Maser Operation at Signal Frequencies Higher than Pump Frequency*

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Summary—**Methods using harmonic spin coupling for operating solid-state masers with signal frequencies higher than the pump frequency are discussed. Expressions for the population inversion ratios are presented, and the maximum signal-to-pump-frequency ratios are calculated.**

Experimental data is presented on a ruby maser which is operated using the symmetrical method. Amplification was obtained at signal frequencies from 10,320 to 10,740 Mc, using pump frequencies ranging from 9580 to 9670 Mc.

An experiment in which maser operation is obtained simultaneously at two frequencies is described.

SOLID-STATE masers are ordinarily operated with the pump frequency two to ten times the signal frequency. This relationship—a relatively high pump frequency—permits good spin-population inversion (and hence satisfactory gain-bandwidth products) at the signal frequency.[1]

A major obstacle to the development of masers with very high signal frequencies has been the lack of availability of satisfactory pump power sources in the millimeter-wave and submillimeter-wave regions. One approach toward circumventing the unavailability of satisfactory millimeter pump sources has been the pulsed maser. With the pulse technique, transient maser amplification of signals up to 70 kMc has been obtained.[2]

Because transient methods have obvious disadvantages, there is a need for a continuous-wave maser in which the pump frequency is kept as low as possible relative to the signal frequency.

One method of keeping the pump frequency low is the method used in the "X-X maser." In this maser, the pump frequency is only about 15 per cent higher than the signal frequency.[3] This method of operation can probably be used in the millimeter-wave region by using maser materials which have suitably large zero-field splittings—for example, emerald, and Cr^{+++} or Fe^{+++} doped titania.

As the signal frequency is raised higher and higher, it becomes increasingly desirable to use a pump frequency lower than the signal frequency. Several methods for achieving continuous-wave maser operation with a signal frequency higher than the pump frequency will be discussed in this paper. Operating data on a maser operating in such a manner will be given.

The low-pump-frequency maser operation discussed here involves spin-coupling of transitions that are equal in frequency or harmonically related. In the harmonic-spin-coupling process, simultaneous spin flips cause energy transfer between transitions that are harmonically related in frequency; all, or almost all, of the Zeeman energy of the spin system is conserved.[4–8] Saturation of the lowest-frequency transition by applied pump power will result in a simultaneous saturation effect in the harmonically related transition. This spin-coupling mechanism will alter the spin population distribution between the energy levels in such a manner that a negative temperature is obtained at a signal frequency higher than the pump frequency.

The Symmetrical Method

A maser material is required having four (or more) energy levels. Let these energy levels be numbered from 1 to 4 in order of increasing energy, and let the notation f_{ij} represent the frequency corresponding to the energy difference between the ith and the jth levels. An operating point (in terms of magnetic field strength and orientation with respect to the crystal C-axes) is chosen at which $f_{14}=2f_{23}$. The method of operation is as follows.

The spin population distribution among the various levels before application of pump power is approximately as shown in Fig. 1(a). The f_{23} transition is saturated by means of pump power from an external pump source at frequency f_{23}. The saturation of the f_{23} transition will result in the simultaneous saturation (at least partial) of the f_{14} transition through the mechanism of harmonic spin coupling. Thus, the spin populations of

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1 N. Bloembergen, "Proposal for a new type solid-state maser," *Phys. Rev.*, vol. 109, pp. 324–327; October 15, 1956.

2 S. Foner, L. Momo, A. Mayer, and R. Myers, "Pulsed field millimeter wave maser," in "Quantum Electronics," Columbia University Press, New York, N. Y., p. 487; 1960.

3 F. R. Arams, "Low field X-band ruby maser," PROC. IRE, vol. 46, p. 1373; August, 1959.

4 N. Bloembergen, S. Shapiro, P. Pershan, and J. O. Artman, "Cross-relaxation in spin systems," *Phys. Rev.*, vol. 144, pp. 445–459; April 15, 1959.

