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Production of coherent radiation by atoms and molecules

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From the time when man first saw the sunlight until very recently, the light which he has used has come dominantly from spontaneous emission, like the random emission of incandescent sources. So have most other types of electromagnetic radiation - infrared, ultraviolet, or gamma rays. The maximum radiation intensities, or specifically the power radiated per unit area per unit solid angle per unit frequency bandwidth, have been controlled by Planck's black-body law for radiation from hot objects. This sets an upper limit on radiation intensity - a limit which increases with increasing temperature, but we have had available temperatures of only a few tens of thousands or possibly a few millions of degrees.

Radio waves have been different. And, perhaps without our realizing it, even much of our thinking about radio waves has been different, in spite of Maxwell's demonstration before their discovery that the equations governing radio waves are identical with those for light. The black-body law made radio waves so weak that emission from hot objects could not, for a long time, have been even detected. Hence their discovery by Hertz and the great use of radio waves depended on the availability of quite different types of sources - oscillators and amplifiers for which the idea of temperature and black-body radiation even seems rather out of place. For example, if we express the radiation intensity of a modern electronic oscillator in terms of temperature, it will typically be in the range 10^{10} to 10^{30} degrees Kelvin.

These two regimes, radio electronics and optics, have now come much closer together in the field known as quantum electronics, and have lent each other interesting insights and powerful techniques.

The development of radar stimulated many important applications of electronics to scientific problems, and what occupied me in particular during the late 1940's was microwave spectroscopy, the study of interactions between microwaves and molecules. From this research, considerable information could be obtained about molecular, atomic, and nuclear structure. For its success, coherent microwave oscillators were crucial in allowing a powerful

high-resolution technique. Consequently it was important for spectroscopy, as well as for some other purposes, to extend their range of operation to wavelengths shorter than the known limit of electronic oscillators, which was near millimeter. Harmonic generation and some special techniques allowed interesting, though rather slow, progress. The basic problem with electronic amplifiers or oscillators seemed to be that inevitably some part of the device which required careful and controlled construction had to be about as small as the wavelength generated. This set a limit to construction of operable devices¹. It was this experimental difficulty which seemed inevitably to separate the techniques which were applicable in the radio region from those applicable to the shorter waves of infrared or optical radiation.

Why not use the atomic and molecular oscillators already built for us by nature? This had been one recurring theme which was repeatedly rejected. Thermodynamic arguments tell us, in addition to the black-body law of radiation, that the interaction between electromagnetic waves and matter at any temperature* cannot produce amplification, for radiation at the temperature of matter cannot be made more intense by interaction of the two without violating the second law. But already by 1917, Einstein had followed thermodynamic arguments further to examine in some detail the nature of interactions between electromagnetic waves and a quantum-mechanical system. And a review of his conclusions almost immediately suggests a way in which atoms or molecules can in fact amplify.

The rate of change of electromagnetic energy confined in a region where it interacts with a group of molecules must, from Einstein's work, have the form

$$\frac{dI}{dt} = AN_b - BIN_a + B'IN_b \quad (1)$$

where N_a and N_b are the numbers of molecules in the upper and lower of two quantum states, which we assume for simplicity to be nondegenerate (that is, single). A and B are constants, and thus the first and second terms represent spontaneous emission and absorption, respectively. The third term represents emission from the upper state produced by the presence of a radiation intensity I , and is hence called stimulated emission.

At equilibrium, when

$$\frac{dI}{dt} = 0, \quad I = \frac{AN_b}{BN_a - B'N_b}$$

* Strictly speaking, at any *positive* temperature. Negative absolute temperatures can be defined as will be noted below.

Rather simple further thermodynamic reasoning shows that $B' = B$ and gives the ratio A/B . While Boltzmann's law $N_b = N_a e^{-W/kT}$ requires $N_b < N_a$ at any temperature T , it is immediately clear from Eqn. 1 that if $N_b > N_a$, dI/dt will always be positive and thus the radiation amplified. This condition is of course one of nonequilibrium for the group of molecules, and it hence successfully obviates the limits set by black-body radiation. The condition $N_b > N_a$ is also sometimes described as population inversion, or as a negative temperature², since in Boltzmann's law it may be obtained by assuming a negative absolute temperature.

Thermodynamic equilibrium between two states of a group of atoms requires not only a Boltzmann relation $N_b = N_a e^{-W/kT}$ but also a randomness of phases of the wave functions for the atoms. In classical terms, this means that, if the atomic electrons are oscillating in each atom, there must not be a correlation in their phases if the entire group can be described as in temperature equilibrium. Einstein's relation (Eqn. 1) in fact assumed that the phases are random. And, if they are not, we have another condition which will allow the atoms to amplify electromagnetic waves, even when $N_b < N_a$. This represents a second type of loophole in the limits set by the black-body law and thermodynamic equilibrium, and one which can also be used alone or in conjunction with the first in order to produce amplification.

Thermodynamic arguments can be pushed further to show that stimulated emission (or absorption) is coherent with the stimulating radiation. That is, the energy delivered by the molecular systems has the same field distribution and frequency as the stimulating radiation and hence a constant (possibly zero) phase difference. This can also be shown somewhat more explicitly by a quantum-mechanical calculation of the transition process.

Stimulated emission received little attention from experimentalists during the 1920's and 1930's when atomic and molecular spectroscopy were of central interest to many physicists.

Later, in the 1940's, experiments to demonstrate stimulated emission were at least discussed informally and were on the minds of several radio spectroscopists, including myself. But they seemed only rather difficult demonstrations and not quite worth while. In the beautiful 1950 paper of Lamb and Retherford on the fine structure of hydrogen³ there is a specific brief note about "negative absorption" with reversal of population. And a year later Purcell and Pound⁴ published their striking demonstration of population inversion and stimulated emission. As a matter of fact, population inversion and its effects on radiation had already shown up in a somewhat less accented form in

the resonance experiments of Bloch⁵ and others. But all these effects were so small that any amplification was swamped by losses due to other competing processes, and their use for amplification seems not to have been seriously considered until the work of Basov and Prokhorov⁶, Weber⁷, and of Gordon, Zeiger, and Townes^{8,9} in the early 1950's.

My own particular interest came about from the realization that probably only through the use of molecular or atomic resonances could coherent oscillators for very short waves be made, and the sudden discovery in 1951 of a particular scheme* which seemed to really offer the possibility of substantial generation of short waves by molecular amplification.

