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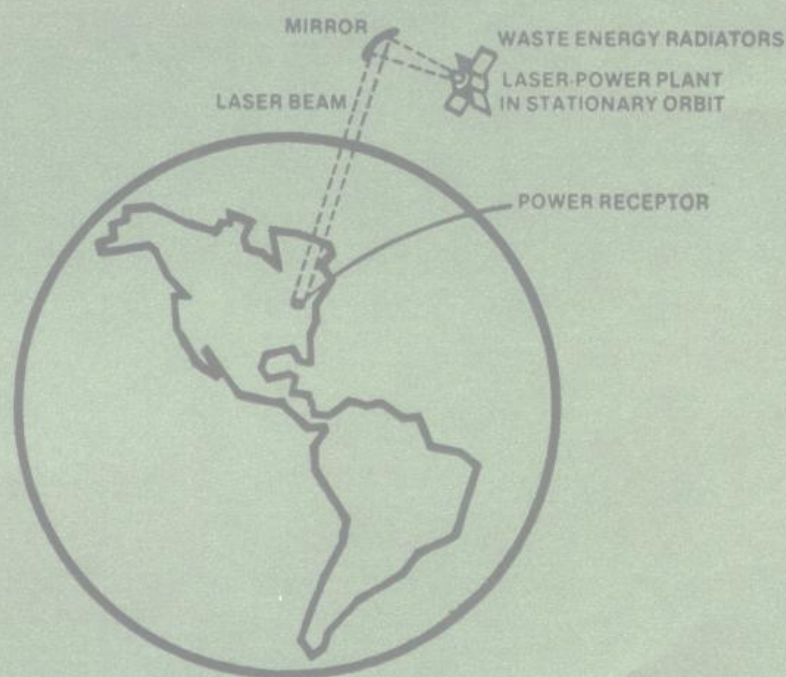
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HIGH ENERGY LASERS

and Their Application

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PROFESSIONAL STUDY SEMINAR

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16. Fenstermacher, C. A., Nutter, M. J., Leland, W. T. and Boyer, K., "Electron-Beam-Controlled Electric Discharge as a Method of Pumping Large Volumes of CO_2 Laser Media at High Pressure," _Appl. Physics Letters, 20_, No. 2, January 1972, pp. 56-60.

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High-Power Carbon Dioxide Lasers

Lasers in which the active medium consists of a low-pressure gas of carbon dioxide molecules have been used to produce the most powerful continuous laser beams achieved to date

by C. K. N. Patel

Until quite recently it was generally assumed that the most powerful lasers that would ever be built would be solid-state lasers, for the simple reason that in a solid the "lasing" particles are much more concentrated than they are in a gas. Nevertheless, it was recognized almost from the beginning that solid-state lasers have their drawbacks. With respect to two important criteria of laser performance—the spectral purity and the spatial coherence of the output light beam—solid-state lasers rate rather poorly. Moreover, most high-power solid-state lasers operate only in the pulsed mode; in other words, their power output consists of short, intense bursts of light rather than a continuous beam. In contrast the original atomic-gas lasers produced continuous beams with excellent spectral purity and spatial coherence, but their power output was very low compared with the power output from the solid-state lasers.

This situation has changed entirely with the advent of the molecular-gas lasers. The outstanding example of this new class of lasers is the carbon dioxide laser, which can produce a continuous laser beam with a power output of several kilowatts while at the same time maintaining the high degree of spectral purity and spatial coherence characteristic of the lower-power atomic-gas lasers. A carbon-dioxide laser was recently used to produce an infrared beam with an output power of 8.8 kilowatts—the most powerful continuous laser beam achieved to date. The significance of such a power output is vividly demonstrated by the fact that a focused infrared beam of a few kilowatts is capable of cutting through a quarter-inch steel plate in a matter of seconds [*see illustration at right*].

Because of their high power output in the infrared region of the electromagnetic spectrum, the carbon dioxide gas lasers have opened up a whole new range of wavelengths for the study of nondestructive optical interactions with gases, liquids and solids. Such optical interactions include nonlinear processes whereby one can generate a coherent source of infrared radiation that is continuously tunable over a range of frequencies. There are in addition a variety of other applications for which high-power carbon dioxide lasers promise to be useful. Perhaps the most important potential application is in the area of optical communications and optical radar. The carbon dioxide laser is particularly suited for use in both terrestrial and extraterrestrial communication systems because the infrared beam it produces is only slightly absorbed by the atmosphere. In this article I shall attempt to explain the physics underlying the operation of this new type of high-power gas lasers.

In general a gas laser consists of a low-pressure gas-filled vessel (called the laser tube) located between two mirrors that form an "optical cavity." The gas in
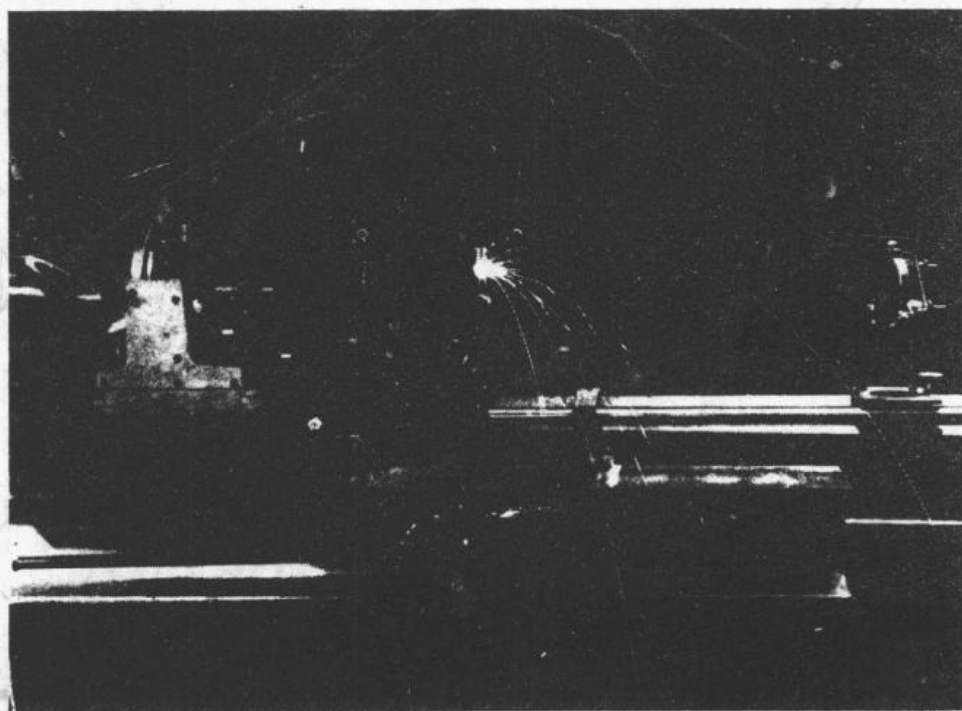
HIGH-POWER INFRARED BEAM from the Redstone Arsenal carbon dioxide laser is shown burning a hole through a quarter-inch-thick stainless-steel plate—a job that takes about 10 seconds. The infrared beam emerges through the slanted Brewster window at the end of the laser tube (*left*) and passes through a partially reflecting end mirror before striking a concave mirror (*right*), which focuses it on the steel plate (*center*). The thermal applications of the high-power carbon dioxide lasers hold considerable promise for industry but are regarded as secondary to the potential use of such lasers in optical communication systems.

the tube (called the laser medium) can consist of atoms, metallic vapor or molecules. Laser action is usually obtained in the gas by subjecting it to an electric discharge; the energetic electrons provided by the discharge collide with the active gas particles, exciting them to higher energy levels from which they spontaneously descend to lower energy levels, emitting their excess energy in the form of photons, or light quanta. In order to achieve the optical "gain" that characterizes laser action it is necessary that the "population density" of particles in the upper energy level exceed that in the lower energy level. This condition is known as population inversion, since it is the inverse of the normal, or nonexcited, state of affairs. In order to achieve a high output power on a given transition between a pair of energy levels, it is also necessary that the absolute number of atoms excited to the upper laser level be large and that the gas particles leave the lower laser level just as fast as they arrive from the upper level. In other words, the "depopulation," or de-excitation, of the particles in the lower laser level is just as important as the excitation of particles from the ground state to the upper laser level, since a particle that has already contributed to the laser output must return to the ground state before it is available again for excitation to the upper level in order to produce additional laser power.

The energy expended by particles in dropping from the lower laser level back to the ground state contributes nothing to the power output of the laser. Hence a certain amount of energy is wasted for every particle that makes the laser transition. This fact suggests an obvious yardstick for judging the efficiency of a particular laser system. The amount of energy wasted by a particle in returning from the lower laser level to the ground state is equal to the difference between the energy needed to excite the particle to the upper laser level and the energy of the photon of light that is emitted when the particle makes the transition from the upper laser level to the lower laser level. It follows that the ratio of these two quantities—the emitted energy divided by the excitation energy—is a measure of the efficiency with which a given laser system can operate. The situation in which every particle that is excited to the upper laser level contributes one photon of laser radiation is of course ideal; it assumes that other mechanisms, such as transitions to other lower energy levels, are negligible for de-exciting a particle in the upper laser level. Thus the ratio of the energy of the emitted photon to the energy of excitation is actually the absolute maximum efficiency (or, as it is sometimes called, the quantum efficiency) of the laser system.