5 W. H. Higa, "Excitation of an L-band ruby maser," in "Quantum Electronics," Columbia University Press, New York, N. Y., p. 298; 1960.

6 J. E. Geusic, "Harmonic spin coupling in ruby," *Phys. Rev.*, vol. 118, pp. 129–130; April 1, 1960.

7 W. S. C. Chang, "Spin lattice relaxation via harmonic coupling," in "Quantum Electronics," Columbia University Press, New York, N. Y., p. 346; 1960.

8 W. B. Mims and J. McGee, "Spin-spin energy transfer and the operation of three-level masers," PROC. IRE, vol. 47, p. 2120; December, 1959.

levels 2 and 3 have been equalized to the average value of their equilibrium populations. The same will be true of levels 1 and 4. The spin population distribution, assuming that all spin-lattice relaxation times are equal, is shown in Fig. 1(b). Note that a negative temperature is obtained in the f_{12} transition.[5,6]

The negative temperature at f_{12} and the saturated f_{23} transition combine to produce a negative temperature at a frequency f_{13}, which is higher than the pump frequency f_{23}. In terms of the spin populations of the various energy levels, since $n_2 > n_1$, and $n_2 = n_3$, it follows that $n_3 > n_1$, so that maser action can be obtained at f_{13}. Thus, a way has been found to obtain operation with a signal frequency higher than the pump frequency. Maser amplification can thus be obtained simultaneously at two signal frequencies, f_{12} and f_{13}, which are related by the expression $f_{\text{sig}_1} + f_{\text{pump}} = f_{\text{sig}_2}$. (This circumstance will be discussed further later on.)

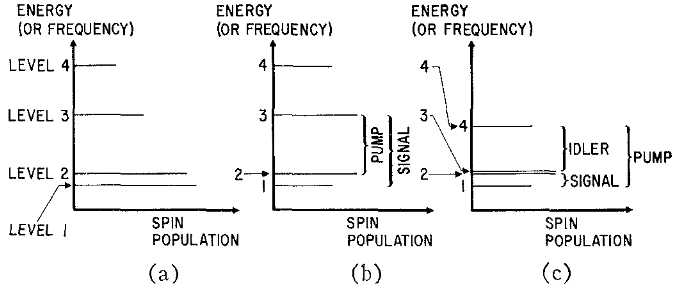


Fig. 1—Population distribution in symmetrical method. (a) Thermal equilibrium, pump power off. (b) Pump power (at f_{23}) on. (c) Analogous three-level technique illustrating that f_{34} is idler transition.

Comparison of this symmetrical method with the standard Bloembergen three-level maser[1] shows that f_{34} plays the role of the idler transition, and that Fig. 1(b) is analogous to the Bloembergen three-level maser technique if we think of the f_{23} transition as the degenerate intermediate energy level; this is shown in Fig. 1(c) and is verified by the following analysis.

Analysis:

We can solve for the steady-state (negative) population difference, $\Delta n_{13} = n_1 - n_3$, using the usual linearized rate equations[1] and the conditions $n_2 = n_3$ caused by pump saturation and $n_1 = n_4$ caused by saturation by second-harmonic spin coupling. A more general procedure is to add to each of the standard rate equations one term representing the spin coupling between the harmonically related transitions. (All other possible simultaneous multiple spin-flip processes are neglected.) Let w_c be the harmonic-spin-coupling transition probability; w_{ij}, the spin-lattice transition probability between the ith and jth energy levels; and n, the order of the spin-coupling harmonic. The condition $n_1 = n_4$ is not used, since the degree of saturation of the f_{14} transition will depend upon the parameter w_c/w_{ij}. For $w_c/w_{ij} = \infty$, the results are identical with those calculated for the first method. The two pertinent rate equations (since we are pumping between levels 2 and 3) are

