Conclusions

A multiple-beam, deflection modulation system appears to be a rather simple method of producing a high-current, high-harmonic content beam with a large cross-sectional area. Unfortunately, the coupling structures suggested to be used with these beams have rather low interaction resistance so that the efficiency is poor. However, the quasi-optical approach using systems large compared to the wavelength is quite attractive. If better couplers could be devised, large amounts of pulsed power would be possible.

Acknowledgment

The authors wish to thank Prof. M. D. Sirkis for his advice and helpful discussions. The suggestions Dr. O. T. Purls (Watkins-Johnson Company) on the electron gun used were of great help. The technical assistance of J. F. Lowe and morale-building humor of A. B. Wilson are fondly remembered.

Multiple Quantum Effects at Millimeter Wavelengths*

R. H. PANTELL†, MEMBER, IEEE, AND R. G. SMITH, MEMBER, IEEE‡

Summary—**When a quantum mechanical system interacts with a radiation field it may do so by multiple as well as single quantum processes. These multiple quantum processes give rise to nonlinear effects such as harmonic generation and parametric amplification and oscillation. The density matrix formulation is used to describe these multiple quantum processes. Two- and three-level systems are considered as forms of harmonic generators and some of the desired properties of the materials to be used are described. Two methods of generating submillimeter radiation starting with optical signals are also discussed.**

I. Introduction

SINCE the first work on the maser by Gordon, Zeiger and Townes [1] much interest has been focused on the possible uses of the quantum properties of matter in the field of electronics. Various types of gaseous and solid-state amplifiers and oscillators have been successfully operated in both the microwave and optical regions, and operation in the millimeter and submillimeter range seems to be possible. The basis for operation of all of these systems is the maser principle, which states simply that if the normal population distribution of an allowed transition is inverted, then there will be a net emission rather than a net absorption by the quantum system. This net emission makes possible amplification and oscillation. These processes are all essentially single quantum processes.

In addition to these familiar applications of the interaction of radiation with a quantized system, there are processes in which more than a single quantum of radiation is involved in the interaction. Such processes are called multiple quantum interactions. These processes allow various types of frequency mixing effects similar in nature to those in classically describable nonlinear elements. For this reason these quantum mechanical processes may also be called nonlinear. They are generally strong field effects and may be used for various applications such as parametric amplification and oscillation, harmonic generation, and modulation. The requirements of the quantum system for these nonlinear effects differ from those of maser applications, especially in the fact that population inversion is not necessary for most of the cases considered so far. These new requirements may make possible the use of many new materials not particularly suited for maser applications.

The millimeter and shorter wavelength range appears particularly well suited for such applications, as many atoms, molecules and crystals have strong spectra in this region.

The phenomenon of multiple quantum effects will be described in more detail and an approach which is well suited to the solution of such higher order quantum mechanical processes will be presented. This will be followed by a discussion of several possible applications.

II. An Approach to the Problem of Multiple Quantum Transitions

For the types of problems dealt with here, the perturbation theory rate equation method does not appear as useful as some other approach which more exactly solves the equations of motion. A method of attack which is particularly well suited for such problems is the density matrix approach. A detailed discussion of the density matrix is given in [2]–[4] and its application to radiation problems in [4]–[8]. It is essentially

* Received January 21, 1963; revised manuscript received June 13, 1963. The research reported in this paper was supported jointly by the U. S. Army Signal Corps, the U. S. Air Force, and the U. S. Navy (Office of Naval Research) under Contract Nonr. 225(48). The paper was presented at the millimeter and Submillimeter Wave Conference, Orlando, Fla., January, 1963.

† Standard Telecommunication Laboratories, Harlow, Essex, England. On leave from Stanford University, Stanford, Calif.

‡ Bell Telephone Laboratories, Murray Hill, N. J. Formerly with Microwave Laboratory, W. H. Hansen Laboratories of Physics, Stanford University, Stanford, Calif.

a method for taking into account both the quantum mechanical and statistical properties of matter. For our use here the density matrix ρ may be described by its equation of motion

$$i\hbar \frac{\partial}{\partial t} \rho = [\mathcal{H}, \rho], \tag{1}$$

and by the prescription for obtaining the expectation value of an observable

$$\langle Q \rangle = Tr(\rho Q). \tag{2}$$

In (1) $\mathcal{H}$ is the total Hamiltonian operator composed of the unperturbed term $\mathcal{H}_0$ and a perturbation term $\mathcal{H}'$ which is due to the interaction of the system with radiation fields; ρ is the density matrix operator; $[\mathcal{H}, \rho]$ is the usual commutator defined as $[\mathcal{H}, \rho] = \mathcal{H}\rho - \rho\mathcal{H}$. Eq. (1) folows from Schrödinger's equation. Eq. (2) defines the expectation value of an observable Q as the trace of the product of the matrices of the density operator ρ and the operator corresponding to the observable. These matrices may be taken with respect to any complete set of functions but it will be found expedient to take as the basic functions the eigenfunctions of $\mathcal{H}_0$. In matrix form (1) becomes

$$i\hbar \frac{\partial}{\partial t} \rho_{nm} = [\mathcal{H}, \rho]_{nm}. \tag{3}$$

Finally, relaxation times may be phenomenologically added to (1) or (3) to account for various relaxation processes such as spin-spin and spin-lattice interactions.