In practice the efficiency of an operating gas laser is considerably lower than its quantum efficiency, since no perfect means exist for selectively exciting the gas particles from the ground state to the upper laser level. Take the case of excitation by means of a collision between an atom and an energetic electron in a gas discharge. The electron must have a certain energy to excite the atom to the upper laser level. Unfortunately in a gas discharge the electrons do not all have the same kinetic energy; instead they are distributed continuously over a wide range of kinetic energies. Hence one cannot help but excite atoms not only to the upper laser level but also to other levels (either higher or lower than the upper laser level), from which they would not contribute to the laser output. The result is that only a fraction of the input electric power needed to produce the discharge is effective in exciting the atoms to the upper laser level. If we define the working efficiency of a laser as the ratio of the output power of the laser beam to the input power of the electric discharge, then the working efficiency will always be much lower than the quantum efficiency. The closer a laser approaches the ideal system in terms of the selectivity of the excitation mechanism, the closer the working efficiency will approach the quantum efficiency. Or, in a somewhat different perspective, a high quantum efficiency, combined with a selective excitation mechanism, is the prescription for obtaining a high working efficiency in a practical laser.

The first gas laser was operated at the Bell Telephone Laboratories in 1961. It operated on a transition between two

OXYGEN   CARBON   OXYGEN
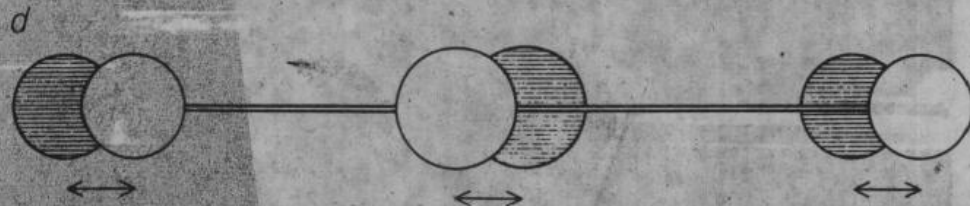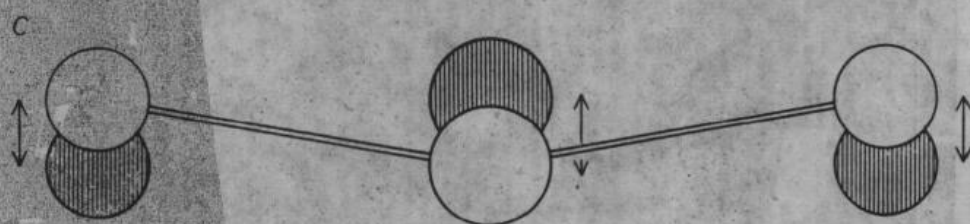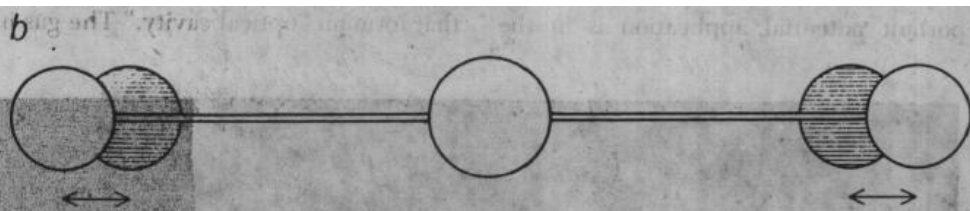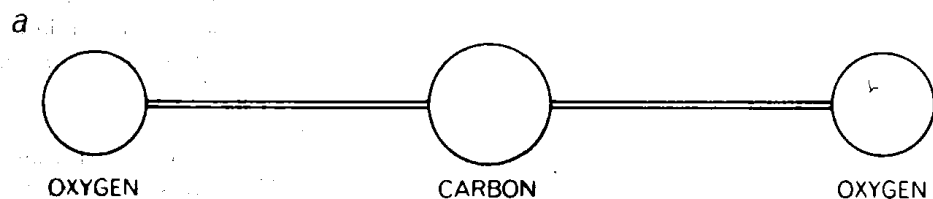
CARBON DIOXIDE MOLECULE (a) is linear and symmetric in configuration and has three degrees of vibrational freedom. In the symmetric stretch mode (b) the atoms of the molecule vibrate along the internuclear axis in a symmetric manner. In the bending mode (c) the oscillation of the atoms is perpendicular to the internuclear axis. In the asymmetric stretch mode (d) the atoms vibrate along the internuclear axis in an asymmetric manner. The vibrational state of the molecule is accordingly described by three quantum numbers, v_1, v_2 and v_3, and is usually written in the form $(v_1 v_2 v_3)$, where v_1 describes the number of vibrational quanta in the symmetric stretch mode, v_2 the number of vibrational quanta in the bending mode and v_3 the number of vibrational quanta in the asymmetric stretch mode.

excited states of atomic neon and produced a strong laser oscillation at a wavelength of 1.15 microns. Laser action has since been obtained using almost all the elements and covering the wavelength range from 2,000 angstrom units (.2 micron) in the ultraviolet region of the spectrum to 133 microns in the infrared region.

The energy-level spectra of molecular gases are considerably more complicated than those of atomic gases. In addition to the familiar electronic energy levels, a molecule can also have energy levels arising from the vibrational motion and the rotational motion of the molecule [see illustration at right]. Thus for a given electronic configuration of a diatomic (two-atom) molecule, say, there are several almost equally spaced vibrational energy levels, and for each of the vibrational levels there are a number of rotational levels. The spacings of the electronic energy levels for molecules are comparable to those for atoms, but the vibrational and rotational spacings are typically smaller by factors of 20 and 500. As a result the energy-level scheme of a molecular gas is extremely complicated.

The first molecular-gas laser oscillation was obtained from electronic transitions of a number of diatomic gases. Obviously, however, one can also have transitions between two different vibrational levels of the same electronic level of the molecule. Such transitions in turn actually occur between two rotational levels belonging to the two different vibrational levels. Moreover, because of the increasing spacing of the rotational levels in a vibrational level and because of a quantum-mechanical selection rule that in the simplest case allows only those transitions involving a change in the rotational angular momentum equal to $\pm h/2\pi$ (h is Planck's constant), such transitions between two vibrational levels result in a vibrational-rotational band [see illustration on next page]. The center of the band corresponds to the spacing between the vibrational levels in the absence of any rotational energy; the transitions on the long-wavelength side correspond to a change of $+h/2\pi$ in angular momentum and are called the P-branch transitions, whereas those on the short-wavelength side involve a change of $-h/2\pi$ in angular momentum and are called the R-branch transitions. As the illustration shows, the P-branch and R-branch transitions are almost equally spaced. These vibrational-rotational transitions, which usually result in infrared emission, are the basis of all the current breed of high-power molecular-gas lasers.
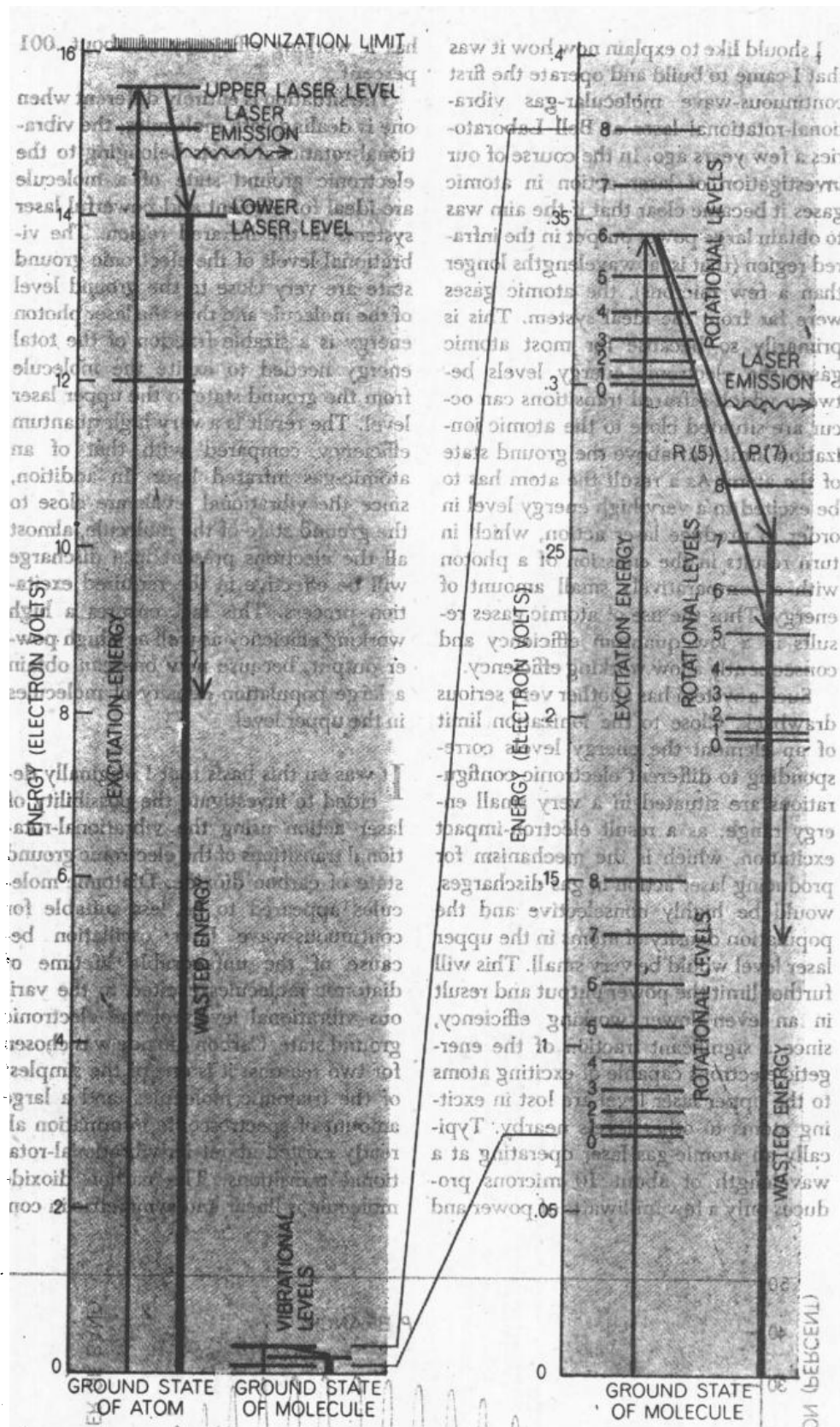
ENERGY-LEVEL DIAGRAMS of an atom and a molecule are compared. In an atom the electronic energy levels between which infrared transitions can occur are situated near the atomic ionization limit—far above the ground state of the atom. As a result the atom has to be excited to a very high energy in order to produce laser action, which in turn results in the emission of a photon with a comparatively small amount of energy. Thus the use of atomic gases wastes a great deal of energy and results in a low quantum efficiency. In a molecule, on the other hand, the vibrational levels of the electronic ground state are very close to the ground level of the molecule; hence the photon energy is a sizable fraction of the total energy needed to excite the molecule from the ground state to the upper laser level. This results in a much higher quantum efficiency. The enlargement at right shows that the vibrational levels of the molecule's electronic ground state are in turn composed of a number of rotational energy levels due to the rotation of the molecule. The number with each level indicates that level's rotational angular momentum in units of $h/2\pi$. Two of the allowed infrared transitions between the rotational levels belonging to two different vibrational levels are indicated.

