

Three major requisites for the operation of a conventional solid-state paramagnetic maser are a low-temperature environment, a source of pumping radiation, and a steady, homogeneous, magnetic field. For amplification restricted to a number of discrete ranges throughout the spectrum, however, a magnet is not required. Following some background material that explains these requisites, this paper discusses the feasibility of a maser without a magnetic field, using trivalent iron sapphire, and cites some advantages of this mode of operation.

ZERO-FIELD MASER

A. W. Nagy and G. E. Friedman

In order to follow the intricacies of the zero-field maser and to better understand the direction of the work done in this field at APL, a discussion of the principles and utility of the maser will first be presented. In any communications system the objective is to pass information between a transmitter and a distant receiver. If the system were perfect, a receiver could reconstruct the transmitted signal independently of the transmitter power by merely supplying enough amplification to the electromagnetic wave appearing at the receiver's antenna. In fact, however, this situation cannot be fully realized; electronic equipment and the medium between the transmitter and receiver have intrinsic, unavoidable signal-contaminating processes, which set a definite limit on how small a signal may become and still be reconstructed by the receiver. In general the transmitter, receiver, and the propagating medium add to the desired signal an undesired signal whose properties can most often be described only statistically. In addition there may be unwanted signals added to the desired signal from other transmitters operating with a favorable view of the receiver antenna. All of these additional signals can be lumped under the category of noise.

In designing a communications system careful consideration must be given to each type of noise in order to reduce interfering effects to a minimum. Unwanted signals from other transmitters may be eliminated by careful selection of the operating frequency and a judicious choice of modulation and demodulation schemes. The importance of background noise may be reduced by good antenna

design, and the ultimate in receiver noise performance may be obtained by incorporation of high-gain, low-noise amplifiers in the receiver immediately following the receiving antenna. It is in the latter area of receiver noise that the maser is of special significance.

In recent years electronics has made great strides in the development of low-noise R-F amplifiers, partly through the improvement of existing devices (for example, traveling wave tubes and klystron amplifiers) and partly through the development of completely new devices such as the parametric amplifier and the maser. Of all these devices, the maser comes closest to the achievement of a noiseless device, although some workers have shown recently that noise performance comparable to masers can be obtained using liquid-helium-cooled parametric amplifiers. The maser, however, is found in those roles in communications in which the very ultimate in system noise characteristics is necessary. The receivers involved in the space probes to the moon and to Venus, the receiver used in the Telstar satellite ground station at Andover, Maine, the receivers used in many radio astronomy installations, and the receivers used in the project West Ford experiment all employed masers of various designs as R-F preamplifiers.

To appreciate the level of noise encountered in a maser R-F preamplifier, some very simple noise-theory concepts and definitions will be found useful. Consider an amplifier with a resistive termination on the input at the standard temperature T_0 of 290°K and terminated on the output by some load. This load will have power dissipated in it due

to two sources, one being the power n_g transmitted through the amplifier due to the thermal noise generated by the input resistor, the second being the noise n_a due to the amplifier itself. The total power n_t in the load resistor is then $n_t = n_g + n_a$. The noise figure F is defined as the ratio n_t/n_g , i.e., the ratio of power in the load due to the input termination and amplifier to the power that would appear in the load if the amplifier were noiseless. Therefore, the goal in low-noise amplifier design is to achieve a noise figure as close to 1 as possible ($F = 1$ means $n_a = 0$; that is, the amplifier is noiseless). Typical noise figures for low noise microwave amplifiers are 2–10.

For narrow-band amplifiers the noise powers n_a and n_g can be related to some fixed amplifier characteristics such as gain and bandwidth, and to equivalent generators (resistors at an appropriate temperature) connected to the input. In this way the amplifier's noise figure F and the temperature T_a of an equivalent noise generator at the input may be related by $T_a = T_o (F-1)$. It is, therefore, completely equivalent to describe an amplifier by either its noise figure F or its noise temperature T_a , in which for either case the temperature of the source is taken as T_o . Therefore, an amplifier with a noise figure of 2 has an equivalent temperature T_o of 290°K. On the other hand, well designed, low-temperature maser amplifiers, exclusive of antenna or input waveguide noise, have noise temperatures less than 10°K, approaching the ideal case of absolute zero.

The Maser

Unlike conventional electron devices, which rely on the interaction between electron charge and an electromagnetic field, the maser utilizes the energy associated with the quantum states of the molecules, ions, or particles of a material. The underlying principles of the microwave maser, as well as its more recent development, the optical maser or laser, rest on some fundamental concepts of quantum theory. The reduction of these principles to a practical device was accomplished in greater part through the experimental procedures developed in electron paramagnetic resonance and microwave spectroscopy of the solid state. Thus, the maser has been cited as a classic example of how basic research may lead rapidly and unexpectedly to major advances in technology.

If a system of a large number of ions is examined to determine the different states of energy occurring in the system, quantum theory and experiment have shown that the energies fall into discrete levels. An ion in one energy level may go to either a higher or lower level (usually only the

next higher or next lower unless special conditions exist) by either gaining or losing exactly the energy difference between the two levels. This exchange of energy may take place via many mechanisms, two of importance being (1) thermal agitation and (2) interaction with an electromagnetic field whose frequency, f , is related to the energy difference by $\Delta E = hf$, where h is Planck's constant.

Thermal agitation is the source of noise in the maser since it can induce an ion in an upper energy state to fall to a lower one. During this process the ion gives up a quantum of energy at the frequency $\Delta E/h$, which may be radiated out of the material. As this radiation is completely uncorrelated with any incident signal field, it appears in an external circuit as noise. For solid-state maser operation the material must be immersed in a bath of extremely low temperature. The noise-producing thermal vibrations are thereby greatly reduced.

It is known, moreover, that the discrete energy levels associated with a quantum mechanical system are not equally populated, but rather the ions (if a system of ions is under discussion) are distributed among the levels in such a way that under ordinary conditions any lower level is more heavily populated than any level above it. The relative populations of any two levels are mathematically expressed by Boltzmann's law,

$$N_j = N_i e^{-[(E_j - E_i)/kT]},$$

where N_j is the number of ions in level j of energy E_j , k is Boltzmann's constant, and T is the absolute temperature.

