+1). \quad (9)$$

At the operating temperature of 1.25°K, the density of spins in the upper three levels of the chromium quartet is approximately  $10^{19}/\text{cm}^3$ . Assuming the  $w$ 's are equal, as experimental results indicate, using approximately a 10% filling factor and the relation  $g(\nu)_{\text{max}} = 2T_2$  ( $T_2 = 6 \times 10^{-9}$  sec), we have for the  $G^{\frac{1}{2}}B$  product a value  $2.6 \times 10^6 \text{ sec}^{-1}$ , which is in reasonable agreement with the experimental value,  $1.8 \times 10^6 \text{ sec}^{-1}$ .

It is also possible to compare the power output of the maser oscillator with the theoretically predicted output. Assuming full and homogeneous saturation of the resonance at  $\nu_{31}$ , and a large signal amplitude at  $\nu_{32}$ , the magnetic power is given by<sup>1</sup>:

$$P_M = (Nh^2\nu_{32}/3kT)(w_{21}\nu_{21} - w_{32}\nu_{32}). \quad (10)$$

Taking  $N = 3.9 \times 10^{19}$ , the population of the upper three levels at 1.25°K, and  $w_{21} = w_{32} \doteq 1/2T_1$ , we obtain the value 8.7 microwatts, which is to be compared with the experimentally obtained four microwatts. Cavity and coupling losses will account for much of the discrepancy between the two values.

It should be pointed out that a long spin-lattice relaxation time, which permits amplification with low

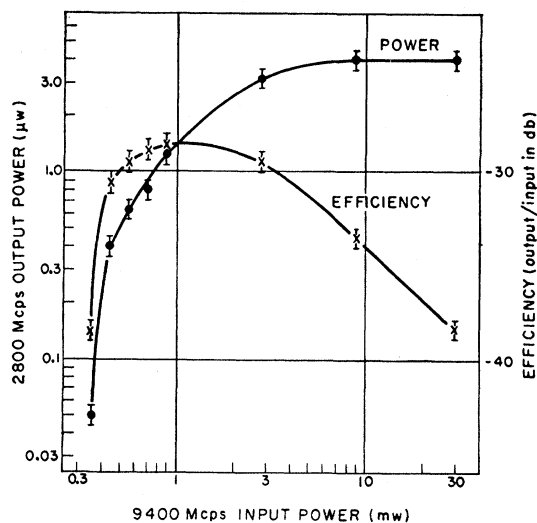


FIG. 10. Power output characteristics of maser oscillator.

saturation power, limits in a like manner the maximum output of the maser as an oscillator.

The operation of a solid state maser of this type places somewhat stringent requirements upon the paramagnetic material. At least three energy levels are necessary, and the paramagnetic ion must be in a field of sufficient asymmetry to produce mixed states and allow  $\Delta m_s = 2$  quantum jumps. Not only should the interionic distances be great enough to reduce the spin-spin interaction to a point where saturation is practicable, but also there must be a sufficient number of spins participating in the maser action to overcome the cavity and coupling losses. Moreover, the residual orbital momentum should not be too great in order to assure a long spin-lattice relaxation time at low temperatures. Further, one would like substantial control over the separation of the energy levels by means other than changing the magnitude of the dc magnetic field (e.g., by varying the orientation of the magnetic field relative to the crystalline electric field), in order to have more freedom in the choice of both saturating and operating frequencies. These requirements severely limit the choice of a maser salt among paramagnetic substances with characteristics reported in the literature.<sup>6,7</sup> Of the two working substances investigated in detail by us, zinc fluosilicate containing a small percentage of  $\text{Ni}^{++}$  and potassium cobalticyanide containing a small percentage of  $\text{Cr}^{+++}$ , only the latter could be used for successful operation of the maser described here.

The other paramagnetic salt mentioned by Bloembergen, gadolinium ethyl sulfate, was not tried because it offered much less flexibility of choice of saturating and operating frequencies. Moreover, as outlined by

<sup>6</sup> B. Bleaney and K. W. H. Stevens, *Reports on Progress in Physics* (The Physical Society, London, 1953), Vol. 16, p. 108.

<sup>7</sup> K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 304.