I should like to explain now how it was that I came to build and operate the first continuous-wave molecular-gas vibrational-rotational laser at Bell Laboratories a few years ago. In the course of our investigation of laser action in atomic gases it became clear that if the aim was to obtain large power output in the infrared region (that is, at wavelengths longer than a few microns), the atomic gases were far from the ideal system. This is primarily so because for most atomic gases the electronic energy levels between which infrared transitions can occur are situated close to the atomic ionization limit—far above the ground state of the atom. As a result the atom has to be excited to a very high energy level in order to produce laser action, which in turn results in the emission of a photon with a comparatively small amount of energy. Thus the use of atomic gases results in a low quantum efficiency and consequently a low working efficiency.

Such a system has another very serious drawback. Close to the ionization limit of an element the energy levels corresponding to different electronic configurations are situated in a very small energy range; as a result electron-impact excitation, which is the mechanism for producing laser action in gas discharges, would be highly nonselective and the population density of atoms in the upper laser level would be very small. This will further limit the power output and result in an even lower working efficiency, since a significant fraction of the energetic electrons capable of exciting atoms to the upper laser level are lost in exciting atoms to other levels nearby. Typically an atomic-gas laser operating at a wavelength of about 10 microns produces only a few milliwatts of power and

has a working efficiency of about .001 percent.

The situation is entirely different when one is dealing with molecules; the vibrational-rotational levels belonging to the electronic ground state of a molecule are ideal for efficient and powerful laser systems in the infrared region. The vibrational levels of the electronic ground state are very close to the ground level of the molecule and thus the laser photon energy is a sizable fraction of the total energy needed to excite the molecule from the ground state to the upper laser level. The result is a very high quantum efficiency compared with that of an atomic-gas infrared laser. In addition, since the vibrational levels are close to the ground state of the molecule, almost all the electrons present in a discharge will be effective in the required excitation process. This fact ensures a high working efficiency as well as a high power output, because now one can obtain a large population density of molecules in the upper level.

It was on this basis that I originally decided to investigate the possibility of laser action using the vibrational-rotational transitions of the electronic ground state of carbon dioxide. Diatomic molecules appeared to be less suitable for continuous-wave laser oscillation because of the unfavorable lifetime of diatomic molecules excited to the various vibrational levels of the electronic ground state. Carbon dioxide was chosen for two reasons: it is one of the simplest of the triatomic molecules, and a large amount of spectroscopic information already existed about its vibrational-rotational transitions. The carbon dioxide molecule is linear and symmetric in con-

figuration and has three degrees of vibrational freedom [see *illustration on page 24*]. In one degree the atoms of the molecule vibrate along the internuclear axis in a symmetric manner. This mode of vibration is called the symmetric stretch mode and is denoted v_1. In another symmetric mode of vibration the oscillation of the atoms is perpendicular to the internuclear axis. This mode is called the bending mode and is denoted v_2. Finally, there is an asymmetric stretch mode of vibration along the internuclear axis; this mode is denoted v_3. By the rules of quantum mechanics the energies of the vibrations are quantized and are all different.

In the first approximation these three modes of vibration are independent of one another. As a consequence the carbon dioxide molecule can be excited to have any linear combination of the three individual modes of vibration. Therefore the vibrational state of the molecule must be described by three quantum numbers, v_1, v_2 and v_3, which represent the quanta of the v_1, v_2 and v_3 modes of vibration to which the molecule is excited. The description of a given vibrational level would accordingly take the form $(v_1 v_2 v_3)$, where v_1 describes the number of vibrational quanta in the symmetric stretch mode, v_2 the number of vibrational quanta in the symmetric bending mode and v_3 the number of vibrational quanta in the asymmetric stretch mode.

In the energy-level diagram of some of the low-lying vibrational states of carbon dioxide [see *illustration on opposite page*] the rotational substructure of each of the vibrational levels has been excluded in order to keep the diagram relatively uncluttered. The rotational levels are spaced much closer than the vibrational
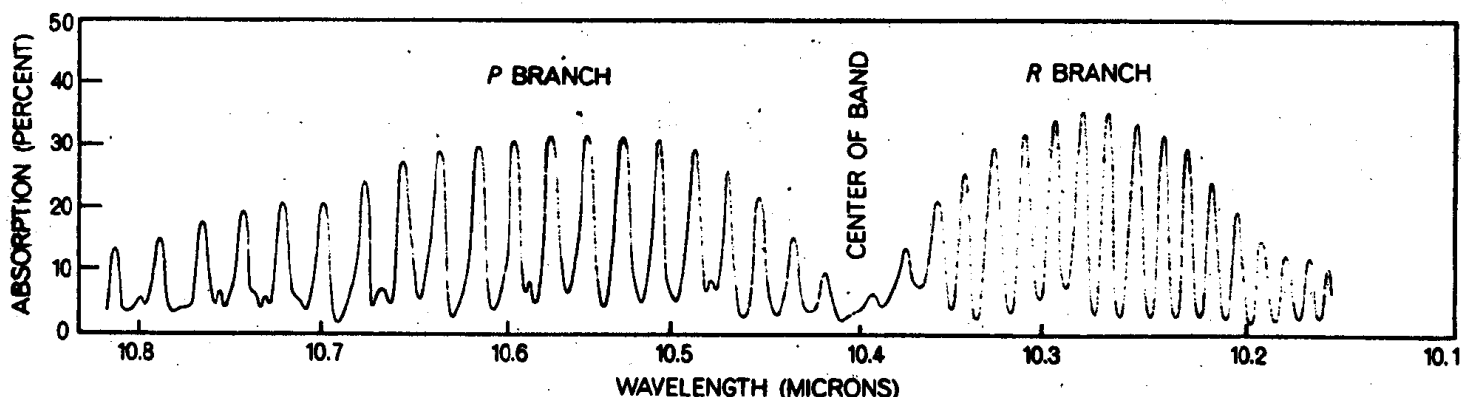
LASER OSCILLATION arising from transitions between two rotational energy levels belonging to two different vibrational levels of the same electronic level of carbon dioxide leads to the emission of infrared light at a number of different wavelengths, which form what is called a vibrational-rotational band. The curve shows the positions of the transitions as observed in the absorption spectroscopy of unexcited carbon dioxide gas. The center of the band corresponds to the spacing between the vibrational levels in the absence of any rotational energy; the transitions on the long-wavelength side correspond to a change of $+h/2\pi$ in rotational angular momentum and are called the P-branch transitions, whereas those on the short-wavelength side involve a change of $-h/2\pi$ in rotational angular momentum and are called the R-branch transitions (h is Planck's constant). The band shown here produces 10.6-micron radiation.

states. The various vibrational levels with different quanta in modes v_1, v_2 and v_3 form almost equally spaced ladders, although only the lowest states (those with only one or two quanta of vibrational energy) are shown. For a number of reasons, such as the lifetime of carbon dioxide molecules in various states and the probability of excitation by electron impact from the ground state, the level designated 001 is suitable for the upper laser level, and the 100 and 020 levels form the lower laser levels. The molecules that arrive at the lower levels decay to the ground state through radiative and collision-induced transitions to the lower 010 level, which in turn decays to the ground state. The 001 → 100 vibrational-rotational transitions produce infrared radiation near 10.6 microns, and the 001 → 020 transitions produce infrared radiation near 9.6 microns. Accordingly the quantum efficiency of a 001 → 100 laser would be nearly 40 percent, whereas that of a 001 → 020 laser would be about 45 percent. It is this high quantum efficiency and the possibility of selective excitation to levels that are close to the ground level that originally made the system attractive to investigate and that has made it possible for us to reach practical efficiencies on the order of 20 to 30 percent.

In our earliest experiments the laser tube was filled with pure carbon dioxide at a pressure of about one torr (one millimeter of mercury). The electric discharge was produced by applying a high-voltage direct current across a section of the tube. In such a discharge a large number of collisions occur between energetic electrons and the carbon dioxide molecules. A few of the most energetic electrons cause the carbon dioxide molecules to dissociate, that is, to break up into carbon and oxygen atoms. The threshold for this process, however, is quite high, and the number of electrons possessing this large amount of kinetic energy is very small. The lower-energy electrons, which far outnumber the high-energy electrons, cause the carbon dioxide molecules to be excited to various vibrational levels. As it happens, the electrons preferentially excite the carbon dioxide molecules to the $00v_3$ levels, that is, to the almost equally spaced levels of the v_3 ladder.