$$\frac{dn_1}{dt} = -w_{12}\left[\Delta n_{12} - \frac{Nh}{4kT_0}f_{12}\right] - w_{13}\left[\Delta n_{13} - \frac{Nh}{4kT_0}f_{13}\right] - w_{14}\left[\Delta n_{14} - \frac{Nh}{4kT_0}f_{14}\right] - w_c[\Delta n_{14} - n\Delta n_{23}] \quad (1a)$$

$$\frac{dn_4}{dt} = w_{14}\left[\Delta n_{14} - \frac{Nh}{4kT_0}f_{14}\right] + w_{24}\left[\Delta n_{24} - \frac{Nh}{4kT_0}f_{24}\right] + w_{34}\left[\Delta n_{34} - \frac{Nh}{4kT_0}f_{34}\right] + w_c[\Delta n_{14} - n\Delta n_{23}]. \quad (1b)$$

In these equations N is the total spin population, h is Planck's constant, k is Boltzmann's constant, and T_0 is the bath temperature. The form of the harmonic spin coupling terms in (1) is obtained by linearizing (for second-harmonic) terms of the form[4] ($n_2^2 n_4 - n_3^2 n_1$.) For all w_{ij} equal, we obtain for the steady-state

$$\Delta n_{13} = -\frac{Nh}{4kT_0} \cdot \left[\frac{w_{ij}(f_{23} - 2f_{13}) + w_c\left[\left(\frac{n+1}{2}\right)f_{23} - f_{13}\right]}{2w_{ij} + w_c}\right]. \quad (2)$$

For $w_c/w_{ij} = \infty$, this reduces to

$$\Delta n_{13} = -\frac{Nh}{4kT_0}\left(\frac{f_{34} - f_{12}}{2}\right). \quad (3)$$

This expression is independent of n and has the same form as that for a standard Bloembergen three-level maser, which—for all w_{ij} equal—is

$$\Delta n_{\text{signal}} = -\frac{Nh}{4kT_0}\left(\frac{f_{\text{idler}} - f_{\text{signal}}}{2}\right). \quad (4)$$

Hence, it is evident that f_{34} and f_{12} act as the idler and signal transitions, respectively [Fig. 1(c)], and that we wish to maximize f_{34}.

Taking (2), normalizing it to the Boltzmann thermal equilibrium population difference $(\Delta n_{13})_0$, and rewriting it in terms of the signal frequency, f_{13}, and pump frequency, f_{23}, we obtain

$$\frac{\Delta n_{13}}{(\Delta n_{13})_0} \equiv \frac{T_0}{T_{s_{13}}} = 1 - \frac{\left(n + 1 + 2\frac{w_{ij}}{w_c}\right)\frac{f_{23}}{f_{13}}}{2\left(1 + 2\frac{w_{ij}}{w_c}\right)} \quad (5)$$

where $T_{s_{13}}$ is the spin temperature of the f_{13} transition. $T_0/T_{s_{13}}$ must be negative in order to obtain amplification, and the obtainable gain is proportional to $T_0/T_{s_{13}}$. As (5) shows, for $n > 1$, amplification can be obtained with a signal frequency f_{13} larger than the pump frequency f_{23}.

Using (5) we solve for the maximum ratio of signal-frequency-to-pump-frequency, with n and w_c/w_{ij} as parameters, by setting $T_0/T_{s_{13}}$ equal to zero. The results are shown in Table I. As can be seen, for $w_c/w_{ij}=\infty$, the maximum ratio of signal-frequency-to-pump-frequency is 1.5 for $n=2$, and 2.0 for $n=3$. The maximum signal frequency is seen to be dependent upon w_c/w_{ij}. This is intuitively reasonable, because the f_{14} (second-harmonic) transition is not saturated for finite w_c/w_{ij}. As expected, the maximum ratio increases with n. It is seen that w_c/w_{ij} need not be very high to permit maser amplification at frequencies higher than the pump frequency. However, because w_c is an implicit function of n, it may be difficult to achieve high signal-frequency-to-pump-frequency ratios. To maximize w_c/w_{ij}, it is desirable to go to as low a bath temperature as possible, since w_c is independent of and w_{ij} is proportional to bath temperature. This parameter—the ratio of the w's—can also be changed by varying the concentration of the paramagnetic ion in the maser crystal.