The diagonal components ρ_{nn}, when normalized such that $\sum \rho_{nn} = 1$, may be interpreted as the probability of occupancy of the nth state. When considering a macroscopic system which contains N microsystems, the quantity $N\rho_{nn}$ is then the average number of systems in state n. The off-diagonal terms, ρ_{nm}, $m \neq n$, are a measure of the extent to which levels n and m are connected, this coupling being caused by the external radiation fields. If the quantity ρ_{nm} is small, then the coupling of levels n and m is small; if it is large, then they are strongly coupled. We shall now qualitatively examine these terms in order to see how multiple quantum processes may come about.

Consider now two levels of the system, l and k. From (3) the equation of motion for ρ_{lk} is given by

$$i\hbar \frac{\partial}{\partial t} \rho_{lk} = \sum_j (\mathcal{H}_{lj}\rho_{jk} - \rho_{lj}\mathcal{H}_{jk}), \tag{4}$$

where we have expanded the commutator and the matrix products appearing in (3). By separating the Hamiltonian into the unperturbed term $\mathcal{H}_0$ and the term due to the interaction with the fields $\mathcal{H}'$, we have

$$i\hbar \frac{\partial}{\partial t} \rho_{lk} + (E_k - E_l)\rho_{lk} = \sum_j (\mathcal{H}_{lj}'\rho_{jk} - \rho_{lj}\mathcal{H}_{jk}'),$$

where now the terms on the right hand side contain only the perturbation Hamiltonian. We may further separate the terms on the right-hand side to give

$$i\hbar \frac{\partial}{\partial t} \rho_{lk} + (E_k - E_l)\rho_{lk} = \mathcal{H}_{lk}'(\rho_{kk} - \rho_{ll}) + \rho_{lk}(\mathcal{H}_{ll}' - \mathcal{H}_{kk}') + \sum_{j \neq l,k} (\mathcal{H}_{lj}'\rho_{jk} - \rho_{lj}\mathcal{H}_{jk}'). \tag{5}$$

From the form of the left-hand side of (5) we see that the "natural" frequency of ρ_{lk} is $\Omega_{lk} = (E_k - E_l)/\hbar$, and hence terms on the right-hand side near this frequency will be most effective in driving ρ_{lk}. We may now qualitatively visualize the various forms of this coupling.

The first term on the right-hand side of (5), $\mathcal{H}'_{lk}(\rho_{kk} - \rho_{ll})$, describes the direct coupling of levels l and k due to a perturbation $\mathcal{H}_{lk}'$. Such a perturbation will arise when there is a matrix element between the two states (such as an electric or magnetic dipole moment) and an applied radiation field whose frequency ω is near Ω_{lk}. This direct coupling gives rise to the usual linear effects involving only a single quantum. Inclusion of terms to this order is sufficient to describe the usual maser theory.

The second term involves a form of self coupling whereby some state l is coupled to itself (*i.e.*, $\mathcal{H}_{ll}' \neq 0$). Such a coupling will arise, for example, when a level possesses a permanent moment. Although this term may be important in many cases of interest, we shall not consider it further; rather, we shall turn to the final terms which describe the indirect coupling processes giving rise to the nonlinear effects described here.

Consider the summation

$$\sum_{j \neq k,l} (\mathcal{H}'_{lj}\rho_{jk} - \rho_{lj}\mathcal{H}_{jk}')$$

which is a summation over all the rest of the states of the system of terms of the form $\mathcal{H}'_{lj}\rho_{jk}$. A term of this form states that if level k is somehow coupled to j, $\rho_{jk} \neq 0$, and there exists a perturbation coupling the levels l and j, $\mathcal{H}_{lj}' \neq 0$, then levels l and k are indirectly coupled via level j. If ρ_{jk} is coupled in first order, then from (5) it is seen that ρ_{jk} is proportional to $\mathcal{H}_{jk}'$. Therefore, ρ_{lk} is proportional to $\mathcal{H}_{lj}'\mathcal{H}_{jk}$ and will have a frequency dependence corresponding to the algebraic sum of the frequencies of $\mathcal{H}_{lj}'$ and $\mathcal{H}_{jk}'$. This is an example of a second order coupling. In principle, any order of coupling may arise. For such a coupling to be possible it is necessary that there exist allowed transitions between the various levels such that a progression between the levels may be formed, *i.e.*, $l \rightarrow j$, $j \rightarrow j'$, $j' \rightarrow j''$, $\cdots j^n \rightarrow k$.