It should be remembered that the upper level for the laser oscillation at 10.6 microns is the one with v_3 equal to 1. Does this mean that the carbon dioxide molecules that are excited to the higher states of $00v_3$ (those with v_3
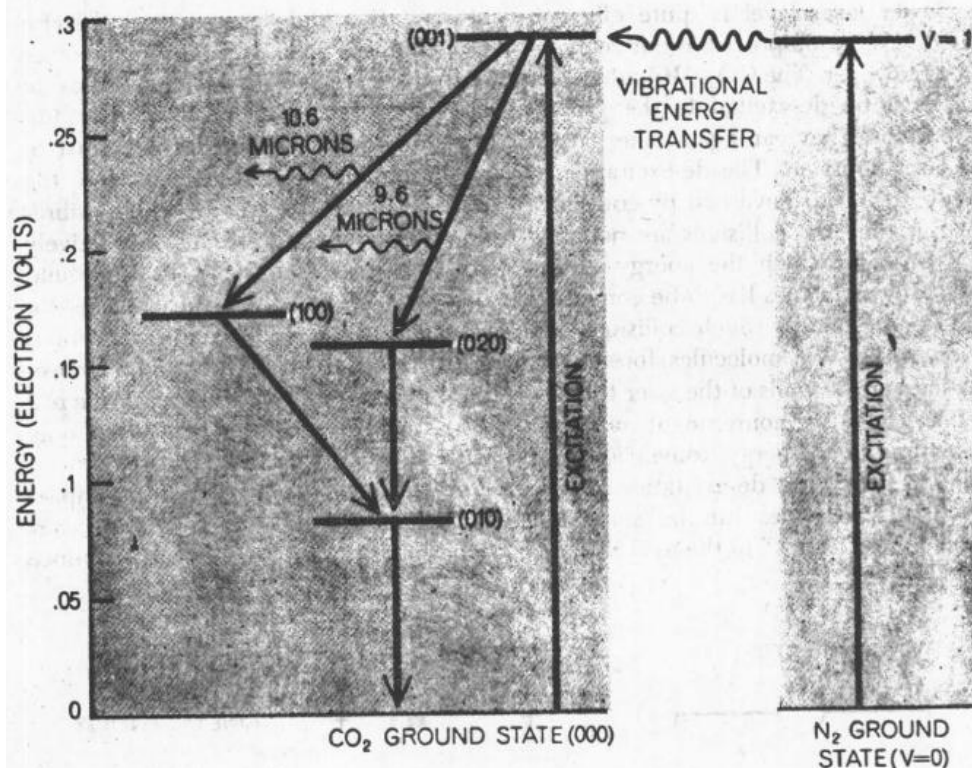
ADDITION OF NITROGEN GAS to a carbon dioxide laser results in the selective excitation of the carbon dioxide molecules to the upper laser level. Since nitrogen is a diatomic molecule it has only one degree of vibrational freedom; hence one vibrational quantum number (v) completely describes its vibrational energy levels. Nitrogen molecules can be efficiently excited from the $v = 0$ level to the $v = 1$ level by electron impact in a low-pressure nitrogen discharge. Since the energy of excitation of the $N_2(v = 1)$ molecule nearly equals the energy of excitation of the CO_2 (001) molecule, an efficient transfer of vibrational energy takes place from the nitrogen to the carbon dioxide in collisions between $N_2(v = 1)$ molecules and CO_2(000) molecules. In such a collision the nitrogen molecule returns from the $v = 1$ level to its ground state by losing one quantum of its vibrational energy, thereby exciting the carbon dioxide molecule from its ground state to the 001 level. The carbon dioxide molecule can then radiatively decay to either the 100 level or the 020 level, in the process emitting infrared light at 10.6 or 9.6 microns respectively.

greater than 1) will not contribute to laser action, thereby reducing the efficiency and power output of the system? In reality this does not happen because the $00v_3$ levels are almost equally spaced, and as a consequence a collision between a $CO_2(00v_3)$ molecule and a $CO_2(000)$ molecule results in an efficient transfer of vibrational energy from the excited molecule to the unexcited molecule. The $CO_2(00v_3)$ molecule loses one quantum of v_3 vibrational energy and becomes a $CO_2(00v_3 - 1)$ molecule, while the $CO_2(000)$ molecule gains that quantum of energy and becomes a $CO_2(001)$ molecule, or in other words a molecule in the upper laser level [see upper illustration on next two pages].

This process is resonant in the sense that there is a redistribution of the energy of the excited molecule without any loss of the total internal energy by its conversion into kinetic, or thermal, energy. This means that the efficiency of converting the $CO_2(00v_3)$ molecules into $CO_2(001)$ molecules with no loss of energy is very high. Therefore in practice

one should be able to excite carbon dioxide molecules to the required upper laser level quite efficiently by electron impact in a gas discharge.

The CO_2(001) molecules can now, for example, emit a laser photon at 10.6 microns and go to the 100 level, from which they have to be returned to ground state before the molecule can be utilized again for producing a laser photon. The molecules at the lower laser level are de-excited essentially through collisions with other molecules. Again the possibility of resonant vibrational energy transfer plays an important role. The lower laser level has nearly twice the energy required to excite the carbon dioxide molecule to the 010 vibrational level. As a result a collision that involves a CO_2(100) or CO_2(020) molecule with a CO_2(000) molecule will efficiently redistribute the vibrational energy between the two molecules by exciting both of them to the CO_2(010) level [see lower illustration on next two pages].

Because of the resonant nature of this collision the vibrational de-excitation of

the lower laser level is quite efficient. The de-excitation process is not yet complete, however. The $CO_2(010)$ molecules still must be de-excited to the ground state before they can again take part in the laser emission. The de-excitation of $CO_2(010)$ is also governed by collisions, but this time the collisions are nonresonant ones in which the energy of the $CO_2(010)$ molecules has to be converted into kinetic energy. Such collisions can involve other CO_2 molecules, foreign gas particles or the walls of the laser tube.

Because of the nonresonant nature of this vibrational energy conversion into kinetic energy the de-excitation of the $CO_2(010)$ molecules can be slow and cause a "bottleneck" in the overall cycle

of excitation and de-excitation, thereby reducing the efficiency and the power output. Even for the pure carbon dioxide laser I originally tested, the de-excitation mechanism was sufficiently fast to allow strong laser oscillation on the $001 \rightarrow 100$ and the $001 \rightarrow 020$ vibrational-rotational transitions respectively at 10.6 and 9.6 microns. It was found that because of their larger emission probability the 10.6-micron transitions are stronger than the 9.6-micron transitions by about a factor of 10. For the rest of the article we shall be concerned only with these 10.6-micron transitions.

It is quite clear that electron-impact excitation that occurs in a pure carbon dioxide discharge cannot produce

the highly selective excitation of the molecules to the upper laser level that is required for obtaining a practical efficiency approaching the quantum efficiency of the system. The reason is that the electrons can also excite the carbon dioxide molecules to levels other than the $00v_3$ level, causing a reduction in efficiency as well as power output. For high efficiency what is needed is some form of selective excitation of the carbon dioxide molecules to the upper laser level. Such a selective excitation occurs when nitrogen gas is added to the carbon dioxide laser.