Electromagnetic radiation at a frequency connecting two energy levels induces transitions of lower-level ions to the higher level and vice versa with equal probability. It may be noted that for solid-state masers, these energy states are those associated with the spin magnetic moment of the electrons of the ion. Each transition (this phenomenon is known as resonance) from a lower to an upper state removes one quantum of energy from the field. As a result of the larger number of ions in the lower state there will be more upward transitions than downward, leading to a net decrease in the energy of the incident radiation. Obviously, a process of this sort does not result in amplification. However, if through some mechanism the populations of the two levels could be reversed, i.e., if the upper energy level were made to have more ions than the lower, a net increase in the incident field would take place, since now there would be more downward transitions than upward. This process, which increases the strength of the electromagnetic field in synchronism with the exciting signal, results in coherent amplification.

Of the number of techniques that have been developed to achieve the necessary population inversion between a pair of energy levels, those that employ an additional level (or levels) are of the most general interest. Suppose, for example, that a system of ions placed in a solid material possesses the three energy states $E_1 < E_2 < E_3$, where each state statistically has $N_1 > N_2 > N_3$ ions, as graphically shown in Fig. 1A. Now, if very strong radiation (the pump energy) at the frequency corresponding to the energy difference $E_3 - E_1$ is coupled to the material, the populations of states 1 and 3 tend to equalize, while that of state 2 remains essentially the same; this results in the situation depicted in Fig. 1B. Thus, for the case illustrated, state 3 is occupied by more ions than state 2, enabling a weak signal of frequency $(E_3 - E_2)/h$ entering the material to be enhanced.

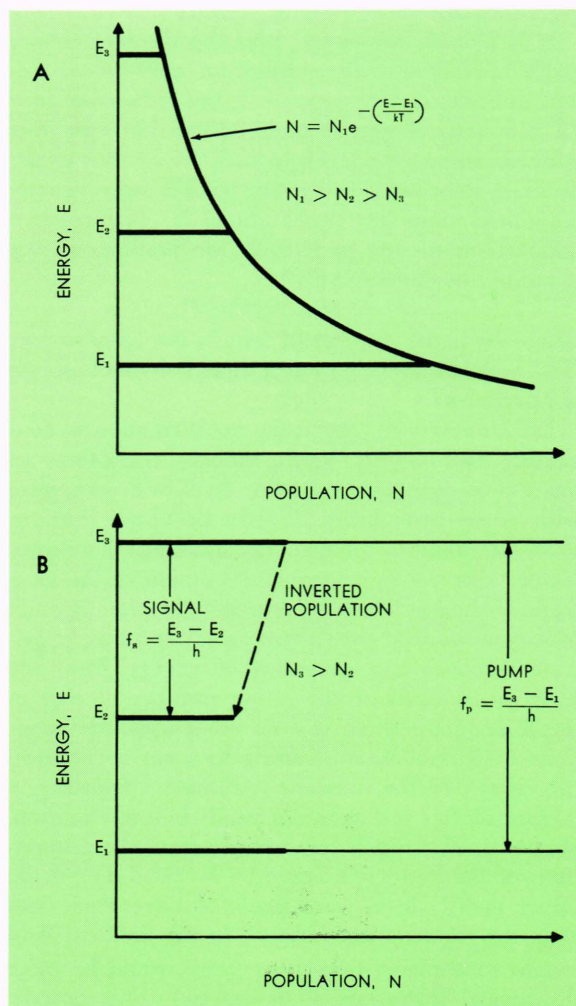


Fig. 1—Ion population in a paramagnetic sample, (A) in equilibrium (no pumping), and (B) with inversion between levels 2 and 3.

The central consideration in devising a maser is the establishment of a continuously inverted population between a pair of energy levels. Any physical process operating in the material, which tends to reduce this essential population difference or to return the ions to their normal-state distribution, also tends to destroy the amplifying ability of the material. Thermal vibrations induce transitions among the ions and are the primary phenomenon that counters the desired population inversion. A quantity called the relaxation time, which is a measure of the time required for the ions to return to their undisturbed distribution after the pump has been turned off, gives an excellent measure of the effect of the material's temperature on the ability of the pump to maintain the necessary population inversion. Data published for ruby,¹ which is the Al_2O_3 crystal with a Cr^{3+} ion impurity, show that the relaxation time is over 1000 times as long at 4°K as it is at 90°K . At the higher temperature the upper-state ions return to lower states as fast as the pump can elevate them, making continuous maser action nearly impossible. Most operational masers function in the region of 4.2°K , using liquid helium as the bath. Thus, the low temperature demand is simultaneously a blessing and a curse, being responsible both for the maser's phenomenally low noise behavior and for its most stringent operational requirement.

Although the detailed picture of the nature of the energy levels of the maser material can be seen only through the methods and mathematics of quantum mechanics, some very useful general notions about these levels can be given in rather gross terms. The maser substance itself is generally a single crystal in which a small number of the host ions have been replaced in the crystal structure by an appropriate foreign ion. The ubiquitous maser crystal, ruby, is formed by substituting chromium ions for roughly 0.05% of the aluminum ions in sapphire. Each Cr^{3+} ion has three electrons whose spin (the spin about the electron's axis) is not cancelled by that of other electrons. It is the interaction of these three electrons with the static fields present in the crystal, both electric and magnetic, that gives rise to the energy states that are the heart of the maser. The electric fields are due to the crystal itself, that is, they originate on the charges of the Al^{3+} and O^{2-} ions that surround the Cr ion. The physical spatial form of the field is crucial since on it rests, in part, the ability of the ion to make the essential direct transitions from energy level 1 to energy level 3.

¹ J. H. Pace, D. F. Sampson, and J. S. Thorp, "Spin-Lattice Relaxation Times in Ruby at 34.6 kMc/sec," *Phys. Rev. Letters*, 4, Jan. 1960, 18-19.

If a static magnetic field is added to the ever present electric field, the energy levels will react in a number of ways: energy levels may be split into additional levels; the energy difference between the levels may change; and the mixture of the ions of different spin angular momentum comprising a given energy level may be altered. All masers in operational installations require this external, static magnetic field. In the pages following, a description of a maser needing no magnetic field will be given.

The maser so far described is seen to possess at least two major requisites for a preamplifier, namely, it amplifies and it is essentially noiseless. The next consideration for a useful amplifier is its bandwidth. The primary goal and chief effort of most of the investigators seeking a practical maser for operational use has been to enhance its gain-bandwidth product, or more accurately to increase its bandwidth at a gain level sufficiently high to insure a negligible noise contribution from all the following stages of the receiver. The bandwidth of the maser is limited by the properties of the maser material itself as well as by the method of coupling the signal radiation into and out of the material. A band of frequencies centered about the frequency f_0 , corresponding to the energy difference $E_3 - E_2$ of Fig. 1A permeating the maser material, would be absorbed (no pump power applied) in varying percentages that decrease with deviation of frequency from that of f_0 . This frequency spread or intrinsic material linewidth (ruby's linewidth is about 60 mc/s) is essentially the limiting bandwidth of the system, and only masers employing traveling wave structures have been capable, therefore, of approaching full utilization of the material's resonance linewidth.