TABLE I

SYMMETRICAL METHOD: MAXIMUM RATIO OF SIGNAL FREQUENCY TO PUMP FREQUENCY

	$w_c/w_{ij}=\infty$	10	4	2	1
$n=2$	1.5	1.33	1.17	1.0	0.83
3	2.0	1.75	1.5	1.25	1.0
4	2.5	2.17	1.83	1.5	1.17
n	$\frac{n+1}{2}$	$\frac{n+1.2}{2.4}$	$\frac{n+1.5}{3}$	$\frac{n+2}{4}$	$\frac{n+3}{6}$

Eq. (5) also shows that the inversion improves as the signal-frequency-to-pump-frequency ratio is decreased. In fact, it continues to increase even when the signal frequency becomes lower than the pump frequency. When the signal frequency is less than the pump frequency, the energy-level arrangement could be called harmonic push-pull pumping (Fig. 2). Thus, even when the signal is less than the pump frequency, the improved inversion (and hence increased gain-bandwidth product) makes the harmonic-coupling technique attractive in comparison with the standard three-level Bloembergen maser technique.

ALTERNATIVE SYMMETRICAL METHOD

Suppose the energy levels are arranged in such a manner that $f_{14}=2f_{23}$ (as previously), but $f_{34}<f_{12}$ (rather than $f_{34}>f_{12}$, as previously). It is still possible to obtain maser amplification at a signal frequency (now f_{24}) higher than the pump frequency (f_{23}). Here, f_{12} is the idler transition. Thus, just as in the standard Bloembergen three-level maser, maser operation can be obtained with the idler transition higher in energy than the signal transition, or vice versa.

THE ASYMMETRICAL METHOD

In this method, we again require four (or more) energy levels numbered from 1 to 4 in order of increasing

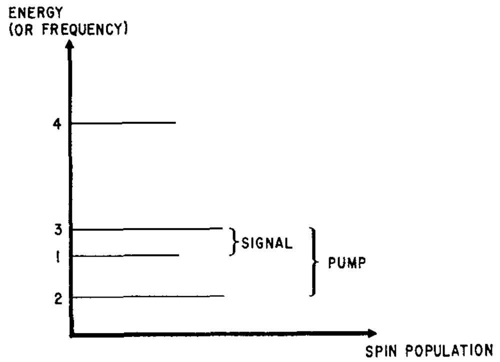
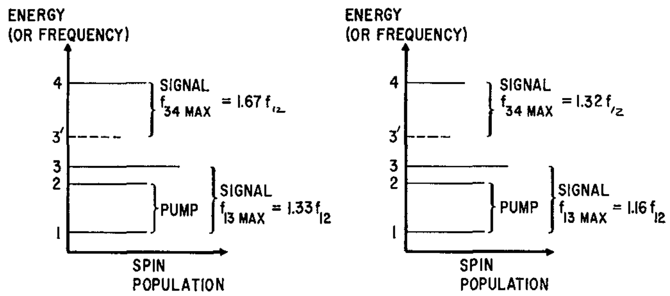


Fig. 2—Population distribution for harmonic push-pull pumping.



(a) (b)

Fig. 3—Population distribution for asymmetrical method. (a) Complete saturation of f_{24} transition. (b) Incomplete saturation of f_{24} transition $w_c/w_{ij}=10$; $n=z$.

energy. We chose an operating point at which $f_{24}=2f_{12}$. The method of operation is as follows (see Fig. 3).

The f_{12} transition is saturated by means of pump power from an external pump source at frequency f_{12}. The harmonic spin-coupling mechanism results in the simultaneous saturation (at least partial) of the f_{24} transition. The populations of levels 1, 2, and 4 will be essentially equalized to their average value. The spin population distribution will then be as shown in Fig. 3(a).