Basic Maser Principles

The crucial requirement for generation, which was also recognized by Basov and Prokhorov, was to produce positive feedback by some resonant circuit and to ensure that the gain in energy afforded the wave by stimulated molecular transitions was greater than the circuit losses. Consider a resonant microwave cavity with conducting walls, a volume V , and a quality factor Q . The latter is defined by the fact that power lost because of resistance in the walls is

$$\frac{E^2 V \nu}{4Q}$$

where \bar{E}^2 is the electric field strength in the mode averaged over the volume and ν is the frequency. If a molecule in an excited state is placed in a particular field of strength E , the rate of transfer of energy to the field is

$$\left(\frac{\bar{E}\mu}{\hbar}\right)^2 \frac{h\nu}{3\Delta\nu}$$

when the field's frequency coincides with the resonance frequency ν between the two molecular states. Here μ is a dipole matrix element for the molecular transition and $\Delta\nu$ is the width of the molecular resonance at half maximum (if a Lorentz line shape is assumed). Hence for N_b molecules in the upper state and N_a in the lower state the power given the field in the cavity is

$$(N_b - N_a) \left(\frac{\bar{E}\mu}{\hbar}\right)^2 \frac{h\nu}{3\Delta\nu}$$

If the molecules are distributed uniformly throughout the cavity, E^2 must be averaged over the volume. For the net power gain to be positive, then,

$$(N_b - N_a) \left(\frac{\bar{E}\mu}{\hbar} \right)^2 \frac{h\nu}{3\Delta\nu} \geq \frac{\bar{E}^2 V \nu}{4Q}$$

This gives the threshold condition for buildup of oscillations in the cavity

$$(N_b - N_a) \geq \frac{3h V \Delta\nu}{16\pi^2 Q \mu^2} \quad (2)$$

There is by now an enormous variety of ways in which the threshold condition can be met, and some of them are strikingly simple. But the system which first seemed to give an immediate hope of such an oscillator involved a beam of ammonia molecules entering a resonant cavity, as shown in Fig. 1. The

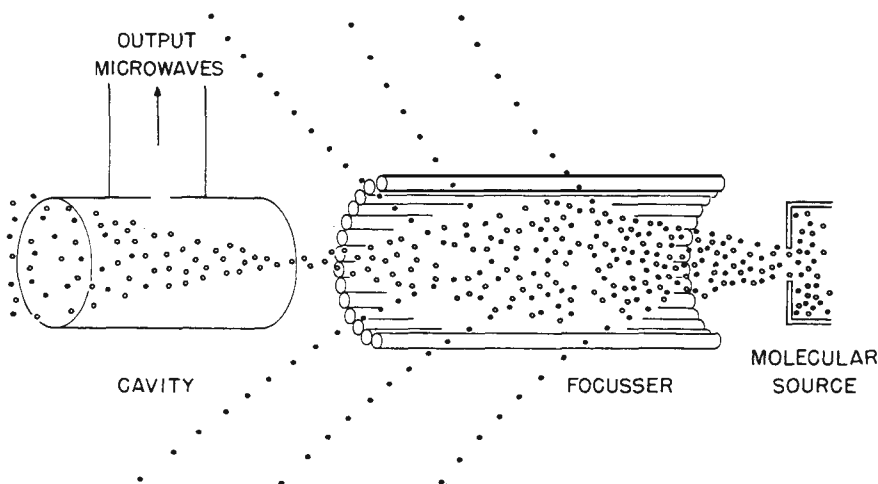


Fig.1. The ammonia (beam-type) maser. Molecules diffuse from the source into a focuser where the excited molecules (open circles) are focused into a cavity and molecules in the ground state (solid circles) are rejected. A sufficient number of excited molecules will initiate an oscillating electromagnetic field in the cavity, which is emitted as the output microwaves. Because of energy given to the field, some molecules return to the ground state toward the end of their transit through the cavity.

transition used was the well-known inversion transition of ammonia at 23,870 Mc/sec. A "focuser", involving inhomogeneous electric fields, tends to remove molecules in the ground state from the beam and to focus molecules in the excited state along the axis of the beam and into the cavity, thus ensuring

that $N_b \gg N_a$. J. P. Gordon played a crucial role in making operable the first such system in 1954, after 2,5 years of experimental work^{9,10}, and H. J. Zeiger was a valuable colleague in the first year of work and early designs. We called this general type of system the maser, an acronym for *m*icrowave *a*mplification by stimulated *e*mission of *r*adiation. The idea has been successfully extended to such a variety of devices and frequencies that it is probably well to generalize the name - perhaps to mean *m*olecular *a*mplification by stimulated *e*mission of *r*adiation. But in the radio-frequency range it is sometimes called the *r*aser, and for light the term *l*aser is convenient and commonly used. Maser amplification is the key process in the new field known as quantum electronics - that is, electronics in which phenomena of a specifically quantum-mechanical nature play a prominent role.

It is well known that an amplifier can usually be made into an oscillator, or *vice versa*, with relatively minor modifications. But it was only after experimental work on the maser was started that we realized this type of amplifier is exceedingly noise-free. The general reason for low noise can be stated simply. The molecules themselves are uncharged so that their motions, in contrast to motions of electrons through vacuum tube amplifiers, produce no unwanted electromagnetic signals. Hence a signal introduced into the resonant cavity competes only with whatever thermal noise is in the cavity as the result of thermal radiation from the cavity walls, and with spontaneous emission from the excited molecules. Spontaneous emission can be regarded for this purpose as that stimulated by a fluctuating field of energy $h\nu$. Since $kT \approx 200h\nu$ for microwaves in a cavity at room temperature, the thermal radiation kT in the cavity is much more important than spontaneous emission. It is then only the thermal radiation present which sets the limit to background noise, since it is amplified precisely as is the signal.

The above discussion also shows that, if the cavity is at 0°K and no extraneous noise enters the cavity with the input signal, the limiting noise fluctuation is determined by the spontaneous emission, which is equivalent to only one quantum of energy in the cavity. It can be shown, in fact, that masers can yield the most perfect amplification allowed by the uncertainty principle.

The motion of an electromagnetic wave is analogous to that of a mechanical harmonic oscillator, the electric and magnetic fields corresponding to position and momentum of the oscillator. Hence the quantum-mechanical uncertainty principle produces an uncertainty in the simultaneous determination of the electric and magnetic fields in a wave, or equivalently in determination of the total energy and phase of the wave. Thus one can show that, to

the extent that phase of an electromagnetic wave can be defined by a quantum-mechanical operator, there is an uncertainty relation¹¹

$$\Delta n \Delta \phi \geq 1/2 \quad (3)$$

Here Δn is the uncertainty in the number of photons in the wave, and $\Delta \phi$ is the uncertainty in phase measured in radians.

Any amplifier which gives some representation of the phase and energy of an input wave in its output must, then, necessarily involve uncertainties or fluctuations in intensity. Consider, for example, an ideal maser amplifier composed of a large number of molecules in the upper state interacting with an initial electromagnetic wave, which is considered the signal. After some period of time, the electromagnetic wave will have grown to such magnitude that it contains a very large number of quanta and hence its phase and energy can be measured by classical means. By using the expected or average gain and phase relation between the final $\epsilon\lambda\chi\tau\rho\mu\alpha\gamma\nu\epsilon\tau\iota\chi$ wave and the initial signal the maser amplifier thus allows a measurement of the initial wave.

A calculation by well-established quantum-mechanical techniques of the relation between input and output waves shows that this measurement of the input wave leaves an uncertainty just equal to the minimum required by the uncertainty principle¹¹. Furthermore, the product $\Delta E \Delta H$ of uncertainties in the electric and magnetic fields has the minimum value allowed while at the same time $(\Delta E)^2 + (\Delta H)^2$ is minimized. The uncertainty in number n of quanta in the initial wave is

$$\Delta n = \sqrt{n+1}$$

and in phase it is

$$\Delta \phi = \frac{1}{2\sqrt{n}}$$

so that

$$\Delta n \Delta \phi = 1/2 \frac{\sqrt{n+1}}{n}$$

The phase has real meaning, however, only when there are as many as several quanta, in which case $n \Delta \phi \rightarrow 1/2$, the minimum allowed by Eqn. 3. The background noise, which is present with no input signal ($n = 0$), is seen to be equivalent to a single quantum ($\Delta n = 1$) of input signal.