Unfortunately, traveling wave structures are extremely difficult to design and fabricate, and the long, high-quality single crystals necessary for optimum gain-bandwidth product cannot be grown easily. Further, very little progress has been made in scaling the successful C-band, traveling-wave, maser structures and techniques to higher frequencies that are playing an increasingly important role in modern technology.

An alternate method of coupling electromagnetic energy into the maser material employs a microwave cavity designed to resonate at the desired signal frequency when loaded with the active substance. Of course, cavity masers also suffer from ills uniquely their own. Since the cavity is generally more narrow banded than the resonance linewidth, the cavity itself becomes the bandwidth-limiting parameter of the maser. Most of the work done on cavity masers has been to circumvent this

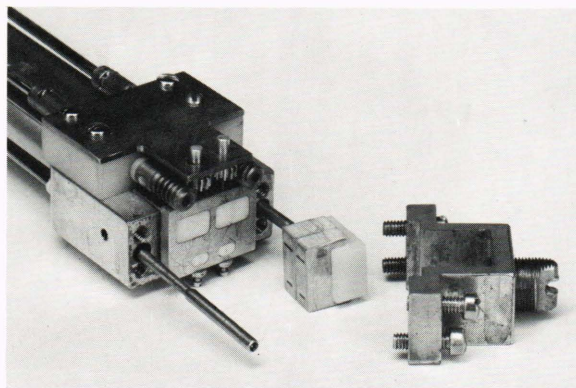


Fig. 2—Two-cavity ruby maser with silver-plated cavities (center) removed from coupling windows. Larger windows couple signal frequency to cavities; smaller windows on the bottom couple the pump frequency.

very problem by use of coupling structures outside the cavity and by use of multiple cavities.

At APL much effort² was spent on a scheme of this kind. A coupling window having a broad resonance was used to link two microwave cavities placed side by side at the end of the waveguide, as shown in Fig. 2. The cavities themselves were formed from solid blocks of ruby machined to the desired dimensions, while silver paint³ applied to the ruby blocks and fired at 600°C became the conducting walls of the cavities. Using an abrasive device, slots were etched in one of the hard silver surfaces to allow energy to enter the cavity. By properly selecting the frequency separation of the cavities, a broad composite response resembling that of a double-tuned amplifier was obtained. Although the geometry becomes very cumbersome, more cavities could be used to give even broader responses. The results of experimentation using the configuration of Fig. 2 are given in Fig. 3, where the line marked $G^{1/4} B = k$ is the theoretical result for a single ruby cavity, and where the line marked $G^{1/6} B = k$ gives the expected result for a pair of cavities.

A final, general consideration in the development of maser amplifiers is the problem of gain stability. If the gain of an amplifier were to vary in an unpredictable way with signal present at the input, the output signal would also fluctuate in a random manner, giving the appearance of noise added to the output. In a device such as the maser, with its nearly perfect noise characteristics, this noise-like phenomenon could become the leading factor in determining the overall noise perform-

² A. W. Nagy and G. E. Friedman, "Reflection Cavity Maser with Large Gain-Bandwidth," *Proc. Inst. Radio Engrs.*, **50**, Dec. 1962, 2504-2505.

³ L. G. Cross, "Silvered Ruby Maser Cavity," *J. Appl. Phys.*, **30**, Sept. 1959, p. 1459.

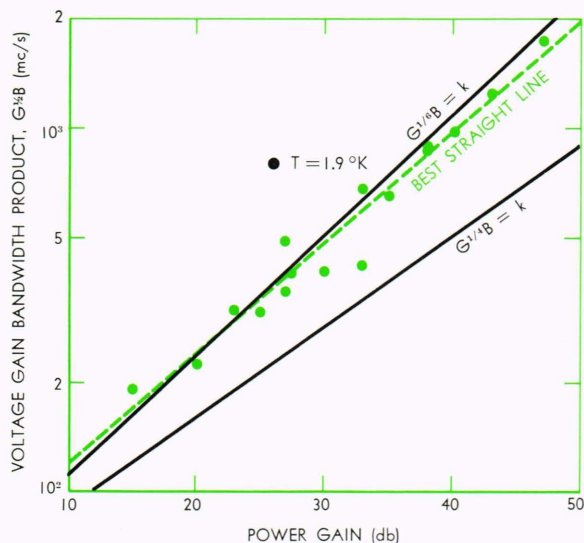


Fig. 3—Variation of gain-bandwidth product with gain for a two-cavity ruby maser at 4.2 °K. Small circles are experimental points. Note improved operation for the single point at 1.9 °K.

ance of a receiver. Investigators discovered very early that the single-cavity maser did not give adequate gain stability while the traveling-wave maser did, and this led to its adoption in many major systems. A recent theoretical report,⁴ though, has indicated that multiple cavities arranged in various ways can give stability surpassing that of a traveling-wave maser.

Details of the Solid-State Maser

In the proposal made eight years ago by Bloembergen⁵ for a three-level solid-state maser, a necessary requirement was that the energy level scheme associated with the paramagnetic ion-host crystal complex have non-zero magnetic dipole transition probabilities between levels not necessarily adjacent. This condition was central to the operation because it required pumping radiation of the proper frequency to equalize the spin populations of two nonadjacent levels, levels 1 and 3, Fig. 4. Transitions between these levels are normally forbidden by quantum mechanical selection rules since they represent a change in the magnetic quantum number of ± 2 . Transitions between consecutively spaced levels ($1 \leftrightarrow 2$, $2 \leftrightarrow 3$) involve a change of $\Delta M = \pm 1$ and are allowed. The rule governing ± 2 transitions, as well as

higher-order transitions, may be broken down if the paramagnetic ion has an energy-level splitting due to the internal crystalline electric field and if an external magnetic field is applied at an angle to the crystalline symmetry axis. For those combinations of magnetic field strength and crystal orientation where the level separations due to magnetic field alone (Zeeman splitting) are comparable to the separations induced by the crystalline field alone (Stark splitting), the electronic spin states are mixed and transitions between all levels are allowed. Since the ± 2 pumping transition is now allowed, a population inversion can be achieved between one of the pumped levels and a third intermediate level. Transitions between nonadjacent levels, which are allowed on the basis of this mechanism, may be considered as examples of noncubic crystalline field mixing. Most masers to date operate by the circumvention of the selection rules in this manner. It may be noted that two other state-mixing schemes have since been disclosed that allow the normally forbidden leap-frog transitions: (1) a rare earth ion in a cubic crystalline field,⁶ and (2) a large hyperfine interaction in a rare earth ion.⁷ The first of these is characterized by four levels degenerate in zero-field (no zero-field splitting) having a large linear Zeeman effect with very large g values. These materials may prove useful for millimeter-wave masers. The second method achieves the mixed quantization through a large magnetic nuclear-electronic interaction present in rare earth ions. Here the paramagnetic may be a powder, but the amplifying frequencies are limited to the order of several gigacycles (gc/s).