A negative temperature can be obtained at a signal frequency f_{13} (and f_{23}) or f_{34}, depending upon the location of energy level 3. The signal frequency f_{13} will be, and the signal frequency f_{34} may be, higher than the pump frequency f_{12}.

Analysis:

We can solve for the steady-state (negative) population differences, Δn_{13} and Δn_{34}, using the usual linearized rate equations and the saturation condition $n_1=n_2=n_4$.

Again, the more general procedure is to add the harmonic spin-coupling terms to the rate equations. In the steady state, assuming that all w_{ij} are equal, we obtain for the population inversion ratios,

$$\frac{T_0}{T_{s_{13}}}=1-\frac{\left(n+2+4\frac{w_{ij}}{w_c}\right)\frac{f_{12}}{f_{13}}}{3+8\frac{w_{ij}}{w_c}} \qquad (6)$$

and

TABLE II
ASYMMETRICAL METHOD: MAXIMUM RATIO OF SIGNAL FREQUENCY TO PUMP FREQUENCY

	$w_c/w_{ij}=\infty$		10		4		2	
	f_{13}	f_{34}	f_{13}	f_{34}	f_{13}	f_{34}	f_{13}	f_{34}
$n=2$	1.33	1.67	1.16	1.32	1.0	1.0	0.86	0.71
3	1.67	2.33	1.42	1.84	1.2	1.4	1.0	1.0
4	2.0	3.0	1.74	2.37	1.4	1.8	1.14	1.29
n	$\frac{n+2}{3}$	$\frac{2n+1}{3}$	$\frac{n+2.4}{3.8}$	$\frac{2n+1}{3.8}$	$\frac{n+3}{5}$	$\frac{2n+1}{5}$	$\frac{n+4}{7}$	$\frac{2n+1}{7}$

$$\frac{T_0}{T_{s_{34}}} = 1 - \frac{(2n+1)\dfrac{f_{12}}{f_{34}}}{3 + 8\dfrac{w_{ij}}{w_c}}. \tag{7}$$

The maximum ratios of signal-frequency-to-pump-frequency as functions of n and w_c/w_{ij} for the asymmetrical method are shown in Table II. The maximum signal-to-pump ratio is different for the f_{13} and f_{34} transitions, being 1.33 and 1.67, respectively, for second-harmonic spin coupling, and 1.67 and 2.33, respectively, for third-harmonic spin coupling. Eqs. (6) and (7) also show that, as in the symmetrical method, harmonic spin coupling significantly improves maser operation over the three-level Bloembergen maser, even when the signal frequency is lower than the pump frequency.

The population ratio of the pump transition f_{14} was calculated assuming that the f_{12} transition is saturated by externally applied pump power. We obtain

$$\frac{T_0}{T_{s_{14}}} = \frac{8 - \dfrac{4}{n+1}}{8 + 3\dfrac{w_c}{w_{ij}}} \tag{8}$$

for the dependence of the degree of saturation of the f_{14} transition upon the w_c/w_{ij} parameter. As expected, $T_0/T_{s_{14}} \to 0$ for $w_c/w_{ij} \to \infty$. How incomplete saturation of the spin-coupled second-harmonic transition reduces the maximum signal frequency is shown in Fig. 3(b). The reduction in population of level 4 combines with an increase of the level 1 and 2 populations to yield only a limited frequency range where inverted populations are obtainable.

THE PUSH-PUSH MASER

This may be considered as a special case of the asymmetrical method with $n=1$. Since $f_{12}=f_{24}$, both transitions are saturated directly by the applied pump power.[9] The population inversion ratios can be obtained directly from (6) and (7) by setting $w_c/w_{ij}=\infty$ and $n=1$. The result shows that the push-push maser produces a significant improvement in population inversion ratio over the three-level Bloembergen maser. However, maser operation at a signal frequency higher than the pump frequency can be obtained for unequal spin-lattice relaxation times only.