A somewhat less ideal maser might be made of N_a and N_b molecules in the upper and lower states, respectively, all interacting with the input signal. In this case fluctuations are increased by the ratio $N_b/(N_b - N_a)$. If the amplifier

has a continuous input signal, a continuous amplified output, and a bandwidth for amplification $\Delta\nu$, the noise power output can be shown to be equivalent to that produced by an input signal¹²

$$N = \frac{h\nu\Delta\nu}{1 - \frac{N_a}{N_b}}$$

The noise power N is customarily described in terms of the noise temperature T_n of the amplifier, defined by $N = kT_n\Delta\nu$. Thus the minimum noise temperature allowed by quantum mechanics is that for a maser with $(N_a/N_b) \ll 1$, which is

$$T_n = \frac{h\nu}{k} \tag{4}$$

This is equivalent to the minimum energy uncertainty indicated above of one quantum ($\Delta n = 1$). In the microwave region, T_n given by Eqn.4 is approximately 1° , whereas the best other microwave amplifiers when maser amplifiers were first being developed had noise fluctuations about 1000 times greater.

It is interesting to compare an ideal maser as a detector with a perfect photodetector, such as a y-ray counter. The y-ray counter can detect a single photon with almost no false signals, whereas a maser must always have a possible false signal of about one photon. But the photodetector gives no information about the phase of the signal; it only counts quanta, which is why the uncertainty principle allows $\Delta n \rightarrow 0$. Unfortunately, there are no perfect photodetectors in the microwave or radio regions, so that the maser is our best available detector for these waves.

The same freedom from noise which makes the maser a good amplifier helps make it a strikingly good source of monochromatic radiation since, when the threshold condition is fulfilled and the maser oscillates, the low noise implies a minimum of random frequency fluctuations.

Consider now a maser oscillator consisting of a group of excited molecules in a resonant cavity. Let the molecular transition frequency be ν_m , its half width at half-maximum intensity $\Delta\nu_m$, and the resonant-cavity frequency be ν_c with a half width $\Delta\nu_c$. If ν_m and ν_c differ by much less than $\Delta\nu_m + \Delta\nu_c$, the radiation produced by the oscillation can be shown to occur at a frequency¹³

$$\nu = \frac{\nu_m Q_m + \nu_c Q_c}{Q_m + Q_c} \tag{5}$$

where the quality factors Q_m and Q_c are $\nu_m/\Delta\nu_c$ and $\nu_m/\Delta\nu_c$ respectively. Thus if the molecular resonance is much sharper than that of the cavity, as in the ammonia-beam maser ($Q_m \gg Q_c$), the frequency of oscillation is¹⁰

$$\nu = \nu_m + (\nu_c - \nu_m) \frac{Q_c}{Q_m} \quad (6)$$

If the cavity is tuned so that $\nu_c - \nu_m$ is small, then the frequency of oscillation coincides very closely with the natural molecular frequency ν_m , and one has an almost constant frequency oscillator based on a molecular motion, a so-called atomic clock.

The frequency ν is not precisely defined or measurable because of noise fluctuations, which produce random phase fluctuations of the wave. In fact, the maser is essentially like a positive feedback amplifier which amplifies whatever noise source happens to be present and thereby produces a more or less steady oscillation. If Q_m or Q_c is high, and the amplifier gain is very large, then the bandwidth of the system becomes exceedingly small. But it is never zero, nor is the frequency ever precisely defined. The average deviation in frequency from Eqn. 5 which these phase fluctuations produce when averaged over a time t is¹⁴

$$\varepsilon = \Delta\nu \left(\frac{W_n}{Pt} \right)^{\frac{1}{2}} \quad (7)$$

where

$$\Delta\nu = \frac{\Delta\nu_c \Delta\nu_m}{\Delta\nu_c + \Delta\nu_m}$$

P is the power generated by the oscillator, and W_n is the effective energy in the source of fluctuations. Where $kT \gg h\nu$ in a cavity at temperature T and resonant frequency ν , the effective energy comes from thermal noise and $W_n = kT$. If the noise fluctuations come from spontaneous emission, as they do when $kT < h\nu$, then $W_n = h\nu$.

It is also useful to state the spectral width of the radiation emitted from a maser oscillator, as well as the precision to which the frequency can be determined. The half width of the spectral distribution is again determined by the same noise fluctuation and is given by^{10,15,16}

$$\delta = \frac{2\pi W_n}{P} (\Delta\nu)^2 \quad (8)$$

where ν , W , and P are the same as in Eqn. 7. This width is typically so small in maser oscillators that they provide by far the most monochromatic sources of radiation available at their frequencies.

Maser Clocks and Amplifiers

Although the ammonia beam-type maser was able to demonstrate the low-noise amplification which was predicted¹⁷, its extremely narrow band-width makes it and other beam-type masers more useful as a very monochromatic source of electromagnetic waves than as an amplifier. For the original maser, the power output P was about 10^9 Watt, and the resonance width about 2 kilocycles, as determined by the length of time required for the beam of molecules to pass through the cavity. Since the frequency of oscillation is 23,874 megacycles, the fractional spectral width, according to Eqn. 8, is $\delta \nu / \nu = 10^{-8}$. In a time $t = 100$ seconds, Eqn. 7 shows that the frequency can be specified to a fractional precision $\delta \nu / \nu = 2 \cdot 10^{-11}$, and of course the precision increases for longer times proportionally to $1/t^{1/2}$.

As a constant-frequency oscillator or precise atomic clock, however, the ammonia maser has an additional problem which is not so fundamental, but which sets a limit on long-term stability. This comes from long-term drifts, particularly of the cavity temperature, which vary ν . These variations can be seen, from Eqn. 6, to « pull » ν . Variations of this type have limited the long-term stability* of ammonia masers to fractional variations of about 10^{-11} ; this still represents a remarkably good clock.

A beam-type maser using the hyperfine structure transition in the ground state of hydrogen, which is at 1420 megacycles, has recently been developed by Goldenberg, Kleppner, and Ramsey¹⁹. In this case, the excited atoms bounce many times from glass walls in the cavity, and thereby a resonance width as small as 1 cycle per second is achieved. Present designs of the hydrogen maser yield an oscillator with long-term fractional variations no larger than about 10^{-13} . This system seems likely to produce our best available clock or time standard.

Masers of reasonably wide utility as amplifiers came into view with the realization that certain solids containing paramagnetic impurities allowed attainment of the maser threshold condition²⁰. Microwave resonances of paramagnetic atoms in solids, or in liquids, had been studied for some time, and many of their properties were already well known. The widths of these re-

sonances vary with materials and with impurity concentration from a small fraction of a megacycle to many hundreds of megacycles, and their frequencies depend on applied magnetic field strengths, so that they are easily tunable. Thus they offer for maser amplifiers a choice of a considerable range of bandwidth, and a continuous range of frequencies.