The usefulness of nitrogen can be explained by referring to the energy-level diagram of the low-lying vibrational
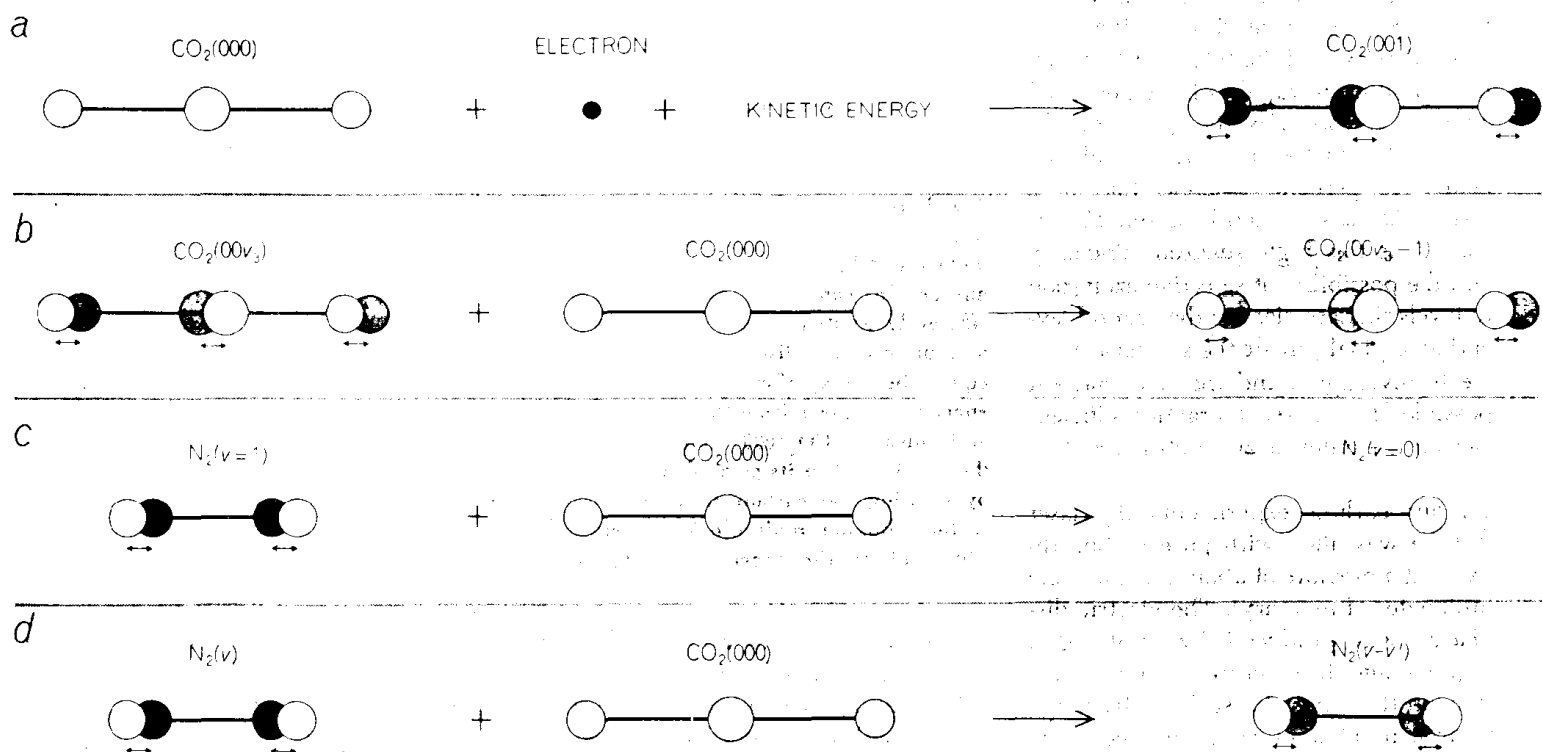
EXCITATION MECHANISMS capable of raising carbon dioxide molecules to the upper laser level (in this case the 001 level) are shown. In an electric discharge the collision of an unexcited, or 000, molecule with an energetic electron can raise the carbon dioxide molecule to the 001 level directly (*a*). Alternatively such a collision can excite the 000 molecule to a $00v_3$ level, where the v_3, or asymmetric stretch, mode has more than one quantum of vibrational energy; in this case subsequent collisions with unexcited molecules result in the transfer of single quanta of vibrational energy to the unexcited molecules, raising them to the 001 level (*b*). In a carbon dioxide-nitrogen laser collisions between vibrationally excited nitrogen molecules and unexcited carbon dioxide molecules
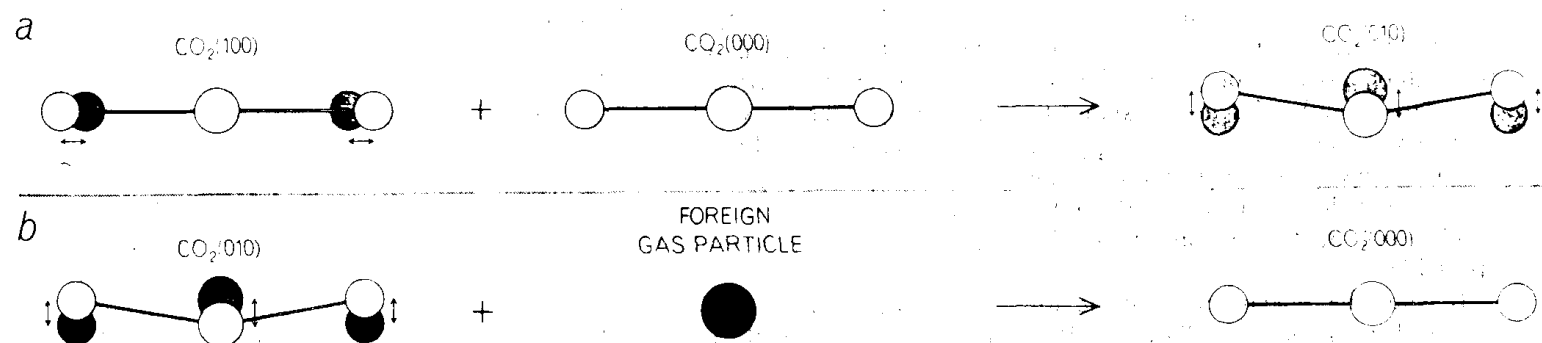
DE-EXCITATION MECHANISMS capable of "depopulating" the lower vibrational levels of carbon dioxide can result in an increased laser power. Two such de-exciting collisions are shown here. In *a* the collision of an excited 100 molecule with an unexcited 000 molecule leaves both molecules at the 010 level. In *b* a molecule at the 010 level can in turn collide with foreign gas particles

levels of the electronic ground state of molecular nitrogen [*see illustration on page 27*]. Since nitrogen is a diatomic molecule, it has only one degree of vibrational freedom; its vibrational energy levels are described by quanta of energy arising from vibrations along the internuclear axis alone. Accordingly one vibrational quantum number completely describes the vibrational levels of the nitrogen molecule. Because nitrogen is a homonuclear, diatomic molecule, molecular nitrogen excited to various vibrational levels of the electronic ground state cannot decay radiatively or through collisions, and it is therefore extremely long-lived.

Nitrogen molecules are efficiently ex-

ELECTRON

$+$ ●

$CO_2(001)$

$+$

$CO_2(001)$

$+$

$CO_2(00v')$

$+$

can raise the carbon dioxide molecules to the 001 level by transferring one quantum of vibrational energy from the nitrogen molecule, which can have initially either one (c) or more than one (d) quanta of vibrational energy going into the collision.

$CO_2(010)$

$+$

FOREIGN GAS PARTICLE

$+$ ● $+$ KINETIC ENERGY

(or with the walls of the laser tube) and thereby return to the ground state, where it is available to be excited once again.

cited from the $v = 0$ level to various higher vibrational levels primarily by electron impact; they can also be excited by cascading from higher electronic states and by the recombination of dissociated nitrogen atoms. In a low-pressure nitrogen discharge one can excite approximately 30 percent of the nitrogen molecules to the $v = 1$ level. Since the energy of excitation of the $N_2(v = 1)$ molecule nearly equals the energy of excitation of the $CO_2(001)$ molecule, one would expect an efficient transfer of vibrational energy from the nitrogen to the carbon dioxide in collisions between the $N_2(v = 1)$ molecule and the $CO_2(000)$ molecule. In such a collision the nitrogen molecule returns from the $v = 1$ level to its ground state by losing one quantum of its vibrational energy and the carbon dioxide molecule is excited from its ground state to the 001 level. Because of the resonant nature of this collision process, the selective excitation of carbon dioxide molecules to the upper laser level should be very efficient.

Furthermore, the higher vibrational levels of the nitrogen molecule are nearly equally spaced, as are the $CO_2(00v_3)$ levels. Hence, in collisions involving $N_2(v)$ and $CO_2(000)$ molecules efficient vibrational energy transfer can take place in which the excited $N_2(v)$ molecule loses v' quanta of vibrational energy and is de-excited to the $N_2(v - v')$ level while the $CO_2(000)$ molecule gains the v' quanta of vibrational energy and is selectively excited to the $CO_2(00v_3 = v')$ level. Since the spacing between the energy levels of the $N_2(v)$ ladder and the $CO_2(00v_3)$ ladder is nearly equal, these collisions involve resonant vibrational energy transfer and the process is very efficient. The $CO_2(00v_3 = v')$ molecules are then converted into $CO_2(001)$ molecules (that is, into the upper-laser-level molecules) through the resonant collisions discussed earlier. In the end one has efficient selective excitation of carbon dioxide molecules to the upper laser level, and one should expect a significant increase in efficiency and power output from a carbon dioxide–nitrogen laser as compared with a pure carbon dioxide laser.

The first experiments to verify this hypothesis were carried out in the system shown on page 32. The gases in the system are continuously flowing. There is no electric discharge in the interaction region, where the laser action is expected to take place. Nitrogen enters through one port and passes through the excitation region, where an electric dis-

charge is produced by means of an oscillating electric field or a high-voltage direct current. The nitrogen molecules are excited to various vibrational levels of the electronic ground state as the nitrogen passes through the discharge region.

Since this is a continuous-flow system, the nitrogen molecules that have been subjected to the discharge are pumped into the interaction region in times that are short compared with the average lifetime of a vibrationally excited nitrogen molecule. Hence the nitrogen gas entering the interaction region will contain a significant fraction of nitrogen molecules that are still excited and that remain in the vibrationally excited levels of the electronic ground state. Carbon dioxide entering through another port mixes with the nitrogen coming through its port. As described above, vibrational energy transfer from nitrogen to carbon dioxide results because of the collisions involving the vibrationally excited nitrogen molecules and the ground-state carbon dioxide molecules. Carbon dioxide molecules are thus selectively excited to the upper laser level. Notice that there is no other form of excitation in the interaction region for exciting the carbon dioxide.

Strong laser oscillation can be obtained in this system on the vibrational-rotational transitions of carbon dioxide even though no discharge is present in the interaction region. After the carbon dioxide molecules have contributed to laser oscillation the continuous-flow system pumps out all the de-excited molecules, and fresh nitrogen discharge products and carbon dioxide enter to continue the laser oscillation. The strength of the laser oscillation proved the effectiveness of using vibrationally excited nitrogen molecules for selective excitation of carbon dioxide molecules to the upper laser level. By mixing nitrogen and carbon dioxide together in a laser tube, with the discharge in the laser region, conversion efficiencies as high as 5 percent have been demonstrated.