EXPERIMENTAL DATA

Continuous-wave maser operation with the signal frequency about 1000 Mc higher than the pump frequency was successfully achieved using the symmetrical method. By using the four energy levels in ruby (chromium-doped aluminum oxide), maser amplification was first obtained at $f_{13}=10{,}590$ Mc, using a pump frequency of $f_{23}=9595$ Mc. The magnetic field of 1675 oersteds was oriented at 90 degrees to the ruby C-axis. The energy levels used are indicated in Fig. 4; they were so arranged that $f_{14}=2f_{23}$.

We established experimentally that the f_{14} transition is saturated because of the pump power applied at frequency f_{23}, and that the frequency ratio need not be exactly integral.[10] It was possible, using a constant pump frequency of $f_{23}=9650$ Mc, to saturate the f_{14} transition over a 600-Mc frequency range (centered near 19,300 Mc) with an optimized magnetic field.

The experimental result that the second-harmonic, f_{14}, transition can be saturated over a range of frequencies with constant pump frequency, is attributable in part to the fact that the frequency of the f_{23} transition varies very little with magnetic field (Fig. 4), changing by less than 40 Mc per 100 oersteds in the region of interest.

The experimental setup consisted of a doubly resonant tunable cavity operating in the TE_{10} waveguide mode. The ruby crystal had a nominal 0.05-per cent residual chromium content. The pump power was 8 milliwatts at a liquid helium bath temperature of 4.2 degrees K. To ascertain that the saturation of the f_{14} transition was primarily caused by harmonic spin coupling, we measured the second-harmonic power output of the pump power source in the 18 to 26 kMc range and found it to be less than 1 microwatt.

A voltage-gain bandwidth product of 11 Mc was measured at a liquid helium bath temperature of 4.2 degrees K. It is expected that larger gain-bandwidth products can be obtained, since the ruby occupied only a small portion of the cavity. The filling factor is estimated to be 50 per cent. The spin population inversion ratio was also measured, and was found to be -0.37.

[9] "Solid-State Research," MIT Lincoln Lab. Quarterly Progress Rept., p. 65; July 15, 1959.

[10] F. R. Arams, "Maser operation with signal frequency higher than pump frequency," PROC. IRE, vol. 47, p. 108; January, 1960.

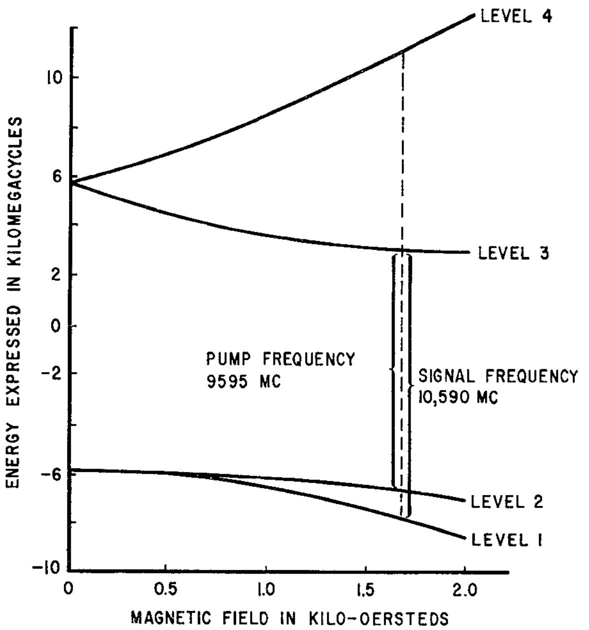


Fig. 4—Energy levels used in ruby in symmetrical method experiment.