From electron paramagnetic resonance studies a number of materials are known that have, even in the absence of an external magnetic field, the

⁶ B. Bleaney, "A New Class of Materials for Bloembergen-Type Masers," *Proc. Phys. Soc.*, **73**, June 1959, 937-939.

⁷ E. S. Sabisky and H. R. Lewis, "Holmium Doped Calcium Fluoride as a Maser Material," *Proc. IEEE*, **51**, Jan. 1963, 53-56.

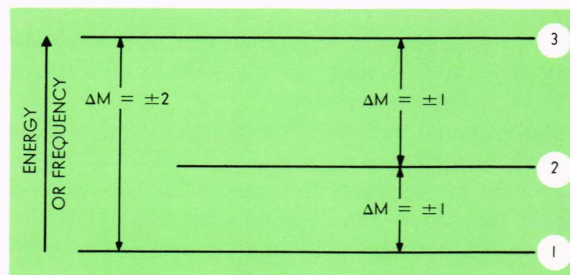


Fig. 4—Three-level maser. The ± 2 transitions represent the pumping levels, the ± 1 , the amplifier levels. Pumping radiation equalizes spin populations of levels 1 and 3 (saturation). Inversion (masing) may follow between levels $3 \rightarrow 2$ or $2 \rightarrow 1$.

⁴ G. Broussard and T. Molnar, "The Influence of Hyperfrequency Circuits on the Performance of a Coupled Cavity Maser," Fifth AGARD Avionics Panel Conference, Oslo, 1961, *Low Noise Electronics*, Pergamon Press, London, 1962, 200-224.

⁵ N. Bloembergen, "Proposal for a New Type Solid-State Maser," *Phys. Rev.*, **104**, Oct. 1956, 324-327.

necessary minimum of three unequally spaced energy levels with the required mixed quantization for cw maser operation.^{8,9,10} In these substances the crystalline field interaction alone splits and mixes the states adequately. Since no magnetic field is used, the pump and amplifying frequencies associated with this type of maser are limited to the values of crystal field splittings intrinsic to the material. The tuning range of the system would then be determined by the linewidths of the transitions involved. Despite this limitation that is not inherent in conventional magnetic field masers, there are a number of three-level materials with zero-field splitting frequencies spread throughout the microwave and millimeter wave ranges. Continued research in materials, moreover, is extending both the range and number of possible operating frequencies that may be used in the zero-field mode.

Requirements for Zero-Field Maser

Although the paramagnetism associated with the unpaired electrons in certain unfilled shells of an atom is a property of compounds of the five transition groups, iron, rare earth, palladium, platinum, and actinide, only the iron and rare-earth groups appear suitable for maser materials. The relevant unfilled electron shells in the iron and rare earths are the $3d$ and $4f$, respectively. To provide the necessary minimum of three energy levels in zero field, the spin of the ion, if integer, should be 1 or 2 since the number of levels is given by $2S + 1$, where S is the spin. For integer plus one-half spins, such as Fe^{3+} (spin $5/2$) and Gd^{3+} (spin $7/2$), the number of levels in zero-field is given by $S + 1/2$, i.e., three and four levels, respectively, for iron and gadolinium. Ions with spins $1/2$ or $3/2$, however, are not suitable for zero-field masers since there are less than three levels in zero field.

In host crystalline electric fields of high symmetry into which the ion may be placed, all the levels calculated by these rules may not be available. Sufficiently low symmetry in the crystalline potential must be present for the required splitting and mixing of the spin states. Figure 5 depicts this situation for the ferric ion as an S -state ion (no orbital angular momentum) having a six-fold degeneracy in zero magnetic field. The free ion electron configuration indicates that the spin magnetic moment is associated with the five uncompensated spins in the $3d$ shell. If the ion is in a crystal field

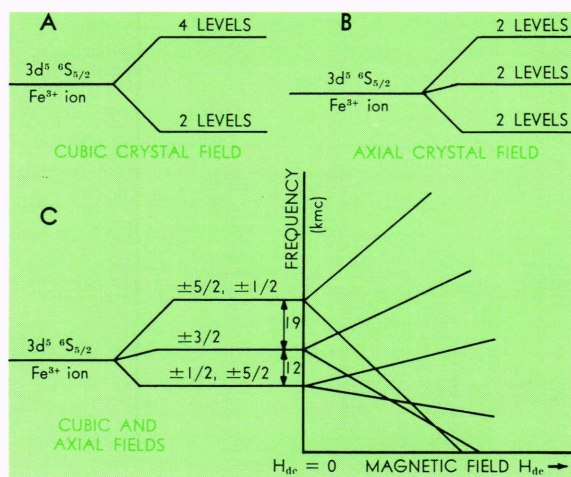


Fig. 5—Energy level scheme for iron sapphire. (A) Inadequate splitting with cubic field alone; (B) three levels (each doublet) but no mixing of states to allow pump transitions; and (C) adequate splitting and mixing of states to allow $\Delta M = \pm 2$ transitions even without a magnetic field ($H_{dc} = 0$). For H_{dc} parallel to the crystal C axis, levels with magnetic fields would appear as shown.

of cubic symmetry, the ground state is split into a low-energy doublet and a higher-energy quadruplet (Fig. 5A).

In a field of lower symmetry, i.e. an axial field, the four-fold degeneracy is partially lifted, resulting in three doublets in zero-magnetic field (Fig. 5B). Although three levels are now available, the spin states are not mixed in a way to allow the $\Delta M = \pm 2$ pump transitions. The necessary admixing is accomplished by the cubic crystal field whenever the spin is 2 or higher. Thus, trivalent iron substituting in Al^{3+} sites in the alpha-corundum (sapphire) lattice satisfies the basic requirement for zero-field operation: three adequately mixed energy levels without an external magnetic field. The two splittings in zero field are approximately 12 gc/s and 19 gc/s.