Scovil, Feher, and Seidel,<sup>8</sup> the relaxation time between the upper two levels had to be drastically altered in order to produce oscillations. This further limits the frequency flexibility. Because of previous experience with the salt,<sup>9</sup> zinc fluosilicate containing a small percentage of  $\text{Ni}^{++}$  was first tried by us in a maser designed to operate at 1400 Mc/sec while saturating at 9000 Mc/sec. We were unable to get this maser to operate for two main reasons: first, because the spin-lattice relaxation time is of the order of  $10^{-4}$  sec, considerable power is required to saturate the resonance; secondly, the line width in the diluted salt was inhomogeneously broadened by a distribution of crystalline electric fields. As a result of the inhomogeneous broadening only  $T_2^*/T_2$  of the paramagnetic spin concentration was available for participation in the maser action, where  $T_2^*$  is the time associated with the total width of the inhomogeneous line.

An attempt to improve the probability of operation by going to a higher amplifying frequency, 2800 Mc/sec, was also unsuccessful.

The nickel ion is still attractive, however, from other points of view. Because it is a spin triplet without nuclear magnetic moment, ions need not be sacrificed by distribution over unused energy levels. Its integral spin  $S=1$  leaves one level,  $m_s=0$ , unchanged by the application of the external magnetic field, thereby enhancing its tunability. Further work is in progress to find a suitable salt and diluent for the nickel ion, as part of a more general program which includes the paramagnetic resonance spectroscopy of other materials.

Published paramagnetic resonance data reveal few

salts which appear to be suitable materials for maser operation. The cyanides were especially attractive because the reduction of line width by dilution is not limited by the proton magnetic moment as it is in the hydrated salts. In the latter case, lines of a few oersteds width can only be achieved by deuteration. Moreover, the spin-lattice relaxation time is long, which when combined with a reduced spin-spin interaction requires little power for saturation of the resonance.

Thus far, no experimental measurement of the noise figure of this maser amplifier has been made, but work is in progress to evaluate this most important characteristic.†

#### VI. ACKNOWLEDGMENTS

We should like to thank Professor Bloembergen for disclosing his concept of a solid-state maser prior to its publication, and for subsequent discussions of related problems. The assistance of Dr. S. H. Autler with some of the experimental work is also gratefully acknowledged. We are indebted to Dr. Harry C. Gatos and Mr. L. J. Gordon of this laboratory, and Dr. C. W. Wolfe of Boston University for furnishing the single crystals of potassium cobaltcyanide containing chromium. The physical preparation and orientation of the samples was done by Dr. S. A. Kulin, and Mr. E. P. Warekois. Assistance in the mechanical design of the cavities was provided by Mr. A. M. Rich. Finally, we should like to acknowledge profitable discussions with Dr. R. H. Kingston and Dr. H. J. Zeiger and the encouragement and continued support of Dr. Benjamin Lax.

† *Note added in proof.*—A preliminary sequence of measurements, giving an upper limit of 20°K for the noise temperature of the maser, has now been completed: McWhorter, Meyer, and Strum, Phys. Rev. **108**, 1642 (1957).

<sup>8</sup> Scovil, Feher, and Seidel, Phys. Rev. **105**, 762 (1957).

<sup>9</sup> J. W. Meyer, Ph.D. thesis, University of Wisconsin, 1955 (unpublished).

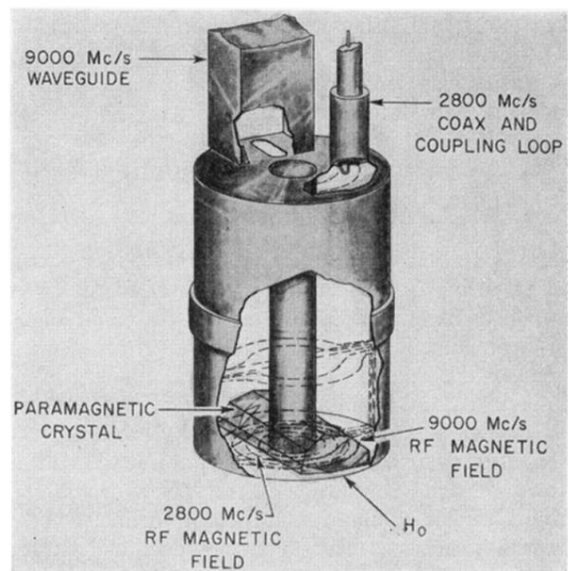


FIG. 4. Dual-frequency maser cavity.