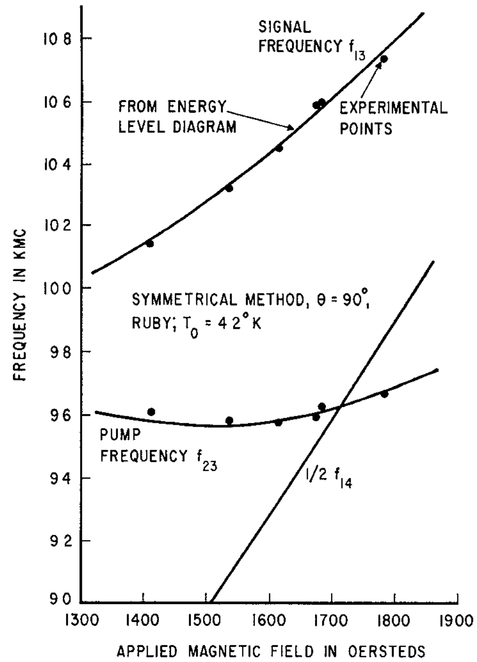


Fig. 5—Frequency tuning range of ruby maser using symmetrical method.

The tuning range was 420 Mc with optimized pump frequency and magnetic field and a liquid helium bath temperature of 4.2 degrees K. Maser amplification was obtained over the 10,320- to 10,740-Mc range, using pump frequencies ranging from 9580 to 9670 Mc. The data are plotted in Fig. 5. The tuning range obtained is also of interest in view of the frequency difference between the pump frequency f_{23} and $\frac{1}{2}f_{14}$, but it is consistent with the observed wide-range harmonic saturation of f_{14} previously described. A greater tuning range should be obtainable at 1.5 degrees K.

An attempt was also made at obtaining maser operation using the asymetrical method. The orientation of 60° between ruby C-axis and applied magnetic field is of particular interest, since the harmonic relationship $f_{24}=f_{12}$ is maintained over a wide range of frequencies,[8] that is, a pump frequency $f_{12}=8.3$ to 12.7 kMc, and a signal frequency $f_{34}=10$ to 15 kMc. Q improvement was observed, but inversion has not yet been obtained, indicating the possibility that additional factors (such as other spin-flip processes) may need to be included that were not considered in the present analysis.

Two-Frequency Maser Operation

In the symmetrical method, a negative spin temperature was obtained at two frequencies simultaneously. One of these is at a relatively low frequency, designated f_{s1}. The second frequency f_{s2} is numerically related to f_{s1} via the pump frequency f_p—that is, $f_{s2}=f_{s1}+f_p$.

A triply resonant cavity experiment was set up, and it was possible to obtain maser amplification at f_{s1} and f_{s2} simultaneously. Operating conditions were: magnetic field = 1650 gauss at 90 degrees to the ruby C-axis; $f_p = 9600$ Mc; $f_{s1}=1005$ Mc; and $f_{s2}=10{,}605$ Mc. When the liquid helium bath temperature was reduced to 1.5 degrees K, maser action was obtained at f_{s1} and f_{s2}, simultaneously. Maser action would get better or worse at both frequencies concurrently as adjustments—such as angular orientation and magnetic field strength—were being made.

The condition $f_{s2}=f_p+f_{s1}$ used here is also used in the parametric amplifier up-converter. In that application, p-n junction diodes or ferromagnetic materials are used, and the input is at frequency f_{s1}, the output is at frequency f_{s2}, and the obtainable power gain is equal to the frequency ratio f_{s2}/f_{s1}.

In the experimental double-frequency maser setup, no interaction between the f_{s1} and f_{s2} signals was observable except when the input power level approached saturation. An input signal at f_{s1} did not affect the f_{s2} output, and vice versa. It is concluded that the absence of coherence between the f_{12} and f_{13} transitions prevents the realization of a maser amplifier up-converter.

Conclusions

Several methods for obtaining CW maser operation with a signal frequency higher than the pump frequency were analyzed. Performance data on the symmetrical method were presented. This maser was tuned over a 420-Mc range. Using the methods described, millimeter-wave masers having pump frequencies lower than signal frequencies should become available in the foreseeable future.

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