A paramagnetic atom of spin $\frac{1}{2}$ has two energy levels which, when placed in a magnetic field, are separated by an amount usually of about $\nu = 2.8 H \text{ Mc}$. Here H is the field in gauss, and from this it is clear that most of the microwave frequency range can be covered by magnetic fields of normal magnitudes. The first paramagnetic masers suggested involved impurity atoms of this type in crystals of silicon or germanium. Relaxation between the two states was slow enough in these cases that a sufficient population inversion could be achieved²⁰. However, before very long a very much more convenient scheme for using paramagnetic resonances was proposed by Bloembergen²¹, the so-called three-level solid-state maser. This system allowed continuous inversion of population, and hence continuous amplification, which was very awkward to obtain in the previous two-level system.

Paramagnetic atoms with an angular momentum due to electron spin S greater than $\frac{1}{2}$ have $2S + 1$ levels which are degenerate when the atom is in free space. But these levels may be split by "crystalline fields", or interaction

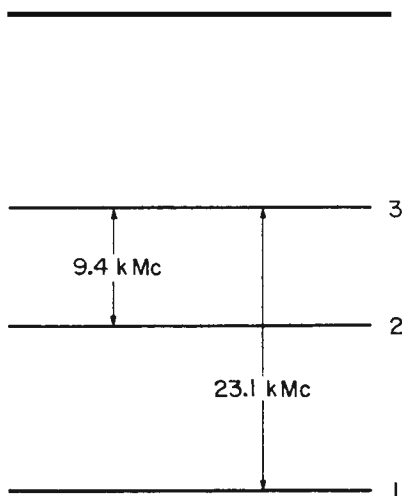


Fig. 2. Energy levels of Cr^{2+} in ruby with a particular crystalline orientation in a magnetic field of 3900 oersteds. For a three-level maser, 23.1 kMc ($23.1 \cdot 10^3 \text{ Mc}$) is the frequency of the pumping field and 9.4 kMc is the frequency of amplification or oscillation.

with neighboring atoms if the atoms are imbedded in a solid, and frequently the splittings lie in the microwave range. The energy levels of such a system, involving a spin of 3/2 and four levels, can be as indicated in Fig. 2 when the system is in a magnetic field. If a sufficiently large electromagnetic wave of frequency ν_{13} (the transition frequency between levels 1 and 3) is applied, the population of these two levels can be equalized or « saturated ». In this case, the ratio of the population of level 2 to that of level 1 or 3 under steady conditions is

$$\frac{n_2}{n_1} = \frac{\frac{1}{T_{12}} e^{-\frac{h\nu_{12}}{kT}} + \frac{1}{T_{23}}}{\frac{1}{T_{12}} + \frac{1}{T_{23}} e^{-\frac{h\nu_{23}}{kT}}}$$

Here T is the temperature of the crystal containing the impurities, and T_{12} and T_{23} are the times for relaxation between the states 1 and 2 or 2 and 3, respectively. For $h\nu_{12} \gg kT$ and $h\nu_{23} \gg kT$, as occurs at very low temperatures or at ordinary temperatures if the levels are separated by optical frequencies,

$$\frac{n_2}{n_1} = \frac{T_{12}}{T_{23}}$$

When $h\nu_{12} \ll kT$ and $h\nu_{23} \ll kT$, which is more commonly the case for microwaves,

$$\frac{n_2}{n_1} = 1 + \frac{h}{kT} \frac{\frac{\nu_{12}}{T_{12}} - \frac{\nu_{23}}{T_{23}}}{\frac{1}{T_{12}} + \frac{1}{T_{23}}} \quad (9)$$

Thus if

$$\frac{\nu_{12}}{T_{12}} > \frac{\nu_{23}}{T_{23}}$$

there is an inversion of population between levels 2 and 1, or if,

$$\frac{\nu_{12}}{T_{12}} < \frac{\nu_{23}}{T_{23}}$$

there is an inversion of population between levels 3 and 2, since the populations n_3 and n_2 have been equalized by the « pumping » radiation. Equation 9 is essentially the result obtained by Bloembergen²¹, who also suggested several promising paramagnetic materials which might be used. Basov and Prok-

horov had already proposed a rather similar three-level « pumping » scheme for application to a molecular beam system²².

The first successful paramagnetic maser of this general type was obtained by Scovil *et al.*²³, using a rare-earth ion in a water-soluble crystal. But, before long, other more suitable crystals such as ruby²⁴ (chromium ions in Al_2O_3) became more or less standard and have provided amplifiers of remarkable sensitivity for radio astronomy, for satellite communication, and for communication with space probes^{2,5}. They have considerably improved the potentialities of radio astronomy, and have already led to some new discoveries^{26,27}. These systems generally require cooling with liquid helium, which is a technological difficulty that some day may be obviated. But otherwise they represent rather serviceable and convenient amplifiers.

A maser amplifier of microwaves can rather easily be built which has a theoretical noise temperature as low as 1° or 2°K , and experimental measurements have confirmed this figure²⁸. However, such a low noise level is not easy to measure because almost any measurement involves attachment of input and output circuits which are at temperatures much higher than 1°K , and which radiate some additional noise into the amplifier. The lowest overall noise temperature so far reported for an entire receiving system²⁹ using a maser amplifier is about 10°K . This represents about 100 times the sensitivity of microwave amplifiers built before invention of the maser. But masers have stimulated other amplifier work, and some parametric amplifiers, using more or less classical properties of materials rather than quantum electronics, now have sensitivities within a factor of about 5 of this figure.

Optical and Infrared Masers, or Lasers

Until about 1957, the coherent generation of frequencies higher than those which could be obtained from electronic oscillators still had not been directly attacked, although several schemes using molecular-beam masers for the far-infrared were examined from time to time. This lack of attention to what had been an original goal of the maser came about partly because the preliminary stages, including microwave oscillators, low-noise amplifiers, and their use in various scientific experiments, had proved so interesting that they distracted attention from the high-frequency possibilities.

But joint work with A. L. Schawlow³⁰, beginning at about this time, helped open the way for fairly rapid and interesting development of maser oscillators

in the far-infrared, optical, and ultraviolet regions - as much as 1000 times higher in frequency than any coherent sources of radiation previously available. It is masers in these regions of the spectrum, frequently called lasers (light amplification by stimulated emission of radiation), which have perhaps provided the most striking new scientific tools and results. Important aspects of this work were clear demonstrations that there are practical systems which can meet the threshold condition of oscillation, and that particular resonator designs allow the oscillations to be confined to certain specific and desirable modes. The resonator analyzed was composed simply of two parallel mirrors-the well-known Fabry-Perot interferometer, but of special dimensions.