It may be interesting to note that in a magnetic field the position of the levels becomes a function of both the magnetic field strength and the orientation of the crystalline symmetry axis (C axis) relative to the magnetic field. The six levels shown in Fig. 5C are those for zero angle between H_{dc} and the C axis. As this angle increases, the levels separate with no crossovers. The magnetic field may be varied appropriately with the crystal orientation to provide maser and pump transitions comparable in frequency to the zero-field splittings. This is the normally operating Bloembergen region where all transitions generally are allowed. For high fields where the Zeeman energy is much larger than the zero-field splittings, the energy

⁸ G. E. Bogle and H. F. Symmons, "Zero-Field Masers," *Australian J. Phys.*, **12**, Mar. 1959, 1-20.

⁹ J. Wakabayashi, "Three-Level Maser Materials: A Survey of Potential Materials, I," Report No. 60, Electronics Res. Lab., Univ. of Calif., Berkeley, Mar. 7, 1962.

¹⁰ L. G. Van Uitert, "Solid State Maser Materials," *Metallurgy of Advanced Electronic Materials*, **19**, 1963, 305-327.

levels diverge and become linear with field. The levels then approach a pure spin-state condition with equal spacing between the levels. Transitions between adjacent levels are still allowed, but the pumping transitions are very weak or highly forbidden. From this it may be seen that the operating frequency range with a given material cannot be extended simply by utilizing the higher separation of levels generated by large Zeeman fields. For higher-frequency masers, materials with higher zero-field splittings must be used.

Trivalent iron has been incorporated into the rutile structure (TiO_2), tin oxide, zinc tungstate, and magnesium tungstate among many others. In rutile, which has been successfully operated as a magnetic-field maser and very recently as a zero-field maser,¹¹ the zero-field splittings are approximately 43 gc/s and 81 gc/s. In the tungstates the splittings are 61 gc/s and 77 gc/s. Thus, these materials are suitable for both zero-field and magnetic-field masers, well inside the upper millimeter-wave region. It may be worthwhile mentioning that a widely successful maser and laser material, ruby, is not suitable for zero-field operation. The spin of the chromium ion is $\frac{3}{2}$, and this gives only two levels, both doublets, in zero field, i.e. a single splitting around 11.5 gc/s. A magnetic field is required to split the doublets, giving four levels, all of which may be used for maser operation. The rare-earth ion gadolinium, Gd^{3+} with spin $\frac{7}{2}$, is another suitable zero-field ion. This paramagnetic was used in the first operating magnetic field maser. Of the eight low-lying levels of this ion in lanthanum ethyl sulfate, only three were used for a maser. Amplification at 9 gc/s was observed with 17 gc/s pumping. Gadolinium compounds, as well as those having the ions Ni^{2+} (spin 1) and Cr^{2+} (spin 2), appear also to be suitable for a zero-field maser.

Although there are other ion-crystal complexes that could qualify on the basis of energy-level structure and state-mixing as zero-field materials, other considerations may make them unsuitable for this mode of operation. The basic and practical criteria for a zero-field maser, most of which must also be satisfied for optimal magnetic field maser operation, may be summarized as follows:

1. In zero magnetic field there must be at least three low-lying energy levels having the desired amplification and pumping frequencies. The amplification frequency will be governed by the application, while the pumping transition should be sufficiently large to allow a high pump/signal frequency ratio. This will optimize the inversion and minimize noise.

2. The spin-lattice relaxation time (T_1) between the three participating levels should be long enough to yield observable spectra at room temperature. This generally indicates a T_1 of the order of milliseconds in liquid helium. A long T_1 will minimize the pump power required for saturation and decrease the magnetic Q of the maser transition, which results in improved gain-bandwidth product (GBP).

3. The optimum concentration of the paramagnetic should be used since this maximizes the GBP, minimizes the spin cross-relaxation and, hence, the linewidth. The pump power required for inversion is then minimal.

4. There should be preferably no nuclear spin since this contributes, through the hyperfine interaction, additional unused energy levels which reduce the inversion ratio in three-level operation by reducing the useful spin concentration. A large nuclear moment may at the same time reduce T_1 .

Although microwave spectroscopy has been performed on a large number of compounds that appear suitable for zero-field maser operation, many of the materials have not been examined at liquid helium temperatures, so that the zero-field splittings are known only approximately. The present purpose is served, therefore, by discussing only the results with iron sapphire. An earlier suggestion¹² and some experimental attempts elsewhere with iron sapphire in small magnetic fields (100-400 gauss) showed the possibility of using this material with no magnetic field. Published results of the GBP, however, were either impractically low in these small fields or not readily measurable even at 2°K.¹³

Exploratory runs at APL on a borrowed sample previously used by workers reporting a GBP of 14 mc/s in a field of 125 gauss,¹⁴ and other samples of iron sapphire obtained commercially, confirmed the low GBP and raised the question, subsequently, of adequacy in the Fe^{3+} concentration. Arrangements were then made with an outside source to undertake the boule growths through a modified flame-fusion process that would minimize the Fe^{3+} boil-off inherent in the conventional Verneuil technique used for incorporating this ion into the sapphire lattice. A number of small boules were grown and tested over an extended period before a workable maser with no magnetic field could be realized.

¹² K. D. Bowers and W. B. Mims, "Three Level Maser Without a Magnetic Field," Paper Presented at *Conference on Electronic Tube Research*, Berkeley, Calif., June 1957.

¹³ K. V. Karlov, Y. P. Pimenov and A. M. Prokhorov, "Paramagnetic Amplifier for the 10-cm Band Using Fe^{3+} Ions in Corundum," *Radio Engrs and Electronic Phys., USSR*, No. 5, May 1961, p. 755.

¹⁴ J. E. King and R. W. Terhune, "Operation of a Zero-Field Maser," *J. Appl. Phys.*, **30**, Nov. 1959, 1844-1845.

¹¹ W. E. Hughes and R. E. Deal, "Zero-Field Millimeter Maser," *Proc. IEEE (Correspondence)*, **52**, July 1964, p. 857.

Experimental Maser Facility

The basic working diagram of the zero-field facility is shown in Fig. 6. The signal and pump waveguides, together with the controls for adjusting the coupling and tuning to the maser cavity, are contained in the center section of the flask. This section is pre-cooled with liquid nitrogen, flushed, and then charged with liquid helium, while the surrounding outer jacket carries several liters of liquid nitrogen. Generally, a 6- to 8-hr run at 4.2°K may be made with two liters of liquid helium. When the space above the liquid helium is evacuated to lower the vapor pressure, temperatures around 1.5°K may be attained within an hour. The lower temperature decreases the spin-lattice relaxation time and generally improves the maser figure-of-merit. However, at the reduced temperature and under conditions requiring several hundred milliwatts of microwave pump power, the boil-off rate of the helium is considerably increased.