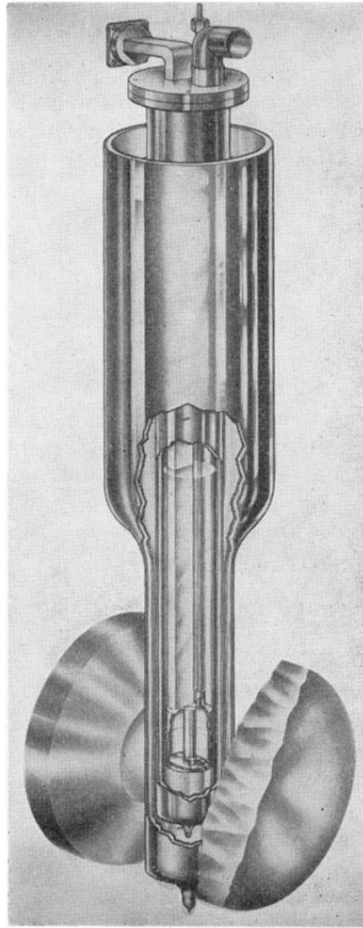


FIG. 5. Low-temperature head and double Dewar system.

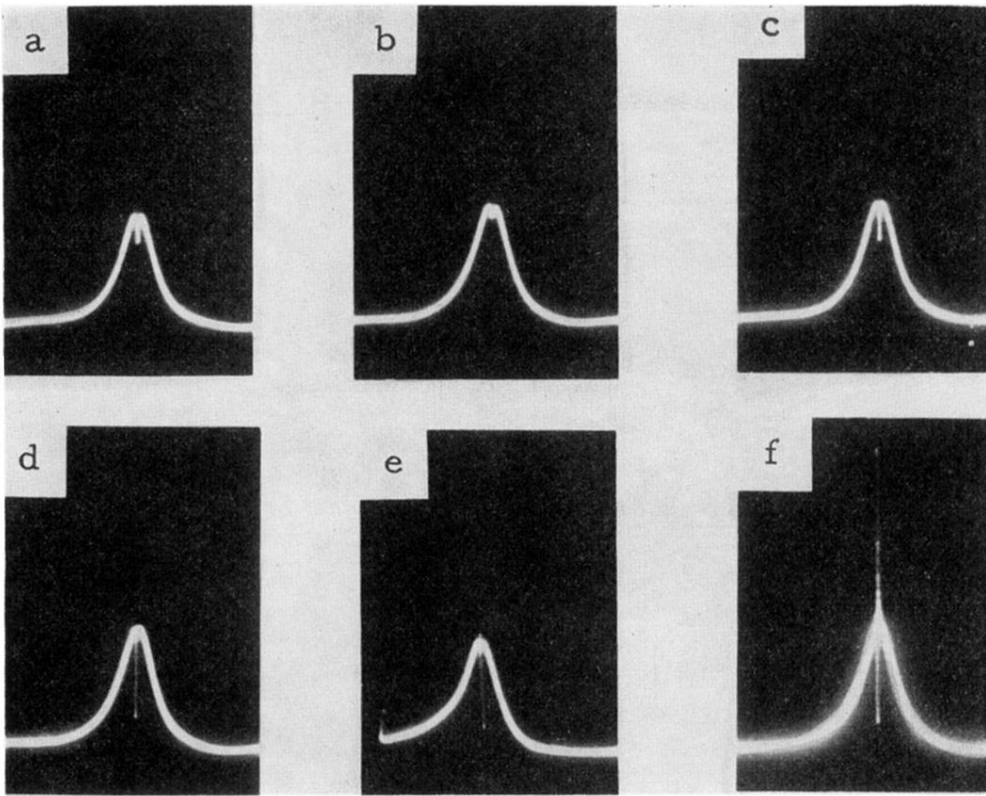


FIG. 9. Oscilloscope display of maser oscillator operation.