As an example, let us consider a four-level system in which transitions are allowed between the following pairs of levels, (1-4), (1-3), (2-3), (2-4); these are shown in Fig. 1 along with the defined frequencies. Directly coupled in first order are those states connected by arrows. In second order we have 4 coupled to 3 via 1, 4 to 3 via 2, 2 to 1 via 3, 2 to 1 via 4. In third order 4 is coupled to 2 via 1 and 3, 4 to 1 via 2 and 3, 3 to 2 via 1 and 4, etc. These processes are easily seen to give rise to frequency mixing effects. For example, suppose we apply three fields at frequencies $\omega \approx \Omega_{41}$, $\omega_2 \approx \Omega_{42}$, $\omega_3 \approx \Omega_{32}$. These will produce first order coupling between the corresponding states. From the preceding discussion we find that levels 1 and 3 are coupled, *i.e.*, $\rho_{13} \neq 0$, the frequency of the driving term being $\omega_1 - \omega_2 + \omega_3$ and the magnitude proportional to the product of the fields at the three frequencies. We may go further if we now calculate the expectation value of the operator giving rise to the matrix element μ_{13} from (2) we find a term $\mu_{13}\rho_{31} + \mu_{31}\rho_{13}$ which will vary at the frequency $\omega_4 = \omega_1 - \omega_2 + \omega_3$. This term (magnetic or electric dipole) will act as a source for electromagnetic fields at the frequency ω_4. Thus in this example our four-level quantum system has acted as a nonlinear element, combining fields at the three frequencies ω_1, ω_2, ω_3 to generate a field at ω_4 defined above, the magnitude of this field proportional to the product of the magnitudes of the applied fields. Although this example is probably impractical it does serve to demonstrate the principles involved.

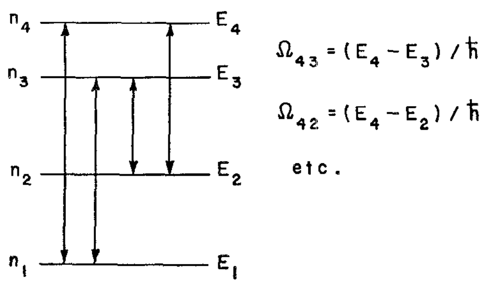

Fig. 1—Four-level system with allowed transitions shown by arrows.

A procedure which has been employed for the solution of such problems is as follows. With a particular idea in mind, such as harmonic generation, and the energy level scheme to be employed (*i.e.*, the number of levels, energy spacings, allowed transitions) the density matrix equations of motion are set up under the assumed conditions. A steady-state solution is found by assuming a harmonic series for the various components of the density matrix, and employing harmonic balance. The set of algebraic equations which results gives the desired steady-state solution to the quantum mechanical problem. By using (2), all the observables of interest may be calculated; of particular interest are those observables which act as the source of radiation fields (electric and magnetic dipole moments). The frequency dependence of these quantities will be that of the applied frequencies and combination thereof. Finally, a classical electromagnetic field theory problem may be solved where the effects of these observables are included as source terms.

Rather than go into more detail, some of the more general results of such an analysis will be included here. This will be followed by a discussion of some applications which might be useful in the millimeter wave range.

Some aspects of multiple quantum processes are as follows:

1) For a strong first order interaction the frequency of the radiation field should be near the natural frequency of the transition involved.

2) For indirect coupling, the frequency of the effective driving term is the algebraic sum of the individual contributions to the coupling. In order to be effective this frequency should be near the natural frequency of the driven transition.

3) The strength of the over-all interaction is proportional to the product of the strengths of the individual interactions. These are in turn proportional to $\mathcal{H}'/\hbar\Delta\omega$ where $\mathcal{H}'$ is the perturbation energy connecting the levels and $\Delta\omega$ is the linewidth of the transition when the applied frequency ω equals the natural frequency Ω, and is equal to the difference $(\omega - \Omega)$ when the applied fields are off resonance. In general the strength of the over-all interaction will be less the higher order the coupling, and will be larger the closer energy is conserved in each step.

4) Energy of the radiation fields and quantum system must be conserved in the over-all process. This may occur either when each individually conserves energy, such as in harmonic generation, or when the increase in field energy equals the decrease in the energy of the quantum system or vice versa. In the latter case, since the quantum system has discrete energies, the frequencies of the radiation fields must satisfy the condition $\sum \pm \omega_i = (E_f - E_i)/\hbar$, where $+\omega_i$ implies absorption of a photon at ω_i, a $-\omega_i$ implies emission, and $E_f - E_i$ is the increase in the energy of the quantum system.

5) The question of population inversion vs no population inversion depends on the particular case. If the radiation field conserves energy and hence the quantum system conserves energy such as in harmonic generations, then the nonlinear effect is independent of the sign of the population difference. If the energy of the quantum system increases (accompanied by an equal decrease in the energy of the field) corresponding to a jump of the quantum system from E_i to E_f, $E_f > E_i$, then levels f and i must be in a normal population state. If the quantum system gives up energy, then these levels must be inverted. For single quantum transitions the above is simply the maser principle.