The microwave components are standard for the relevant frequency ranges; they operate in the signal (amplifier) branch around 12 gc/s, and for the pump transition, at about 31 gc/s. The Helmholtz coils provide a uniform variable field up to 300 gauss at the maser cavity. These fields are used for observing the magnetic resonance (absorption) of various maser samples and for studying the effects on the maser of very small magnetic fields, generally less than 25 gauss.

Since all the experimental work has centered around reflection-cavity configurations, the circulator shown is essential to the maser operation. Both the signal and pump energy are coupled into the tunable maser cavity through screw-tuned irises in a coupling structure similar to that used in the ruby maser. The input signal enters the maser cavity through the circulator, and the reflected amplified output is presented on an oscilloscope where, by standard procedures, the gain and bandwidth of the maser characteristic is determined. An oscilloscope monitors the klystron modes and displays the maser cavity modes resonant at the pumping frequency.

No-Field Maser Operation

Initially, the iron-sapphire boules supplied in the program were too small to allow design and cut of rectangular parallelepiped resonators as with the ruby cavities. Instead, the boule was trimmed, with minimum waste, to make the shape more regular for positioning on the coupling plate. In view of the complex mode character of a resonant cavity of this general form, no attempt was made to

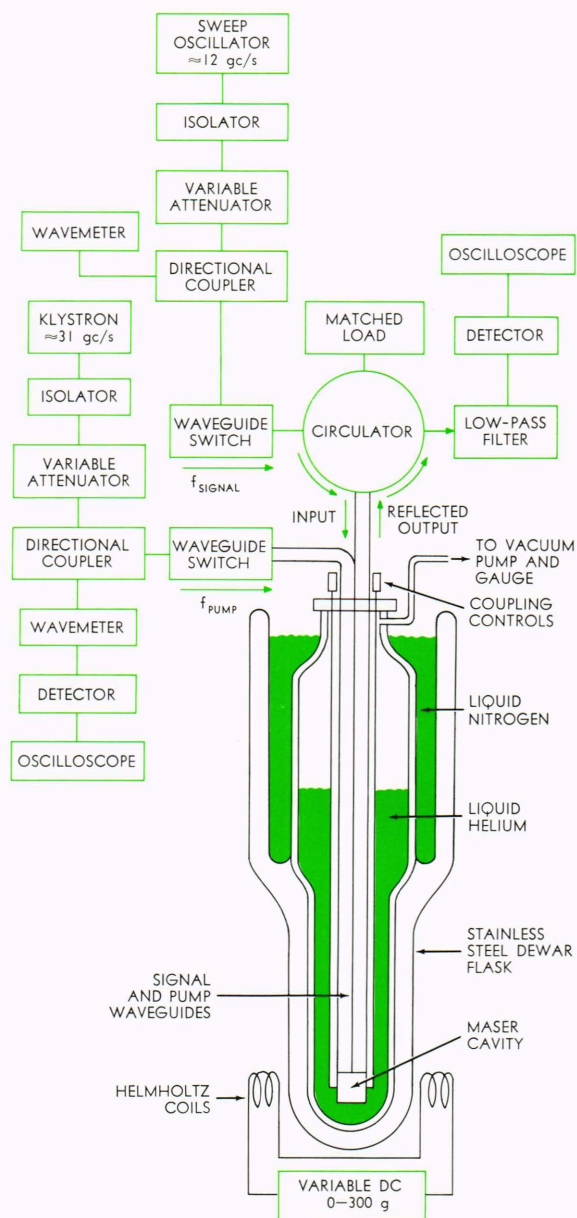


Fig. 6—Experimental schematic for a zero-field maser.

anticipate the mode response prior to the experimental room temperature tests. Figure 7 shows the physical arrangement of the zero-field maser cavity and tuning section. The boule is silver plated by a technique described in the literature,³ and one side is flattened and slotted to permit coupling to the dual iris structure attached to the signal and pump waveguides. The long dimension of the boule (≈ 0.75 in.) and the signal coupling slot are trimmed empirically to put a cavity resonance at the zero-field signal transition frequency. Allowance must be made for operation in liquid helium

since, on cooling, the cavity modes in this type of structure shift upward in frequency several tens of

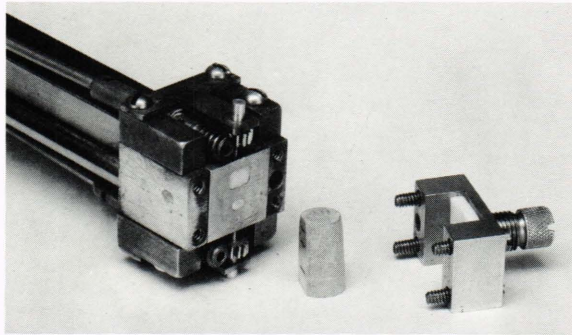


Fig. 7—Cylindrical iron sapphire cavity (center) removed from the coupling plate. First maser operation with no magnetic field was observed with this sample. Signal frequency ≈ 12 gc/s and pump frequency ≈ 31 gc/s.

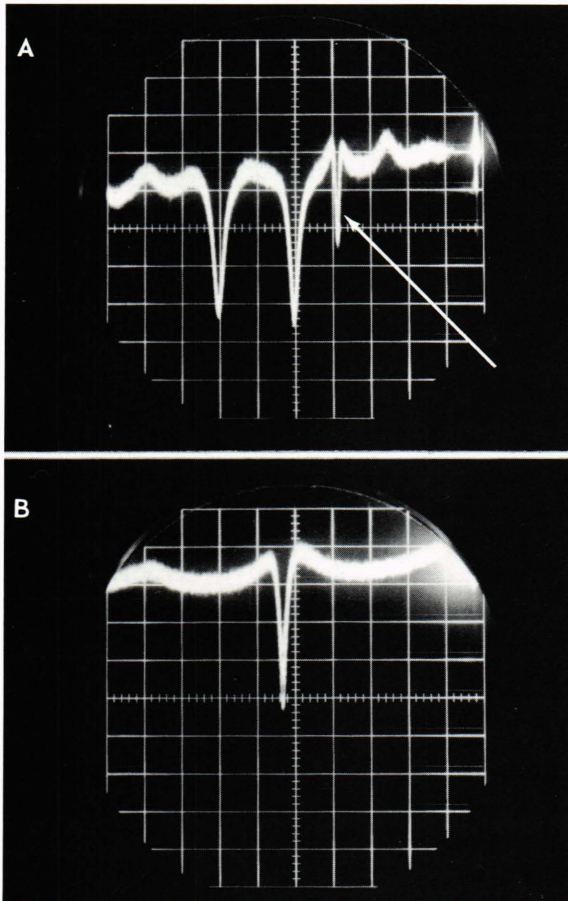


Fig. 8—(A) Three simultaneous inversions in iron sapphire with a magnetic field ≈ 25 gauss; the zero-field mode at right is indicated by the arrow. (B) The magnetic field is completely removed, only the zero-field mode persisting (shown expanded).

megacycles, owing mainly to the change in sapphire dielectric constant. A smaller contribution appears to come from the dimensional changes in the cavity. The shift in the ruby cavities, for example, was generally 60-80 mc/s at an operating frequency of about 9 gc/s.