For light waves, the wavelength is so short that any macroscopic resonator constructed must have dimensions that are large compared with the wavelength. In this case, the electromagnetic field may to some reasonable approximation be considered to travel in straight lines and be reflected from the walls of the resonator. The threshold condition may be written

$$\left(\frac{\mu E}{\hbar}\right)^2 \frac{h\nu (N_b - N_a)}{12\pi \Delta\nu} \geq \frac{E^2}{8\pi} \frac{V}{t} \tag{10}$$

where t is the decay time for the light in a cavity of reflecting walls and volume V . If the light has a random path in the cavity, the decay time can be expressed generally in terms of the reflection coefficient r of the walls, the volume V , the wall area A , and the velocity of light c ,

$$t = \frac{6V}{(1-r)Ac}$$

Hence Eqn. 10 becomes³⁰

$$N_b - N_a \geq \frac{\Delta\nu}{\nu} \frac{h(1-r)Ac}{16\pi^2 \mu^2 V} \tag{11}$$

It can be seen that this critical condition is almost independent of frequency if the fractional line width $\Delta\nu/\nu$ does not change with frequency (as, for example, in the Doppler effect). The reflection coefficient and dipole moment matrix element are not particularly dependent on frequency over the range in question. Hence, if the critical condition can be met for one frequency, it can probably be met over the entire range from the far-infrared to the ultraviolet.

There is a problem with a resonator which is large compared to a wavelength in that there are many modes. Hence, unless the modes in which oscilla-

tions occur are successfully controlled, the electromagnetic field may build up simultaneously in many modes and at many frequencies. The total number of modes in a cavity with frequencies which lie within the line width of the atomic molecular resonance is

$$p = \frac{8\pi^2 V \nu^2 \Delta\nu}{c^3}$$

or about 10^3 for a cavity volume of 1 cm^3 , a frequency in the optical region, and ordinary atomic line widths. But fortunately the possibility of oscillation can be eliminated for most of these modes.

Two small parallel mirrors separated by a distance much larger than their diameter will allow a beam of light traveling along the axis joining them to travel back and forth many times. For such a beam, the decay time t is $L/c(1-r)$, where L is the mirror separation and r the reflectivity. Hence the threshold condition is

$$N_b - N_a \geq \frac{3\Delta\nu}{8\pi^2\nu} \frac{hc(1-r)}{\mu^2 L}$$

This assumes that diffraction losses are negligible. A beam of light which is not traveling in a direction parallel to the axis will disappear from the volume between the mirrors much more rapidly. Hence the threshold condition for off-axis beams will require appreciably more excited atoms than that for axial beams, and the condition for oscillation can be met for the latter without a build-up of energy in off-axis light waves.

Many features of the modes for the electromagnetic wave between two square, plane, parallel mirrors of dimension D and separation L can be approximately described as those in a rectangular box of these dimensions, although the boundary conditions on the enclosed sides of the « box » are of course somewhat different. The resonant wavelengths of such a region for waves traveling back and forth in a nearly axial direction are³⁰

$$\lambda = \frac{2L}{q} \left[1 - \frac{1}{2} \left(\frac{Lr}{Dq} \right)^2 - \frac{1}{2} \left(\frac{Ls}{Dq} \right)^2 \right] \quad (12)$$

where q , r , and s are integers, and $r \ll q$, $s \ll q$. More precise examination of the modes requires detailed numerical calculation³¹. For a precisely axial direction, $r = s = 0$, and the modes are separated by a frequency $c/2L$. If this frequency is somewhat greater than the atomic line width, then only one axial mode can oscillate at a time. The axial wave has an angular width due to

diffraction of about λ/D , and, if this is comparable with the angle D/L , then all off-axis modes (r or $s \neq 0$) are appreciably more lossy than are the axial ones, and their oscillations are suppressed.

If one of the mirrors is partially transparent, some of the light escapes from the axial mode in an approximately plane wave and with an angular divergence, approximately λD , determined by diffraction.

A number of modified resonator designs have been popular and useful in optical masers, in particular ones based on the confocal Fabry-Perot interferometer. However, the plane-parallel case seems to offer the simplest means of selecting an individual mode.

Although a number of types of atomic systems and excitation seemed promising in 1958 as bases for optical masers, optical excitation of the alkali vapors lent itself to the most complete analysis and planning for an operable oscillator. One such system has been shown to oscillate as expected³²; but the alkali vapors are no longer of great interest, because other systems which were at the time much less predictable have turned out to be considerably more useful.

The first operating laser, a system involving optical excitation of the chromium ions in ruby and yielding red light, was demonstrated by Maiman in 1960³³. He took what seemed at first a rather difficult route of inverting the population between the ground state and excited states of the chromium ion. This technique requires that at least half of the very large number of atoms in the ground state must be excited in order to have the possibility of a population inversion. In the case of two normally unpopulated atomic states, the total amount of excitation required is much less. However, Maiman succeeded handsomely in exciting more than half the chromium ions in a ruby with chromium concentration of about 1/2000 by applying a very intense pulse of light from a flash tube. This type of system is illustrated schematically in Fig. 3. Success immediately yielded a very-high-energy maser oscillation because, to get population inversion at all, a large amount of energy must be stored in the excited atomic states. Surfaces of the ruby served as the reflecting mirrors. Collins *et al.*³⁴ quickly demonstrated that the ruby laser showed many of the characteristics predicted for such an oscillator.

The ruby laser is operated normally only in pulses, because of the high power required to reach threshold, and emits intense bursts of red light at power levels between about 1 kilowatt and 100 megawatts. It has given rise to a whole family of lasers involving impurities in various crystals of glasses, and covering frequencies from the near infrared into the optical region.

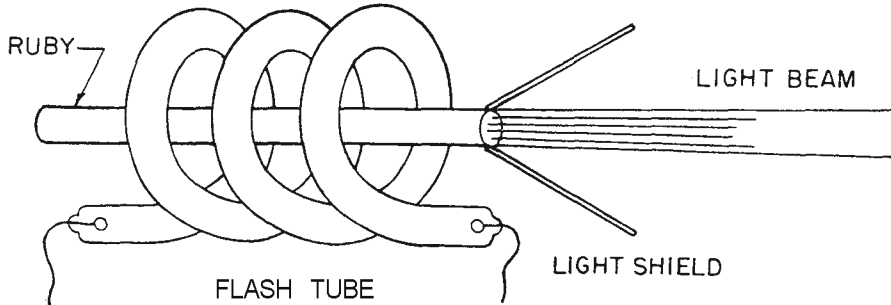


Fig. 3. Schematic diagram of a ruby (optically excited solid-state) laser. When the gas flash tube is activated, electromagnetic oscillations occur within the ruby rod, and some of these light waves are emitted in a beam through one partially reflecting end of the rod.