6) Under the influence of the strong radiation fields required for these higher order effects, the effective natural frequencies of the transitions are changed depending on the magnitude of the RF fields present [9]–[11] Such an effect is particularly critical when the transitions involved have narrow linewidths.

We shall now turn to a discussion of some representative examples of these effects.

III. Applications

One of the most useful applications of a nonlinear element is its use in generating power at frequencies where fundamental sources are not available. We shall discuss several aspects of this problem; namely, harmonic generation in both two and three level systems, and methods of obtaining millimeter wavelengths starting with optical signals. Other possible applications are also mentioned.

A. Harmonic Generation

1) Two-Level System:

a. Principles: The two-level system is the simplest of all quantum mechanical systems, yet it does offer some interesting possibilities for harmonic generation. Let us consider a scheme of operation. Suppose there is a radiation field of frequency ω applied to a two-level system. We shall assume that the levels 1 and 2 are connected by a dipole moment of magnitude μ. In first order, level 1 is coupled to 2; in second order, level 1 is connected to itself via 2; in third order levels 1 and 2 are again coupled. This coupling will be at the frequency 3ω and its magnitude will be proportional to the cube of the applied field, E^3 (or H^3). As a result of this coupling term at 3ω and by using (2) there will be a component of the polarization (or magnetization) at the frequency 3ω. This will act as a source for third harmonic fields.

Thus far, no mention has been made of the relation between the frequency of the applied radiation ω and the natural transition frequency Ω. From an examination of the process it is seen that energy is conserved by the field in the over-all process but it can be conserved in only one of the three intermediate steps, either for $\omega \approx \Omega$ or $3\omega \approx \Omega$. These then are the two possible frequency ranges of operation where we may expect a fairly strong interaction. This will clearly not be as strong an effect as a situation where energy is conserved or nearly conserved at each step.

For an electric dipole system of the above type, the magnitude of this polarization $P(3\omega)$ has been evaluated in [10] and [11], and is given by

$$\underset{\omega \approx \Omega/3}{P(3\omega)} = \frac{\mu^2 c\gamma S E_1^3}{8\pi\Omega^3\hbar^2[1 + T_2^2(\Omega - 3\omega)^2]} \tag{6a}$$

$$\underset{\omega \approx \Omega}{P(3\omega)} = \frac{\mu^2 c\gamma S E_1^3}{64\pi\Omega^3\hbar^2[1 + T_2^2(\Omega - \omega)^2]}, \tag{6b}$$

where μ is the value of the dipole moment for the 1–2 transition; c is the speed of light; γ is the absorption coefficient for the transition as observed in single quantum spectroscopic experiments; E_1 is the magnitude of the fundamental electric field; Ω is the natural transition frequency; $\hbar$ is Planck's constant/2π; and $T_2 = 1/\pi\Delta\nu$, where $\Delta\nu$ is the full linewidth of the transition at half intensity. The quantity S is a saturation parameter and is defined as the ratio of the population difference $(n_1 - n_2)$ under dynamic conditions to its equilibrium value $(n_1^e - n_2^e)$,

$$S = \frac{n_1 - n_2}{n_1^e - n_2^e}. \tag{7}$$

From an examination of (6a) and (6b) it is seen that the harmonic polarization is expressed entirely in terms of known quantities; no phenomenological nonlinearity need be added. The details of this calculation may be found in [10] and [11].

The power generated at 3ω by this harmonic dipole will be proportional to the square of the polarization. It is thus proportional to $\gamma^2 S^2$, which brings up two interesting points. First, since the power is proportional to γ^2 it is independent of the sign of γ and hence independent of whether or not the populations of the states are inverted. This result is in contrast to the population inversion requirement for the maser oscillator. An oscillator or amplifier requires a negative conductance, which is achieved by producing population inversion. A harmonic generator requires a nonlinear conductance or reactance, and this nonlinear element may present a positive conductance at the output frequency. Hence there is not the requirement for population inversion. Secondly, saturation, which occurs when the populations of the two levels tend to equalize and S tends to zero, will limit the output power of such a system. It is found then that the effects of saturation are to be minimized as much as possible, especially when high powers are involved. In the case of a solid this implies a fast spin-lattice relaxation and, in a gas, operation at higher pressures resulting in broader lines.

For practical considerations any system to be used in this manner should have both a large γ and a large μ, the power out being proportional to $\mu^4\gamma^2$ (or to μ^8 since $\gamma \propto \mu^2$). Since electric dipole moments are in general two orders of magnitude larger than magnetic dipoles, the former will be far superior. Many gases show very strong lines in the millimeter range; these appear, at this time, to be the most practical operating substance. In fact, for some types of gas molecules the absorption coefficient varies as Ω^3, making the harmonic polarization (6) independent of frequency. This suggests the extension of these principles to as high a frequency range as the necessary fundamental field strengths are available. Typically these fields need to be of the order of 15,000 volts/cm or greater.