Increasing de-excitation of the lower laser levels by removing the "bottleneck" at the 010 level of carbon dioxide can also result in increased power output as well as higher efficiency from the carbon dioxide laser. Earlier I mentioned that de-excitation of the $CO_2(010)$ molecules takes place by conversion of the energy of the $CO_2(010)$ molecule into kinetic energy during a collision with another particle. The rate at which this de-excitation process proceeds depends on the nature of the other particle. For
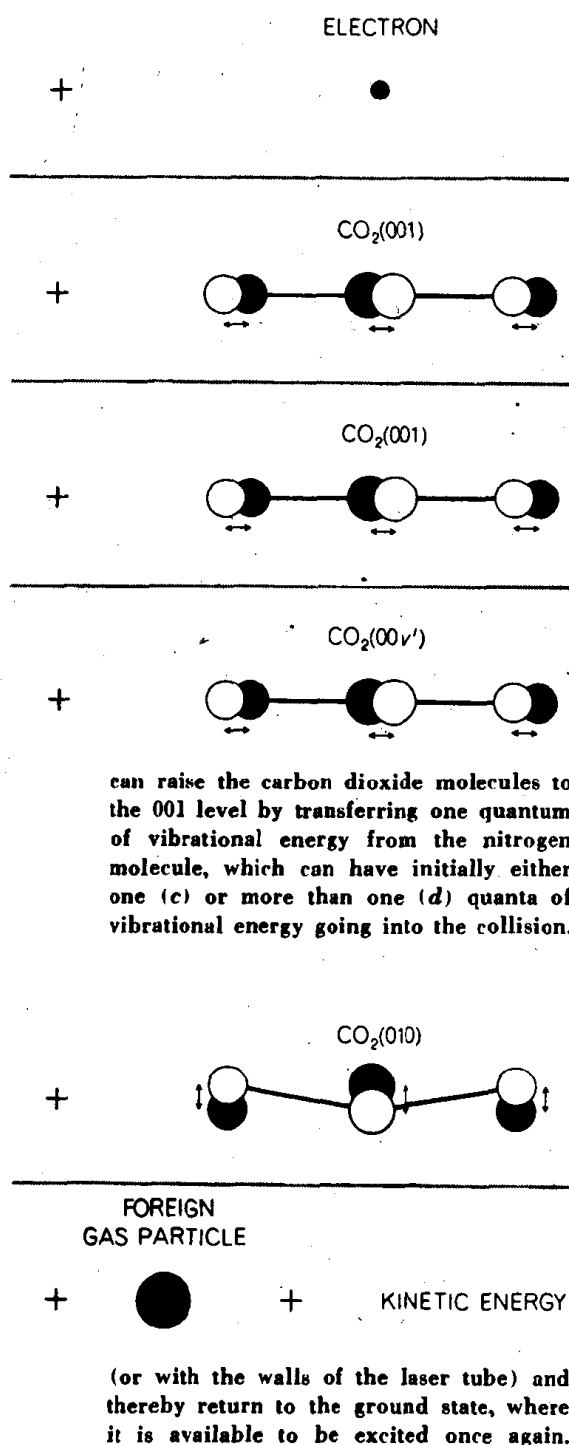
example, carbon dioxide itself has about 100 de-exciting collisions per second at a pressure of one torr, whereas helium atoms have some 4,000 (and water molecules some 100,000) de-exciting transitions per second at the same pressure. Thus we have another method for increasing the power output and efficiency of the nitrogen–carbon dioxide laser system.

It was found that in order to obtain an extremely high continuous power output at a high efficiency it is necessary to use additional gases in the discharge tube. Gases such as oxygen, water vapor, hydrogen and helium give rise to increased power output. The increase is understood in terms of two effects: (1) the increased rate of de-excitation of the lower vibrational levels of the carbon

dioxide molecules and (2) the increase in the rate at which carbon dioxide molecules are excited to the 001 level, either directly by electron-impact processes or indirectly by increasing the excitation rate of the vibrationally excited nitrogen molecules. Both processes for increasing the excitation of the carbon dioxide molecules to the upper laser level are likely if the density of electrons in the discharge is increased and also if the energy distribution of the electrons changes to make it more favorable for exciting the carbon dioxide molecules to the 001 level directly and for faster production of $N_2(v)$ molecules.

Helium seems to be important on both counts and is the most widely used third gas. Water vapor and hydrogen are useful only in terms of the first effect. Car-

bon monoxide seems to be important from both the excitation and the de-excitation points of view. Using a carbon dioxide pressure of three torr, a nitrogen pressure of three torr and a helium pressure of about 20 torr, a continuous-wave power of some 80 watts per meter of discharge length has been obtained at a wavelength of 10.6 microns. The working efficiency in this case is in excess of 20 percent.

At present most of the high-power carbon dioxide lasers have the gases flowing at a slow rate through the laser tube. Some of our early experiments at the Bell Laboratories and more recent experiments at the Philips Research Laboratory in the Netherlands have shown, however, that it is possible to make sealed-off carbon dioxide lasers if sufficient care is taken in preparing the tube and if the proper gas mixtures are used. These lasers are capable of producing just as much power output as the flowing-gas systems, and the efficiency is quite comparable.

Typical carbon dioxide lasers are about two meters long and can produce continuous-wave laser power of about 150 watts. There is nothing to prevent one from making a very long laser in order to obtain much higher power outputs, since the power output increases linearly with length. In fact, workers at the Raytheon Company have constructed a "folded" carbon dioxide laser that is 600 feet long. This laser has produced continuous-wave power as high as 8.8 kilowatts. The power output of the laser has thus finally caught up with the fantasies of science fiction, and the thermal effects of such output are certainly awesome.

The experimental setup shown in the illustration on page 32 is useful in studying laser action in molecular gases that are unstable, that is, gases that dissociate easily under direct-discharge excitation and/or require an extremely selective excitation for continuous laser operation (for example the diatomic gases). The advantage here is that there is no discharge in the laser region and thus the only levels of the active gas that can be excited are those whose energy coincides with the vibrationally excited nitrogen molecules. In this way continuous laser oscillation was achieved on vibrational-rotational transitions of carbon monoxide (CO) at five to six microns, in nitrous oxide (N_2O) on the $001 \rightarrow 100$ transitions near 10.8 microns and in carbon disulfide (CS_2) on the $021 \rightarrow 120$ transitions near 11 microns.


ENERGY →

RELATIVE POPULATION DENSITY ⟶
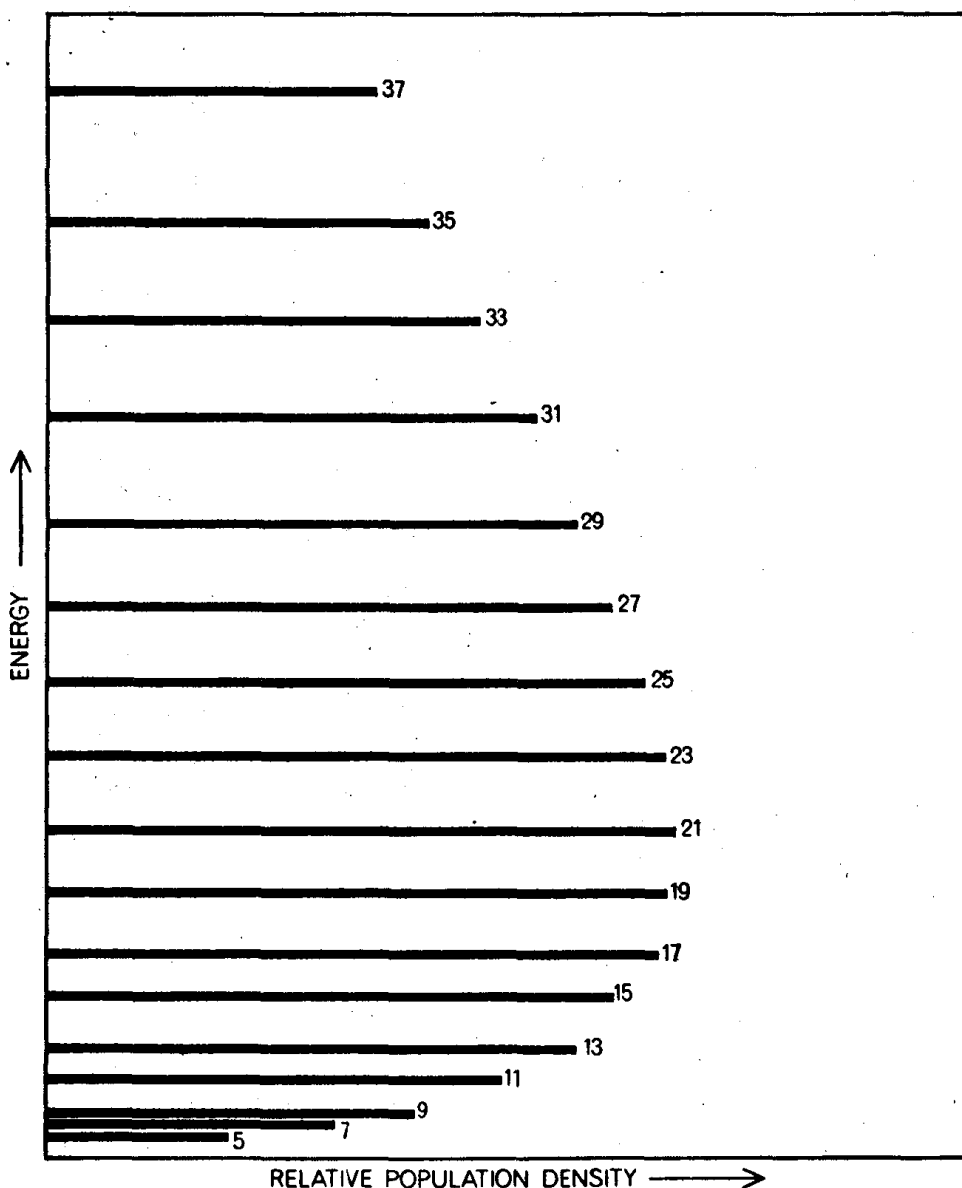
BOLTZMANN DISTRIBUTION of the population densities of the rotational energy levels of the 001 vibrational level of carbon dioxide results from the fact that during its lifetime in a given vibrational level a molecule undergoes a large number of rotational thermalising collisions, hopping around from one rotational level to another about 10 million times per second. The horizontal scale shows the population densities of the rotational levels at about 400 degrees Kelvin. Vertical energy scale shows the position of each rotational level.

The carbon monoxide laser is particularly interesting from the spectroscopic point of view. We are now able to observe transitions among vibrational levels as high as $v = 25$ in the electronic ground state—transitions that have never been observed before. Nitrous oxide, which is similar in its vibrational modes to carbon dioxide, has the capability of generating high continuous-wave power at an efficiency comparable to that of the carbon dioxide system; it has not, however, been investigated in detail as yet. In any case, the technique of selective excitation for obtaining high power output and high efficiency appears to be generally applicable.