Not very long after the ruby laser was developed, Javan, Bennett and Herriott³⁵ obtained maser oscillations from neon atoms excited by collisions of the second kind with metastable helium, in accordance with an idea previously put forward by Javan³⁶. This system, illustrated in Fig. 4, requires only a gaseous discharge through a tube containing a mixture of helium and neon at low pressure, and two reflectors at the ends of the tube. It oscillates at the

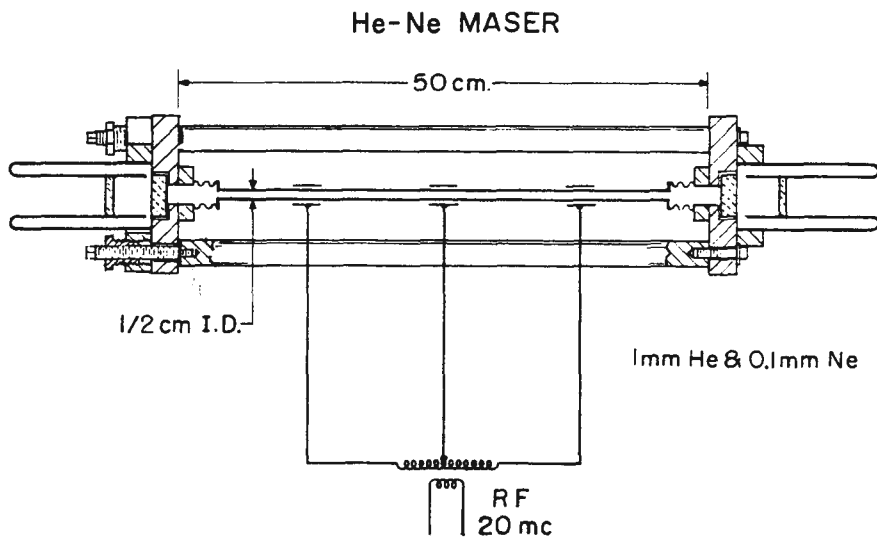


Fig. 4. Schematic diagram of a helium-neon (gas discharge) laser. Electrical excitation can initiate a steady maser oscillation, resulting in an emitted light beam from either end of the gas discharge, where there are reflecting mirrors.

relatively low power of about 1 milliwatt, but approaches ideal conditions much more closely than the ruby system, and affords a continuous source of infrared radiation of great purity and directivity.

The technique of gaseous excitation by electrical discharge has also led to a large family of lasers, producing hundreds of different frequencies from many different gases which range from wavelengths as long as a few tenths of 1 millimeter down into the ultraviolet. For some systems, a heavy discharge pulse in the gas is needed. Others, particularly some of the infrared frequencies in rare gases, oscillate so readily that it seems probable that we have had lasers accidentally all along. Very likely some neon or other rare-gas electric signs have been producing maser oscillations at infrared wavelengths, which have gone unnoticed because the infrared could not escape from the glass neon tubes. Some of these oscillation frequencies represent atomic transitions which were previously undetected; for others, the transition has not yet even been identified.

Another class of lasers was initiated through the discovery³⁷ that a *p-n* junction of the semiconductor gallium arsenide through which a current is passed can emit near-infrared light from recombination processes with very high efficiency. Hall *et al.*³⁸ obtained the first maser oscillations with such a system, with light traveling parallel to the junction and reflected back and forth between the faces of the small gallium arsenide crystal. His results were paralleled or followed immediately, however, by similar work in two other laboratories^{39,40}. This type of laser, illustrated in Fig. 5, is of the general size and cost of a transistor. It can be made to oscillate simply by passage of an electric current, and in some cases the radiation emitted represents more than 50 percent of the input electrical energy - an efficiency greater than that of other man-made light sources.

There quickly developed a large family of semiconductor lasers, some involving junctions and, recently, some using excitation by an external beam of electrons⁴¹. They range in wavelength from about 10 microns, in the infrared, to the center of the visible region.

Normal Raman scattering can be regarded as spontaneous emission from a virtual state, as indicated in Fig. 6. Associated with any such spontaneous emission there must be, in accordance with Einstein's relations, a stimulated emission. Javan showed⁴² the principles involved in using this stimulated emission for a Raman maser. What is required is simply a large enough number of molecular systems which are sufficiently strongly excited by radiation of frequency greater than some Raman-allowed transition.

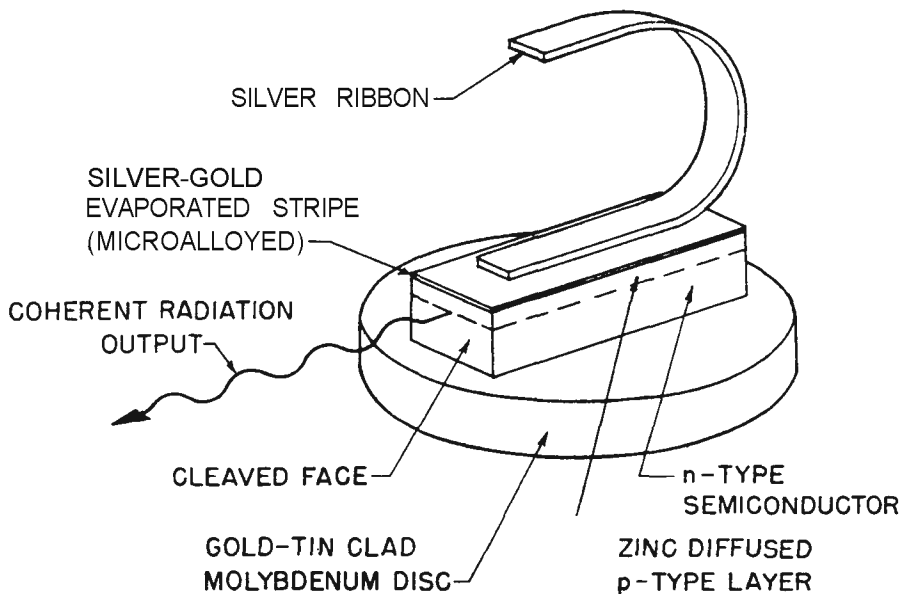


Fig. 5. Schematic diagram of a gallium arsenide (injection, or semiconductor) laser. A small voltage applied between the silver ribbon and the molybdenum disc can produce maser oscillations with resulting emission of coherent infrared radiation.

One might consider the population of the virtual level in a Raman maser (see Fig. 6) to be greater than that of the first excited state, so that there is no population inversion. On the other hand the initial state, which is the ground state, needs to be more populated than the first excited state. One can quite properly consider the amplification process as a parametric one with the molecular frequency as idler, or as due to a mixture of ground and excited states in which there is phase coherence between the various molecules. This is the second type of loophole through the black-body radiation law mentioned earlier. The ammonia-beam maser itself illustrates the case of amplification without the necessity of population inversion. As the ammonia molecules progress through the cavity and become predominantly in the ground state rather than the excited state, they continue to amplify because their oscillations are correlated in phase with each other, and have the appropriate phase with respect to the electromagnetic wave.

Raman masers were first demonstrated by Woodbury and Ng⁴³ as the result of excitation of various liquid molecules with a very intense beam from a pulsed laser. They too have now many versions, giving frequencies which

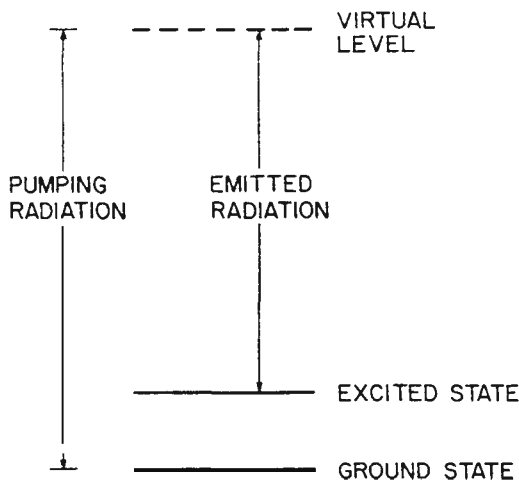


Fig. 6. Representation of energy levels in a Raman maser. This system resembles qualitatively a three-level maser, one of the levels being « virtually », or not characteristic of the molecule when no field is present.

differ from the original driving maser beam by some small integer times a molecular-vibrational frequency. Their action has been considerably extended by Terhune⁴⁴ and treated in a number of theoretical papers^{42,45}.