We shall now briefly consider a possible form for such a device and the experimental evidence for such operation.

b. Applications of Two-level System: Because of the strong fields required in order to obtain significant harmonic output power it appears necessary to use a resonant structure at the fundamental. In order to couple out the maximum power the structure should be resonant at the harmonic also. Akitt, Strain and Coleman [12] have considered a Fabry-Perot type of system which appears well suited for this application because of the high Q's involved. Using such a structure the gas may either completely fill the volume or perhaps selectively fill various sections of the cavity by the use of partitions. Since it is not necessary to have an inverted population for operation, no state separator or focuser is required as in beam type masers, simplifying construction.

For microwave transitions in a gas the absorption coefficient for a given line is independent of pressure. From (6) the polarization is then independent of pressure and hence the power available is also independent of pressure. The effects of saturation are, however, dependent on the linewidth of the transition and hence on the pressure. Higher pressures result in more frequent collisions and hence a faster return to equilibrium. This fast relaxation will result in a lessening of the effects of saturation. Hence we may increase the pressure and reduce saturation without altering the strength of the nonlinearity. This provides for a more versatile operation than could be obtained by using a solid.

Practically, such a harmonic generator would consist of the doubly resonant system filled with the gas with provision for coupling the power in and out. In order to eliminate the possibility of heating of the gas it might be continuously cycled.

Some qualitative aspects of third harmonic generation using a two-level system have been verified experimentally [13] using the inversion transition in ammonia. Harmonic powers on the order of 30 milliwatts were obtained at a frequency of 25.5 kMc using a pumping source at 8.5 kMc in a re-entrant type of microwave cavity. Saturation was found to vary as expected and maximum output power was obtained at a pressure of 300 mm Hg. These results are shown in Fig. 2. Work is now in progress to obtain more quantitative data. Akitt, *et al.* [12] at Illinois are using a Fabry-Perot type of system in conjunction with the strong HCN transition at 88.6 kMc and are attempting to generate both the third and fifth harmonic. This work is described elsewhere [12].

2) *Three-level System:*

Second harmonic generation in a three-level system provides an example of a multiple quantum process in which energy is nearly conserved at all stages of the interaction. Consider a three-level system where the energy levels are approximately equally spaced and where transitions are allowed between all pairs of levels. In this case, the outer levels (1 and 3) are individually connected to the middle level (2) as well as to each other. The natural transition frequencies are shown in Fig. 3. If a radiation field of frequency $\omega \approx \Omega_{31}/2$ is now applied to the system it will excite both the 1–2 and 2–3 transitions strongly, being nearly resonant in both cases. As a result of these first order couplings, levels 1 and 3 will be coupled in second order and a component of the dipole moment will be generated at 2ω if there is a nonzero matrix element between these levels. This component of the dipole moment will then act as a source of radiation fields as explained previously. The strength of this dipole moment will be proportional to the square of the applied radiation field and to the product of the dipole moments at the three transitions. In addition, it is proportional to the difference in populations between levels 1 and 3, $(n_1 - n_3)$. By denoting the field by F_1, the dipole moments by the appropriate subscripts and the resultant dipole by $M(2\omega)$, we have

$$M(2\omega) \propto F_1^2 \mu_{12}\mu_{23}\mu_{13}(n_1 - n_3).$$

The details of the calculation and the exact expressions may be found in [11].

The power generated, being proportional to $M^2(2\omega)$, will then be proportional to $\mu_{12}^2\mu_{23}^2\mu_{13}^2(n_1 - n_3)^2$. From this it is clear that large dipole moments are extremely desirable and that population inversion is not necessary. Further, since the power generated depends on the square of the difference in populations of the two levels, the effects of saturation are to be avoided. This suggests, as in the two-level case, the use of materials with fast relaxation times.

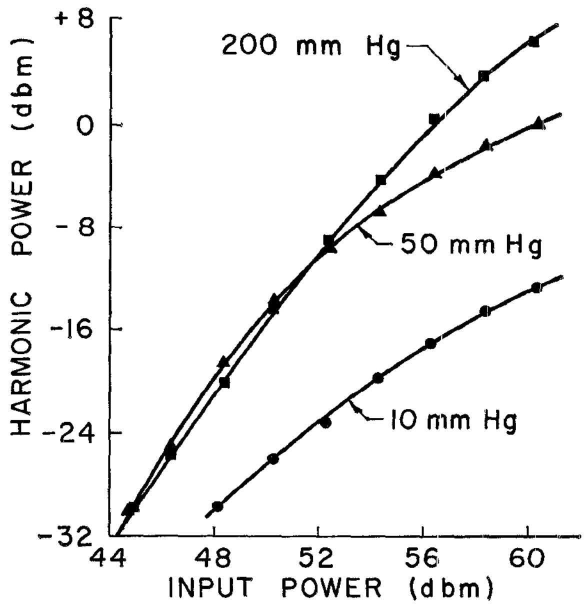

Fig. 2—Third harmonic output power vs input power for various pressure levels. The operating substance is NH_3.