Other techniques for obtaining vibrational excitations in molecules include chemical reactions, heating of gases by flames or burners and optical excitation by matching optical radiation obtained from discharge or flames. These other means have not yet been exploited to any great extent but they do hold promise. The fact that the discharge excitation of the carbon dioxide laser is capable of a conversion efficiency of more than 20 percent, however, poses a formidable challenge to new techniques in terms of practical applications.

So far I have described the mechanisms of excitation and de-excitation that result in the extremely high power output of carbon dioxide lasers, but I have said nothing about the spectrum of the power output. As I have mentioned, the transitions between two vibrational levels occur in the form of a band consisting of P and R branches, because of the closely spaced rotational levels of both the upper and the lower laser vibrational levels. Does this imply that the power output from the carbon dioxide laser occurs at a number of frequencies corresponding to the discrete P-branch and R-branch transitions simultaneously? If the power output were to occur at a number of frequencies, the laser beam would not be truly monochromatic and its usefulness in areas such as communications would be limited. In reality the power output from a high-power carbon dioxide laser can usually be made to occur on a single P-branch transition, usually $P(20)$ at 10.5915 microns, without any trouble, in spite of the fact that the $001 \rightarrow 100$ vibrational band contains a number of possible P-branch and R-branch transitions. This is accomplished by exploiting some rather subtle "competition effects" that take place between the P-branch and the R-branch transitions—a stratagem that in-
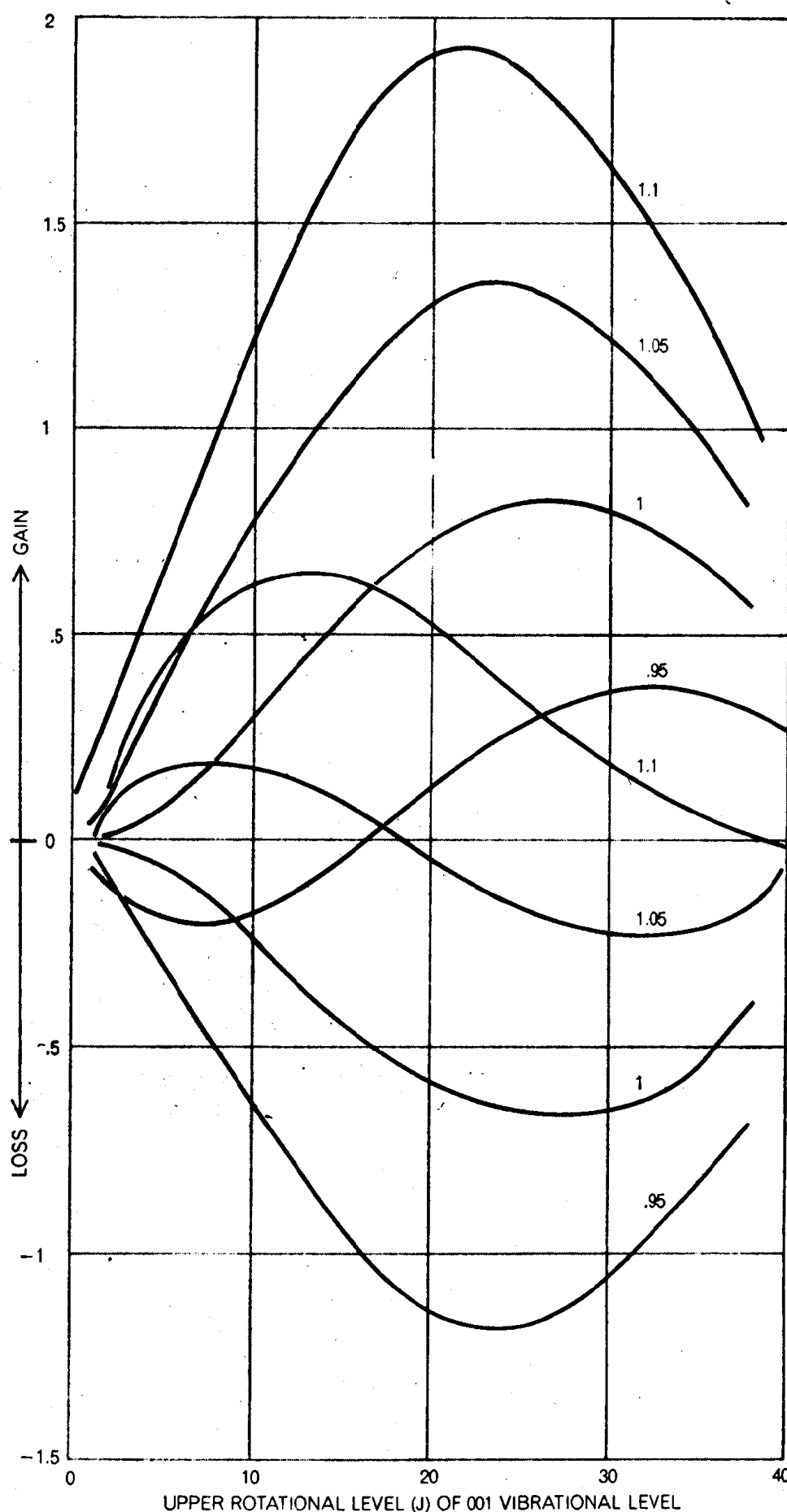
COMPETITION EFFECTS among the various possible vibrational-rotational laser transitions of carbon dioxide usually result in one P-branch transition dominating. This set of curves shows the amount of gain (or loss) on a number of P-branch transitions (*color*) and R-branch transitions (*black*) of a given vibrational band. The number associated with each curve gives the ratio of the total population density in the upper, or 001, vibrational laser level to the total population density in the lower, or 100, vibrational laser level.
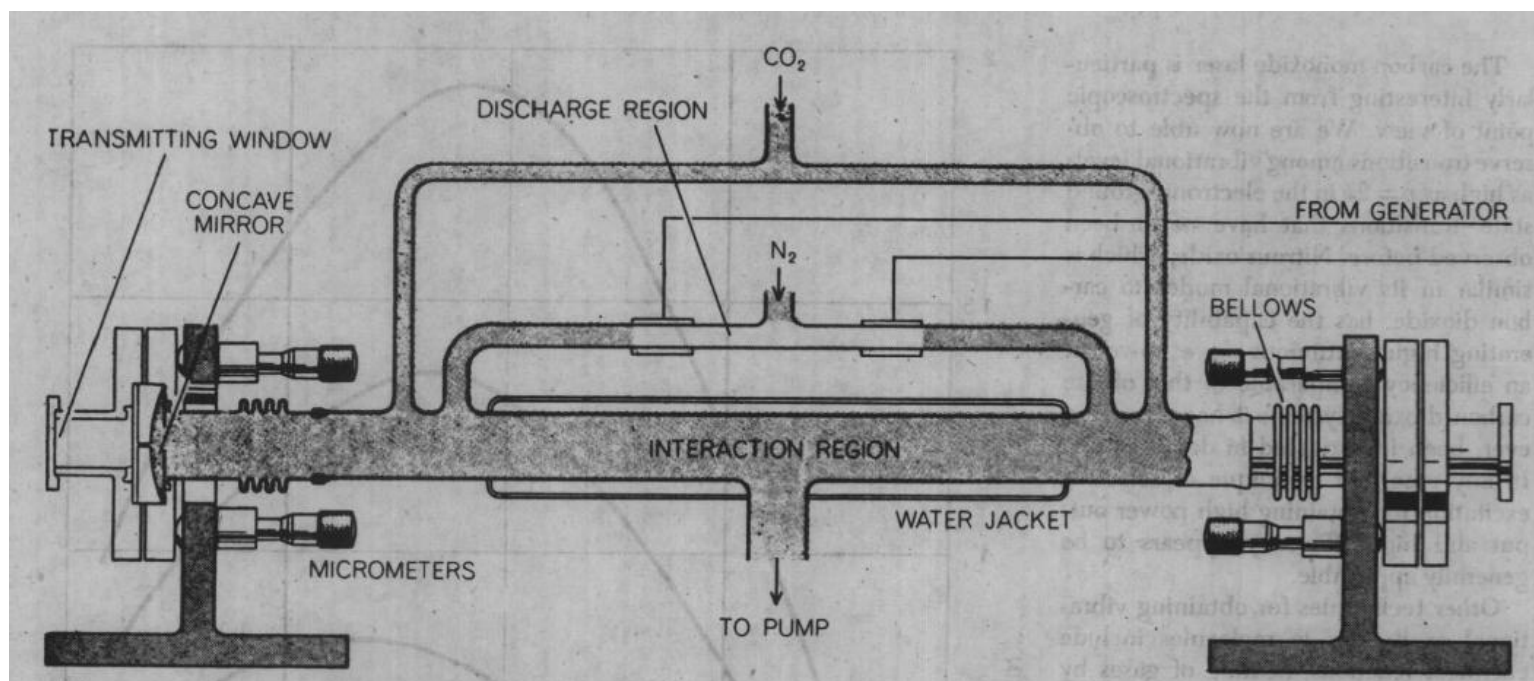
CONTINUOUS-FLOW SYSTEM was used by the author to verify the hypothesis that a carbon dioxide–nitrogen laser would be more efficient than a pure carbon dioxide laser. Strong laser oscillation was obtained in this system on the vibrational-rotational transitions of carbon dioxide even though no electric discharge was present in the interaction region, thereby proving the effectiveness of using vibrationally excited nitrogen molecules for selective excitation of carbon dioxide molecules to the upper laser level.

creases the usefulness of the carbon dioxide laser tremendously.