Present Performance of Lasers

Where now do we stand in achieving the various theoretical expectations for performance of masers?

First, consider the general extension of the frequency range where we have coherent amplifiers and oscillators. This has been increased by a factor of somewhat more than 1000; there are still additional spectral regions where such techniques need to be developed, but the pace has been quite rapid in the last few years. Maser oscillations in the infrared, optical, and ultraviolet regions have now been obtained in many ways and appear easy; new excitation mechanisms and systems are continually turning up. There are still two frequency regions, however, where such sources of radiation are rare or non-existent. One is in the submillimeter region or far-infrared. The region has, in a sense, now been crossed and conquered by maser oscillators. But techniques in this spectral region are still rudimentary, and the frequency coverage with

masers is spotty. Presumably further work will allow interesting explorations in this region and a very fruitful, high-resolution spectroscopy.

Another region where coherent oscillators have not yet been developed is that of still shorter wavelengths stretching indefinitely beyond the near-ultraviolet, where the first such oscillators are now available. It can be shown that a rather severe and fundamental limitation exists as one proceeds to shorter wavelengths because of the continually increasing number of electromagnetic modes in a given volume and the faster and faster dissipation of energy into them by spontaneous emission.

Consider a cavity resonator of fixed volume, fixed-wall reflectivity, and fixed-fractional frequency-width $\Delta\nu/\nu$. Meeting the threshold condition (Eqn. II) in such a resonator requires that there is power which increases as ν^4 , radiated by spontaneous emission into all modes of the system³⁰. In the optical region this dissipated power for typical conditions, whereas at 50 ångströms, in the soft X-ray region, it would be about 10^5 watts. The threshold condition would then be very difficult to maintain. But, by the same token, if it is maintained, the coherent X-ray beam produced would contain many kilowatts of power. It seems reasonable to expect, on this basis, that masers will be developed to wavelengths somewhat below 1 000 ångströms, but that maser oscillations in the X-ray region will be very much more difficult.

Secondly, let us examine the monochromaticity which has been achieved. For the ammonia-beam maser, the variation of microwave oscillations was shown experimentally to agree with the theoretical expression (Eqn. 7) within the experimental precision of about 50 percent. This was done by beating two independent ammonia oscillators together and examining their relative phase variations⁴⁶. A similar procedure can be carried out for two optical oscillators by mixing their two light beams together in a photocell and detecting the beat frequency. However, the technical difficulties in obtaining theoretical performance are rather more demanding than in the case of the ammonia maser. Equation 8 for a typical helium-neon laser predicts a frequency spread of about 10^{-2} cycle per second, or a fraction $3 \cdot 10^{-17}$ of the oscillation frequency of $3 \cdot 10^{14}$ cycles per second.

Almost all masers so far oscillating in the optical or near-infrared region require a sharper resonance, or higher Q, of the cavity than of the atomic resonance. Hence the frequency of oscillation is primarily determined, from Eqn. 5 by the cavity resonance. The frequency of oscillation thus depends on the separation L between mirrors, since from Eqn. 12 $\nu = qc/2L$, where q is

some integer. If, then, the radiated frequencies are to have a fractional bandwidth of about $3 \cdot 10^{-17}$, such as would come from fundamental noise according to Eqn. 8, the mirror separation must not vary by more than this fractional amount. For a mirror separation of 1 meter, the motion allowed would be less than $3 \cdot 10^{-13}$ centimeter - a demanding requirement!

If the mirror separation is held constant by cylindrical rods, L must still vary as a result of thermal excitation of the lowest frequency-stretching modes of the rods. This gives an additional fluctuation which is usually larger than that from spontaneous emission. It produces a fractional motion¹⁷

$$\left(\frac{2kT}{YV}\right)^{\frac{1}{2}}$$

where T is the temperature, V the volume of the separators, and Y their Young's modulus.

In order to examine the monochromaticity of lasers, two helium-neon systems were carefully shock-mounted in an acoustically insulated wine cellar of an unoccupied and isolated house so that acoustic vibrations would be minimized¹⁷. Their pairs of mirrors were separated by heavy invar rods about 60 centimeters long. For this case, the limiting theoretical fluctuations set by thermal motions of the rods corresponded to fractional frequency variations of $5 \cdot 10^{-15}$, or a frequency fluctuation of 2 cycles per second. Light from each laser was sent into a photodetector, and the beat frequency examined electronically. Under good conditions free from acoustic disturbances or thermal transients in the invar spacers, this experiment showed that variations in the laser frequencies over periods of a few seconds were less than 20 cycles per second, or about one part in 10^{13} . This was ten times the limit of thermal fluctuations, but corresponded to detection of motions of the two mirrors as small as $5 \cdot 10^{-12}$ centimeter, a dimension comparable with nuclear diameters. Presumably, with great care, one can obtain results still nearer to the theoretical values.

The narrowest atomic spectral lines have widths of the order of 10^8 cycles per second, so that the laser measured was more monochromatic than earlier light sources by a factor of about 10^6 . Light of this type can interfere with itself after traveling a distance of about 10 000 kilometers. Hence it could in principle measure changes in such a large distance to a precision of one wavelength of light, if there were any optical path so constant. Interference work has been done in several laboratories with laser light over distances of a few hundred meters, which does not require quite such special elimination of acoustic or other disturbances.

A third property of laser light which is of interest is its directivity, or the spacial coherence across the beam. As indicated above, certain modes of oscillation should represent approximately a plane wave of cross section comparable with the mirror diameter D . The helium-neon maser seems to easily allow adjustment so that such a mode of oscillation occurs, and its beam has been shown^{35,48} to have nearly the expected divergence λ/D due to diffraction.

The spacial coherence or planarity of a laser beam implies that the entire beam can be focused by a microscope to a region as small as about $\lambda/2$, or the resolving power of the microscope. Similarly, it may be transmitted through a telescope in a beam whose angular width is simply determined by the angular resolution of the telescope, and hence much less than the angular divergence λ/D as the beam emerges from a small laser. The entire energy is originally created in the ideal laser in a single mode; it can be transmitted into other single modes by optical systems without violating the well-known brightness laws of optics.

This brings us to a fourth important property, the intensity or brightness which can be achieved by maser techniques. As indicated initially, once one has the possibility of coherent amplification, there is no firm limit to intensity because equilibrium thermodynamics and Planck's law no longer are controlling. The only limit is set by the available energy input, heat dissipation, and size of the apparatus used.

If only the 1 milliwatt of power emitted by a helium-neon laser is focused by a good lens, the power density becomes high because the cross-sectional area of the focused spot would be only about $\lambda^2/4$. This gives a power density of $4 \cdot 10^5$ Watt/cm². The effective temperature of such a beam, because of its monochromaticity, is also rather high - about 10^{19} °K for the light of 20-cycle per-second bandwidth.