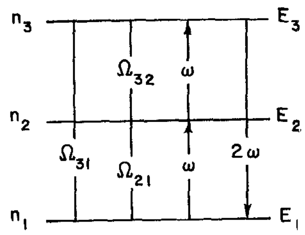

Fig. 3—Energy level diagram for second harmonic generation in a three-level system.

Examples of systems which have the desired characteristics of equally spaced energy levels and the proper selection rules include some paramagnetic materials. The exact behavior of a given ion will, of course, depend on its environment, in particular the strength and symmetry of the crystal fields.

Although no particular materials will be suggested here, some general desired properties of the material may be listed:

1) The zero field splitting should be large and of the order of the working frequency.
2) The crystal symmetry should be such that $\Delta m=2$ transitions are allowed, corresponding to the 1–3 transition.
3) Since the harmonic power varies as the product of the squares of the matrix elements of the three transitions involved, a large effective spin is desired.
4) Since it is undesirable to saturate the levels involved, a short spin-lattice relaxation time is desirable and is required for high power operation.

The latter condition 4) should make possible the consideration of a larger class of materials than for maser applications where saturation of the pump transition is required.

Kellington [14] has experimentally verified the principles involved using a degenerate system, *i.e.*, $\Omega_{32}=\Omega_{21}$. The experiment was performed at room temperature using pink ruby as the substance at a fundamental frequency of 9.425 kMc. Although the output power, 10^{-9} watts, was low, operation was still in the square law region with efficiency increasing proportional to the input power. Had the experiment been performed at liquid helium temperatures, a considerable increase in the efficiency at low power levels should have been expected, due to the increase of the quantity (n_3-n_1). Similar results were obtained using a nondegenerate system where $\Omega_{32}\neq\Omega_{21}$ [11]. Although experimental results to date are encouraging, there is still insufficient data available to make any statement as to the ultimate practicability of such a device.

B. Optical Mixing

With the development of high power optical masers there exists the possibility of mixing two optical frequencies to obtain their difference frequency. This beat frequency may lie anywhere in the range from microwave to infrared. Javan [15] has proposed a scheme for mixing two optical signals in a three-level system which uses the concepts discussed in this paper. This method may be described by considering the energy level diagram shown in Fig. 4.

The transition frequencies Ω_{31} and Ω_{21} correspond to optical frequencies while Ω_{32} is near the desired output frequency. All three possible transitions must be allowed. If two optical signals of frequencies ω_1 and ω_2 are applied where $\omega_1\cong\Omega_{31}$, $\omega_2\cong\Omega_{21}$, then levels 2 and 3 will be coupled in second order, the strength of the coupling being proportional to the product of the fields at the two optical frequencies. This coupling will produce a component of the dipole moment at the difference or beat frequency. As an example Javan considers the R_1 and R_2 lines in ruby. His calculation shows that when light near both these two frequencies (derived from a ruby laser) is incident upon a ruby crystal, the power generated at the difference frequency is at least some five orders of magnitude greater than that expected from mixing in KDP. Were the transition between the R_1 qnd R_2 lines electric dipole rather than magnetic dipole an additional increase of several orders of magnitude would result in the output power. By varying the temperatures of the rubies used and hence their natural frequencies, a variable difference frequency can be achieved.

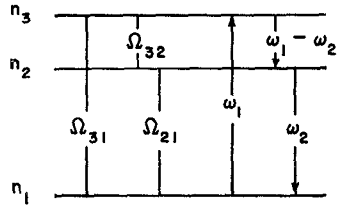

Fig. 4—Energy level diagram of a three-level system used to obtain difference frequency of two applied signals.

No experimental results have been announced to date.

C. Raman Processes

As a final example we shall consider a Raman type process which may be used both for generating signals and as a form of parametric amplifier. There are many possible forms such a system may take; we shall consider only one here. Let us suppose that we have a three-level system as shown in Fig. 5 and that we apply a single frequency ω_1 to this system where $\omega_1>\Omega_{21}$. We shall further assume that transitions are allowed between levels 1 and 3 and 2 and 3. If a radiation field at the frequency $\omega_2=\omega_1-\Omega_{21}$ were present then it would be possible to excite the quantum system from level 1 to 2 with the absorption of a photon at frequency ω_1 and the emission of one at ω_2. The reverse process, absorbing a photon at ω_2 and emitting one at ω_1 is equally probable. If $n_1>n_2$ then there will be more of the first type process and hence a net emission at ω_2. Thus, for a net emission at ω_2, a normal population distribution is desired, consistent with conditions (5) of Section II. With $n_1>n_2$ the signal at ω_2 would be amplified, a net gain occurring if the net emission from the quantum system exceeds the cavity losses. This type of operation is the same as classical parametric amplifier theory where now the quantum system provides the idler frequency, Ω_{21}. If we pump harder at the frequency ω_1, sufficient to overcome both

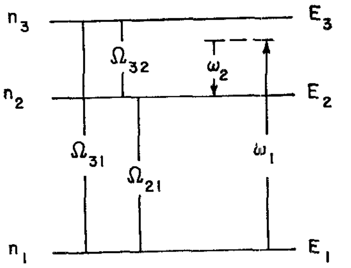

Fig. 5—Energy level diagram for a Raman-type process.

the cavity and coupling losses, then the system will oscillate coherently at the frequency ω_2.