It is important to observe that the energy spacings between the various vibrational levels are usually much greater than the kinetic energy of the molecules (which is on the order of .025 electron volt at room temperature). The spacing of the rotational energy levels, on the other hand, is smaller than the kinetic energy. Thus the population density of a particular rotational level in a given vibrational level is not independent of the population density of other rotational levels, since every single collision can result in an exchange of energy equal to the rotational-level spacings. As a result the molecule can jump around from one rotational level to another very frequently. The frequency of this hopping around (also known as the rotational thermalization rate) is in excess of 10 million hops per second for the gas pressures at which the lasers operate. Since the spacing of the vibrational energy levels is much larger than the kinetic energy of the molecules, however, the vibrational thermalization rate is very small: about 1,000 per second. The vibrational-level lifetime, including radiative and collisional relaxation, is about a millisecond whereas the rotational thermalization time is considerably shorter: about 10^{-7} second. This implies that during its lifetime in a vibrational level a molecule undergoes a very large number of rotational thermalizing collisions. This gives rise to a Boltzmann distribution of the molecules among the

various rotational levels of a vibrational level [see illustration on page 30].

Under the above conditions governing the population densities of the rotational levels one can calculate the amount of gain (or loss) on the various P-branch and R-branch transitions of a given vibrational band [see illustration on preceding page]. From such a calculated set of curves the following useful conclusions can be easily reached: (1) Some P-branch transitions show gain even when the total vibrational population density in the lower laser level exceeds that in the upper level. This situation is called "partial inversion," since the R-branch transitions do not show gain. (2) When the total vibrational population density in the upper laser level exceeds that in the lower laser level, both P-branch and R-branch transitions show gain. This is called "complete inversion." (3) Even for the case of complete inversion, an R-branch transition always has less gain than a P-branch transition starting from the same upper rotational level.

Let us now see what practical effects one can observe from the above conclusions. While it is true that gain occurs on a large number of transitions simultaneously, the existence of a Boltzmann distribution requires that the change of population density of one rotational level affect the population density of all rotational levels in order to maintain the Boltzmann distribution. The rotational transition with the highest gain—in this case transition $P(22)$—will start oscillating first. This will be the strongest P-

branch transition, since the R-branch transitions have lower gain. When this occurs, the rate at which molecules are removed from the $J = 21$ rotational level increases because of the stimulated emission on the $P(22)$ transition. But the requirement of the Boltzmann distribution will result in a transfer of molecules from other rotational levels to the $J = 21$ level and the population density of all the rotational levels decreases even though the laser oscillation on $P(22)$ drains the molecules from the $J = 21$ level. This results in a very strong competition among the possible laser transitions and usually one P-branch transition dominates.

As a result of our newfound mastery of these competition effects, the power output from a high-power continuous-wave carbon dioxide laser can be made to occur on a single rotational transition of the $001 \rightarrow 100$ band, thereby ensuring that the high-power output is both extremely coherent and extremely monochromatic.

Oscillation is possible on the weaker P-branch or R-branch transitions provided there is sufficient gain and a wavelength-selecting element such as a grating or a prism is introduced in the laser cavity to prevent the stronger transition from oscillating. Because of the strong competition, one obtains nearly the same amount of power output on any transition that one selects for oscillation, using the wavelength-selecting device.

In addition, because of the long lifetime of the vibrational levels responsible

for laser oscillation in carbon dioxide, we can store energy in the discharge medium for about a millisecond by blocking the path of the laser beam within the resonator and thereby preventing the laser oscillation. If the block is suddenly removed, then the output from the laser occurs in the form of a sharp pulse whose peak power is usually 1,000 times larger than the average continuous-wave power obtainable from this laser. This mode of operation is called Q-switching. The Q-switching is most easily accomplished by replacing one of the laser-cavity mirrors with a rotating mirror [see illustration below]. The laser operates every time the rotating mirror lines up with the opposite stationary mirror, putting out an infrared pulse at 10.6 microns. With such a Q-switching scheme a carbon dioxide laser capable of producing approximately 50 watts of continuous-wave power will produce nearly 50 kilowatts of pulsed power in bursts approximately 150 nanoseconds long and at a rate of about 400 bursts per second. Such high pulsed-power output with the coherency afforded by gas lasers is particularly useful in nondestructive physical investigations.

An ideal source of coherent radiation is one that can be continuously "tuned," that is, one whose frequency can be changed and controlled continuously. Tuning a high-power molecular laser is not strictly possible, but the number of discrete vibrational-rotational transitions that can be made to oscillate is extremely large. For example, in carbon monoxide alone there are about 200 transitions that oscillate between five and six microns; in carbon dioxide about 100 transitions with high-power output

can be made to oscillate between nine and 11 microns. In short, the high-power molecular lasers, although not continuously tunable, offer a wide range of discrete wavelengths at which one can work.

The high continuous-wave and Q-switched power made available by the carbon dioxide lasers has many applications. Focusing the coherent infrared output into an area of approximately a thousandth of a square centimeter can give an intensity of a million watts per square centimeter for a continuous-wave laser and an intensity of a billion watts per square centimeter for a Q-switched laser. Continuous-wave powers in excess of one kilowatt have obvious thermal applications such as metal cutting and welding. A strange "softening" of granite rock is also reported when the rock is irradiated with a kilowatt of power from a carbon-dioxide laser. The output from the carbon dioxide laser at 10.6 microns, although invisible to the eye, is just as devastating as that from any other powerful laser. The thermal applications of the carbon dioxide lasers command considerable industrial interest.

More important, however, are the nonthermal applications of the carbon dioxide laser. These potential applications include optical communications both on the earth and in space. The main attraction here is the low-loss optical "window" that exists between eight and 14 microns for transmission through the earth's atmosphere. The high efficiency and high power of the carbon dioxide lasers at 10.6 microns make them ideal candidates for such applications. The

carbon dioxide laser is also ideal for use in optical radar systems, again because of low-loss transmission through the atmosphere. Another possibility is the use of a carbon dioxide laser to investigate optical interactions with matter at a wavelength of 10.6 microns, since many semiconductors that are opaque in the visible portion of the spectrum are transparent at this wavelength. Still another application of the high-power carbon dioxide laser is the use of the 10.6-micron radiation as a "pump" for studying nonlinear properties of new materials with the aim of making a really tunable source of infrared radiation. In this connection my colleagues and I have performed a number of interesting experiments, which include second-harmonic generation, parametric amplification of far-infrared radiation, two-photon electron-hole pair production in semiconductors, nonlinearities arising from conduction electrons in semiconductors and Raman scattering from Landau-level electrons in semiconductors. Some of these mechanisms are strong enough to enable us to make a tunable laser oscillator in the infrared portion of the spectrum. Such a tunable laser, pumped with the fixed-frequency carbon dioxide laser, can be used as a local oscillator in an optical communication or radar system. Moreover, such a tunable infrared source would completely revolutionize infrared spectroscopy. The description of these experiments will perhaps be the subject of a future article. In conclusion, it suffices to say that the carbon dioxide lasers have already opened up avenues of physical investigation undreamed of before, and they promise many more fruitful experiments in the future.
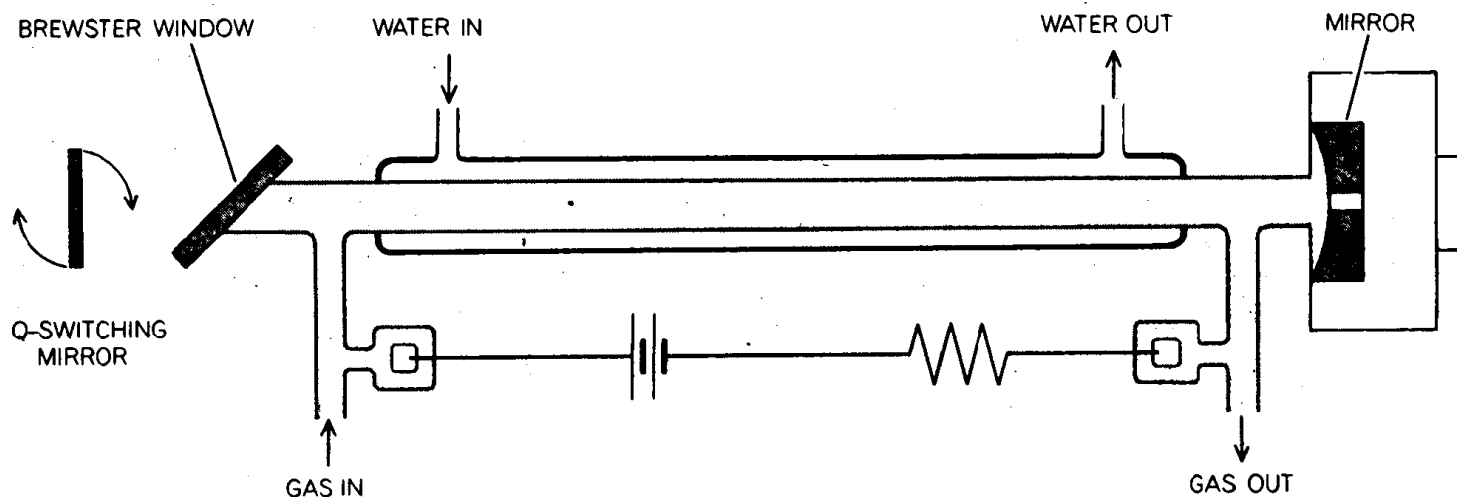
Q-SWITCHING, a technique for operating a normally continuous-wave laser in a pulsed mode, is accomplished by replacing one of the laser-cavity mirrors with a rotating mirror. The laser operates every time the rotating mirror (left) lines up with the opposite stationary mirror (right). A carbon dioxide laser capable of producing approximately 50 watts of continuous-wave power will produce nearly 50 kilowatts of Q-switched power in bursts approximately 150 nanoseconds long and at a rate of about 400 bursts per second.