The pulsed systems, such as ruby lasers in particular, emit much greater power, although they do not quite approach the limits of coherence which the gaseous systems do. Ruby lasers emit a few tenths of a joule to a few hundred joules of energy in pulses from about 10^3 second to 10^8 second in length. The power can thus be as great as 10^9 Watts or more. Effective temperatures of the radiation are of the order 10^{23} °K. The actual limit of power density will generally be set by the limit of light intensity optical materials can stand without breakage or ionization. Power of 10^9 Watts focused to a spot 10^2 millimeter in diameter produces an electric field strength in the optical wave of about 10^9 Volt/cm, which is in the range of fields by which valence electrons are held in atoms. Hence this power ionizes and disrupts all material.

The radiation pressure also becomes large, being about 10^{12} dyne/cm², or 10^6 atmospheres, at such a focal point.

Some Applications of Lasers

It is clear that light in more ideal and in more intense form, which maser techniques have produced, can be expected to find application in wide and numerous areas of technology and of science simply because we find our present techniques of producing and controlling light already so widely applied. Most of these applications are still ahead of us, and there is not time to treat here even those which are already beginning to develop. I shall only mention that in technology lasers have been put to work in such diverse areas as radar, surgery, welding, surveying, and microscopy. A little more space will be devoted here to discussing three broad areas of science to which optical, infrared, and ultraviolet masers are expected to contribute.

Masers seem to provide the most precise techniques for measurement of the two fundamental dimensions of time and length. Over short periods of time maser oscillators clearly give the most oscillations; for longer times the hydrogen maser also seems to provide the most precise clock yet available. Light from optical masers allows new precision in the measurement of distance, and already seems capable of improving our standard of length. This new precision suggests interesting experiments on certain fundamental properties of our space, as well as the application of higher precision to a variety of physical effects. So far, experiments have been done to improve the precision with which the Lorentz transformation can be experimentally verified^{49,50}. It appears that improved precision in measurement of the speed of light can also be expected. If we look some distance in the future, it seems clear that the techniques of quantum electronics will allow direct measurement of the frequency of light, rather than only its wavelength. This can be accomplished by generation of harmonics of a radio frequency, amplification of the new frequency, and further generation of harmonics until the radio region is linked with optical frequencies. This should eventually allow measurement of the velocity of light, c , to whatever precision we define time and length. Or, it will allow the elimination of separate standards of time and of length because c times a standard time will define a standard length with more precision than we can now achieve.

The power of spectroscopy should be considerably increased by use of

masers. In particular, these very monochromatic sources can very much improve spectroscopic resolution and thus allow more detailed examination of the structure of atoms, molecules, or solids. This advance can be particularly striking in the infrared and far-infrared, where present resolution is far less than the widths of atomic or molecular lines. Already some high-resolution spectroscopy has been done with lasers^{51, 52} and still more interesting work of this general type can be expected before long.

A third interesting field for which lasers are important has emerged as a field almost entirely because of the existence of lasers, and is the area where scientific research has so far been most active. This is what is usually called nonlinear optics^{53, 54} although it includes some phenomena which might not previously have been described in this way. We have been accustomed in the past to discussing the progress of light through a passive optical material of more-or-less fixed properties. But, in the intense laser beams now available, interactions between the light and the optical medium are sufficiently large that properties of the medium can no longer be regarded as fixed. The medium distorts, it molecules vibrate, and polarization of electrons in its atoms no longer responds linearly to the applied field. One must now also consider the dynamics medium, and interactions between their two motions. Some of the new phenomena observed are multiple-quanta absorption, which makes absorption depend on intensity^{55, 56}, harmonic generation in optical materials and mixing of light frequencies⁵⁷⁻⁶⁰, excitation of coherent molecular vibrations and stimulated Raman effects⁴²⁻⁴⁵, and stimulated Brillouin scattering^{61, 62}. Only the last two of these will be discussed, partly because they bear on still another kind of maser, one which generates phonons.

The Phonon Maser

Acoustic waves follow equations that are of the same general form as the equations of light and manifest many of the same phenomena. An acoustic wave can produce an atomic or molecular excitation, or receive energy from it by either spontaneous or stimulated emission. Hence, one may expect maser action for acoustic waves if a system can be found in which molecules are sufficiently coupled to an acoustic field and appropriate excitation can be obtained to meet the threshold condition. The first such systems suggested involved inversion of the spin states of impurities in a crystal in ways similar to those used for solid-state electromagnetic masers⁶³. A system of this type has

been shown to operate as expected⁶⁴. However, a more generally applicable technique seems to be Brillouin scattering and its close associate Raman scattering, which utilize phase correlation rather than population inversion to produce amplification. This process can also be viewed as parametric amplification.

Light may be scattered by the train of crests and troughs in an acoustic wave much as by a grating. Since the wave is moving, the scattering involves a Doppler shift. The net result, first analyzed by Brillouin⁶⁵, is that the scattered light is shifted in frequency from the frequency ν_0 of the original beam by an amount

$$\nu = 2 \nu_0 v / c \sin \theta / 2 \quad (13)$$

where v and c are the phase velocities of sound and of light, respectively, in the medium, and θ is the scattering angle. The energy lost, $h\nu$, is given to the scattering acoustic wave of frequency ν . If the light is of sufficient intensity, it can thus give energy to the acoustic field faster than it is lost and fulfill a threshold condition which allows the acoustic energy to build up steadily.

For the very high acoustic frequencies (10^9 to 10^{10} c/ sec) implied by Eqn.13 when θ is not very small, the losses are usually so large that interesting amplification cannot be achieved with ordinary light. But, with laser beams of hundreds of megawatts per square centimeter, it is quite feasible to produce an intense build-up of acoustic waves by this process of stimulated Brillouin scattering^{61, 62} - so intense, in fact, that the acoustic energy can crack glass or quartz. This gives a method of producing and studying the behavior of very-high-frequency acoustic waves in almost any material which will transmit light - a possibility which was previously not so clearly available.

Brillouin scattering by spontaneous emission has been studied for some time. But the intense monochromatic light of lasers allows now much greater precision in work with this technique⁵² and it too is yielding interesting information on the propagation of hypersonic waves in materials.

There is no firm limit to the acoustic frequencies which can be produced by stimulated emission, even though Eqn.13 indicates a kind of limit, for $\theta = \pi$, of $2\nu_0 v / c$. But in the optical branch of acoustic waves the phase velocity v can be very high. In fact, stimulated Raman scattering, or the Raman maser mentioned briefly above, represents excitation of the optical branches of acoustic spectra, and generates coherent molecular oscillations. Quantum-electronic techniques can thus allow interesting new ways to generate and explore most of the acoustic spectrum as well as much of the electromagnetic domain.

Concluding Remarks

In a few years this brief report will no longer be of much interest because it will be outdated and superseded, except for some matters of general principle or of historical interest. But, happily, it will be replaced by further striking progress and improved results. We can look forward to another decade of rapid development in the field of quantum electronics - new devices and unsuspected facets of the field, improved range and performance of masers, and extensive application to science and to technology. It seems about time now for masers and lasers to become everyday tools of science, and for the exploratory work which has demonstrated so many new possibilities to be increasingly replaced by much more finished, more systematic, and more penetrating applications. It is this stage of quantum electronics which should yield the real benefits made available by the new methods of dealing with radiation.

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