Javan [16] has also studied the possibility of using a two-level system for Raman-type processes where now the self-coupling (*i.e.*, a matrix element connecting the state with itself) of one state or the other takes the place of the level 3 used here.

The pump power level required for such oscillations to begin depends on many factors, among which are the intensities of the allowed transitions, the energy difference, $\hbar(\Omega_{31}-\omega_1)$, the losses in the cavity structure at ω_2. and the coupling of the material to the cavity resonant mode (filling factor). By using suitably strong optical transitions and a strong tunable coherent source such as the ruby laser, along with a tunable cavity at ω_2, it should be possible to achieve a variable frequency source in the millimeter and submillimeter range.

Yajima and Shimoda [17], [18] have studied some aspects of this problem in the microwave range. Experimentally, they have been able to achieve net emission but no oscillations in a traveling wave gas device pumping at 50 kMc with a signal frequency of 5 kMc. Shimoda's group is now attempting this at optical frequencies.

Very recently Woodbury [19] and Eckhardt *et al.* [20] at Hughes have observed coherent Raman processes by using the giant pulse optical maser. The output frequency in their experiments ω_2 was close to the pump frequency, ω_1. This is a result of the fact that the frequency Ω_{21} is in the infrared and not in the optical. Further work should make possible the observation and application of this effect at all frequencies.

D. Other Possible Applications

We have listed several methods for the generation of power, both by harmonic generation processes and by the mixing of optical signals. These form only a representative sample of some of the possible applications. They are for the present, however, some of the most important as far as application to the millimeter wave range are concerned because of the lack of sources in this region. Other uses which may someday result are forms of amplifiers, mixers, detectors, limiters, and modulators. For example, the three level scheme for optical mixing might act as a mixer, with one optical signal being a local oscillator while the Raman-type scheme might be used as a form of tunable amplifier.

IV. Summary and Conclusions

A radiation field may interact with a quantum mechanical system through interactions which involve two or more photons as well as the familiar single quantum interaction. These higher order processes are strong field effects and become increasingly important as the signal levels are raised. As a result of these higher order effects the quantum system may look like a nonlinear element, mixing various frequencies applied to it to give sum and difference terms, the strength of these effects being dependent on the frequencies applied and the properties of the material. Using these phenomena a quantum mechanical system may then be used in various applications as a nonlinear element. Examples have been given of several applications which might be of use in the millimeter wave range. As a result of the difference in principle of operation from the linear maser, different requirements are made of the materials which may be used; notable is the inclusion of materials with short relaxation times.

A method of analysis of problems of this type using the density matrix formulation has been described. This formulation is quite general and is applicable in both the microwave and optical regions of the spectrum. The formulation has the advantage that many level schemes can be handled, relaxation terms may be conveniently added phenomenologically, and it provides some insight into the processes involved.

It should be noted that these multiple quantum effects are not limited to the microwave range of the spectrum but are very general, existing all the way from radio frequencies to optical frequencies.

In short, the purposes of this paper have been threefold: to point out the basic nature of these multiple quantum effects, to describe a method of approach which is applicable to problems of this type and to describe some possible applications.

References

[1] J. P. Gordon, H. J. Zeiger, and C. H. Townes, "The maser—new type of microwave amplifier, frequency standard, and spectrometer," *Phys. Rev.*, vol. 99, pp. 1264–1274; August 15, 1955.
[2] D. ter Harr, "Theory and applications of the density matrix," *Rept. Progr. Phys.*, vol. 14, pp. 304–362; 1961.
[3] U. Fano, "Description of states in quantum mechanics by density matrix and operator techniques," *Rev. Mod. Phys.*, vol. 29, pp. 74–93; 1957.
[4] A. A. Vuylsteke, "Elements of Maser Theory," D. Van Nostrand Company, Inc., New York, N. Y.; 1960.
[5] H. Suhl, "Quantum analog of the ferromagnetic microwave amplifier," *J. Phys. Chem. Solids*, vol. 4, pp. 278–292; 1958.
[6] A. M. Clogston, "Susceptibility of the three-level maser," *J. Phys. Chem. Solids*, vol. 4, pp. 271–277; 1958.
[7] P. W. Anderson, "The reaction field and its use in some solid-state amplifiers," *J. Appl. Phys.*, vol. 28, pp. 1049–1053; September, 1957.
[8] S. Yatsiv, "Role of double-quantum transitions in masers," *Phys. Rev.*, vol. 113, pp. 1538–1544; March 15, 1959.
[9] F. Bloch and A. E. Siegert, "Magnetic resonance for nonrotating fields," *Phys. Rev.*, vol. 57, pp. 522–527; March, 1940.
[10] J. R. Fontana, R. H. Pantell, R. G. Smith, "Harmonic generation by means of multiple quantum transitions," in "Advances in Quantum Electronics," J. R. Singer, Ed., Columbia University Press; New York, N. Y.; 1961.
[11] R. G. Smith, Ph.D. dissertation, Stanford University, Stanford, Calif; July, 1963.