The first good iron-sapphire cavity furnished in this program gave the results shown in Fig. 8: three simultaneous inversions in a field of 25 gauss at a C-axis orientation of approximately 18° . The smallest of the inversions (Fig. 8A) is at a frequency of 12.05 gc/s. When the magnetic field was completely removed, only this inversion persisted. Although the GBP of this maser was only a few megacycles, subsequent results with a boule cavity having an Fe^{3+} concentration of about 10^{19} spins/cm² and more nearly optimum coupling, gave a GBP over 200 mc/s. Figure 9 shows a typical double-hump response of a zero-field mode in absorption (9A) and masering with no magnetic field (9B). The expanded response (Fig. 9C) shows a maser with a GBP > 200 mc/s. These appear to be the first reported solid-state maser operations without the use of a magnetic field.¹⁵ It is interesting to note that an additional 25% improvement in GBP has been observed with fields as small as 5-10 gauss, with a C-axis orientation of 90° . The inversion, however, was not a sharp function of the angle.

Since the C-axis orientation of the crystal has no significance in a zero-field maser, a large monocrystalline structure is not required. Thus, the paramagnetic may be an aggregate of particles with random orientation, say, a powder. Figure 10 shows the inversions obtained with a cavity containing the particles of a crushed iron-sapphire boule that had been previously masered as a plated cavity with GBP > 100 mc/s. The particles were contained in a rectangular Teflon* box approximately 2.5 cm³ in volume, located inside a plunger-tuned rectangular waveguide cavity operating in a TE_{10n} mode, and coupled to the signal and pump guides through the plate used to couple the silvered cavities. Although this maser had only a 3-10 db gain, depending on the coupling, it showed a potentially large bandwidth (40 mc/s). No effort was made to optimize the maser circuit parameters in these particular runs since the main objective was to demonstrate the feasibility of maser operation in a powdered paramagnetic with no magnetic field.¹⁶

¹⁵ G. E. Friedman and A. W. Nagy, "Iron Sapphire Maser with No Magnetic Field," *Proc. IEEE*, 51, Feb. 1963, 361-362.

¹⁶ A. W. Nagy and G. E. Friedman, "A No-Field Powder Maser," *Proc. IEEE*, 51, July 1963, p. 1037.

* Trade name of polytetrafluoroethylene, manufactured by E. I. Du Pont de Nemours and Co., Inc.

Advantages of Zero-Field Operation

It seems appropriate to mention briefly some advantages that may be realized from masers operating without a magnetic field and using a powder paramagnetic.

1. In general, powders should be less difficult to prepare than defect-free monocrystalline boules with a well-defined C-axis. In the millimeter range, for example, tolerances in growing, cutting, orienting, and aligning single crystals required to fit into suitably close-tolerance traveling wave structures would pose a variety of difficult problems.

2. The use of a powder would allow a more flexible geometry in a traveling-wave or cavity-maser structure and could result in an optimal filling factor. Strength and mechanical stability in the active maser material need not be a prime requirement.

3. Many maser materials have equivalent but differently oriented paramagnetic ions in the unit crystal cell. Generally only one ion is properly oriented in a magnetic-field maser operation. In a powder all equivalent sites would be useful in producing electronic gain.

4. Elimination of a large, homogeneous magnetic field would simplify the maser package.

The disadvantage of having to operate the maser only around the frequencies set by the zero-field splitting intrinsic to the material, and not at any arbitrary frequency as with magnetic field masers, is partly compensated by the range and number of zero-field frequencies available. The possibilities of extending this technique to the relatively difficult millimeter-wave region appear attractive. The development of low-noise amplifier devices in that part of the spectrum through conventional traveling-wave magnetic-field maser and parametric techniques presents serious problems of structure, design, and operation.

Further Maser Development

The current effort in zero-field masers at the Laboratory centers around the preparation of suitably doped iron-sapphire powder for use in a traveling-wave X-band maser structure and further additional studies in multiple cavity operation. Since these powders have not been available commercially, a small facility was set up to produce experimental quantities via two processes. In the first a very small amount of ferric ammonium sulfate is combined in suitable proportion with aluminum ammonium sulfate. The mixture is calcined at 1000°C for three hours in a quartz beaker,