[12] D. P. Akitt, R. J. Strain, P. D. Coleman, "Millimeter Wave Generation by Nonlinear Quantum Susceptibility," presented at 6.6, Millimeter and Submillimeter Conf., Orlando, Fla.; January, 1963.
[13] J. R. Fontana, R. H. Pantell, R. G. Smith, "Harmonic generation using the ammonia inversion transition," PROC. IRE (*Correspondence*), vol. 50, pp. 469–470; April, 1962.
[14] C. M. Kellington, "Resonant harmonic generation in ruby," *Phys. Rev. Lett.*, vol. 9, pp. 57–58; July 15, 1962.
[15] A. Javan and C. H. Townes, "Quarterly Progress Report No. 66," M.I.T. Research Lab. of Electronics, Cambridge, Mass.; July 15, 1962.
[16] A. Javan, "Transitions a plusieurs quanta et amplification maser dans les systemes a deux niveaux," *Le Journal de Physique et le Radium*, vol. 19, pp. 806–808; November, 1958.
[17] T. Yajima and Koichi Shimoda, "Multiple quantum effect in a three-level gas maser," in "Advances in Quantum Electronics," J. R. Singer, Ed., Columbia University Press, New York, N. Y., 1961.
[18] T. Yajima, "Three-level maser action in a gas, I. theory of multiple quantum transition and Doppler effect in three-level gas maser, II. Experimental study on formic acid," *J. Phys. Soc. Japan*, vol. 16, pp. 1594–1606, August, 1961; vol. 16, pp. 1709–1718, September, 1961.
[19] E. J. Woodbury and W. K. Ng, "Ruby laser operation in the near IR," PROC. IRE (*Correspondence*), vol. 50, p. 2367; November, 1962.
[20] G. Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, D. Weiner, and E. J. Woodbury, "Stimulated Raman scattering from organic liquids," *Phys. Rev. Lett.*, vol. 9, p. 455; 1962.

TE_{01} Mode Components in the 3-mm Region*

ALAN J. SIMMONS†, SENIOR MEMBER, IEEE

Summary—**In the 3-mm region (94 Gc) it is desirable to use waveguide components operating in the low loss TE_{01} mode in circular waveguide rather than in fundamental-mode rectangular waveguide. Because this is a higher mode, mode purity is of major concern. A method of identifying undesired modes and their amplitudes is by means of radiation patterns from the end of the waveguide.**

Components developed to operate in this mode include a transition from rectangular to circular waveguide, standing wave detector, variable attenuator, directional coupler, flexible waveguide, fixed 90° bend and rotary joint.

INTRODUCTION

TRANSMISSION of millimeter-wave energy by means of the TE_{01} mode has the advantage over fundamental-mode rectangular waveguide of lower loss, greater power capacity, larger size and simpler flange couplings. It has the advantage over optical or quasi-optical transmission lines of being a closed shielded system with controllable single-mode propagation which permits launching of the mode with high efficiency. Potential use of this mode has been boosted by recent development of high-power magnetrons whose output is in this mode, and which develop enough power to cause RF breakdown in standard air-filled rectangular waveguide at atmospheric pressure.[1]

* Received January 21, 1963. Revised manuscript received April 5, 1963. This work was sponsored by the U. S. Army Signal Research and Development Laboratory, Fort Monmouth, N. J. under Contract No. DA-36-039-sc-88979. This paper was presented at the Millimeter and Submillimeter Conference, Orlando, Fla., January 7–10, 1963.

† Technical Research Group, Inc., East Boston, Mass.

[1] "Inverted coaxial magnetron delivers 100 kw at K_a-band," *Microwaves*, pp. 46–47; March, 1962.

This paper describes the development of components to operate in the TE_{01} mode in the frequency region around 94 Gc (3.2 mm), which is in the neighborhood of a propagation window in the earth's atmosphere.

The components developed or under development in this program include the following:

1) transition from rectangular to circular waveguide
2) mode filter
3) 90° bends
4) 10-db directional couplers to rectangular waveguide
5) flexible sections
6) standing wave detector
7) variable attenuator
8) low- and high-power terminations
9) manual three-port RF switch
10) rotary joint
11) coupling flanges
12) pressurizing window sections

Most of this paper will be concerned with items 6) and 7) which have certain novel features and represent major design problems. A brief description of the method of design and results achieved on some of the other components will be presented.

The inner diameter of the circular waveguide used is 0.250 inch; cutoff freqency for the TE_{01} mode is 57.5