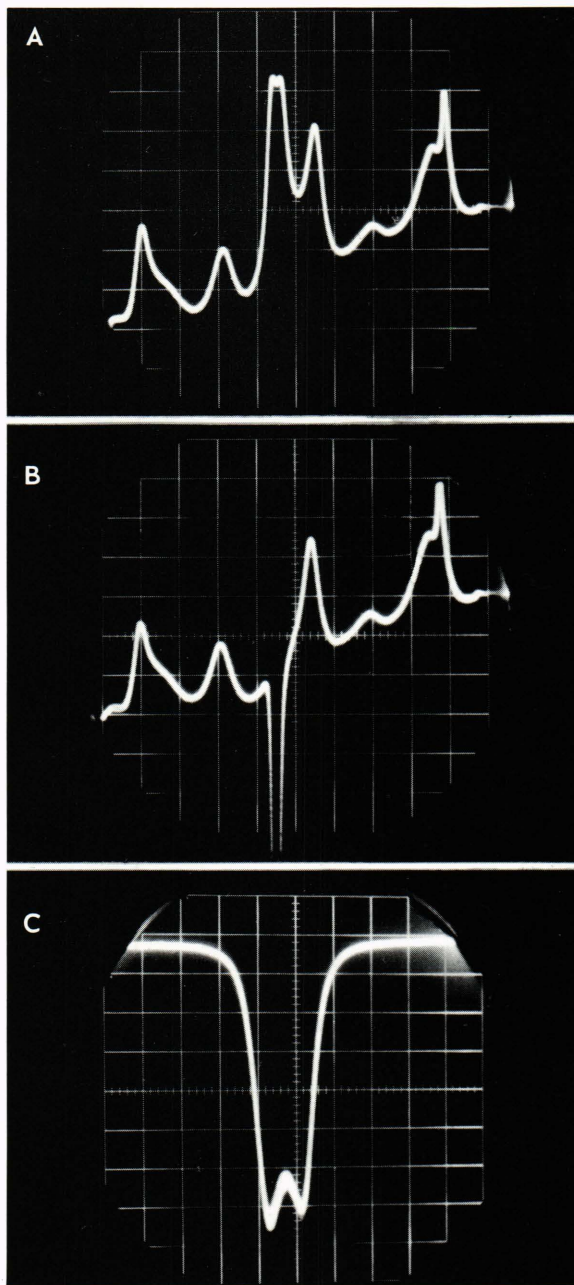


Fig. 9—Improved no-field iron-sapphire maser. (A) Iron sapphire double-hump zero-field line absorption; (B) masering with no magnetic field. Five other cavity modes range from 11.880 gc/s to 12.240 gc/s; (C) no-field maser \approx 12.04 gc/s; gain \approx 30 db; bandwidth \approx 5 mc/s; pump frequency \approx 31.03 gc/s.

which results in substitution of the Fe^{3+} ion for the aluminum in the sapphire lattice. A second calcining for 7 hr at 1300°C in a platinum crucible is believed to effect almost completely the necessary crystal structure conversion from $\gamma\text{-Al}_2\text{O}_3$ to

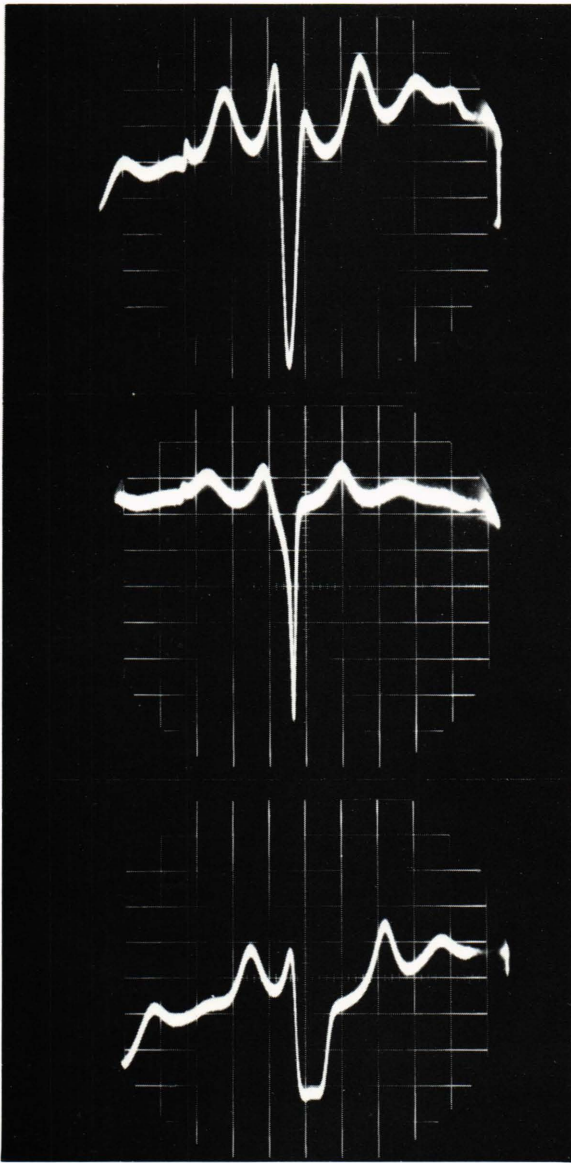


Fig. 10—Inversion in iron-sapphire powder with no magnetic field. Variation of gain with coupling $\approx 3 - 10$ db; bandwidth ≈ 44 mc/s per box; signal ≈ 12.03 gc/s; pump frequency ≈ 31.34 gc/s.

alpha- Al_2O_3 (alpha corundum). The sample is then prepared for a run in the Teflon cavity described above. In the second process a small amount of Fe_2O_3 is ball-mill ground with the desired amount of eta-alumina. Following a crushing to a 20 mesh, the powder is loosely packed into an alumina-lined crucible and fired to 1000°C overnight and at 1400°C for 3.5 hr. A final ball-mill grinding results in a fine powder.

Samples prepared in the preliminary phase of this program, mostly with the ammonium sulfates and with starting concentrations of iron ranging

from 0.5% to 0.003% have been tested. Although no masering was observed, almost all samples showed significant magnetic resonance and interaction with the pump, with a maximum occurring for the 0.01% sample. The interaction extends over a range from 11.90 gc/s to 12.10 gc/s, being maximal in the 12.03- to 12.06 gc/s region. Despite the lack of inversion there appeared little doubt that the interactions were assignable to an Fe^{3+} resonance.

In order to investigate a possible dependence of the inversion on the particle size, a number of isostatically pressed cylinders were formed from the powders and sintered to temperatures around 1500°C , the present maximum capability of the furnace. These cylinders are cut to the approximate shape of the original iron-sapphire boules and then silver plated and slotted. The density of the sample is only 40–50% that of the flame-grown boule. At the present writing, tests on all samples have not been completed. However, it is possible to report an inversion, for the first time, in a plated cavity of 0.01% material. The inversion, requiring several watts of pumping power, was transient; that is, the masering lasted momentarily as the pump frequency was tuned slowly through the zero-field resonance. More recently, with a cylindrical section of the same material in the Teflon cavity, continuous masering was observed with only about one-third the pump power. The masering could be moved over a region of approximately 60 mc/s by plunger-tuning the cavity, and it appeared to be a maximum at 12.03 gc/s with the pump at 31.29 gc/s. This is the first observation of a continuously operating maser in zero field, using a powder prepared as described earlier. In a similar configuration of a 0.02% sample prepared via Fe_2O_3 doping, a weak maser oscillation of an undercoupled mode at 12.04 gc/s was obtained. All these results indicate the feasibility of preparing an iron-doped corundum by both methods although the preferred method, at present, is the one using the sulfates.

The generally broad absorptions associated with the various samples may be due to excessive cavity losses arising from sample shape and particle size. This could account for the high pump power required for inversion. The concomitant heating could have shortened the relaxation time, which would have further degraded the inversion. Variations in powder preparation, such as sintering to a high temperature—around 2000°C —and varying the ferric concentration, will be explored. The objective here is the production of adequate amounts of powder with the maser properties of the crushed flame-grown boule.