Introduction to Laser Physics

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Gas Lasers

1. NEGATIVE ABSORPTION OF OPTICAL RADIATION IN GASES: QUALITATIVE DISCUSSION

In gases, as in solids, a net stimulated emission of radiation occurs only in a condition that is variously referred to as population inversion, temperature inversion, or a state of negative absorption or negative temperature. This is a nonequilibrium condition characterized by the existence of a pair of energy levels of which the higher is more densely populated than the lower. The discussion in Chapter II is equally applicable to solids and gases; therefore, without repeating what was said there, we shall proceed to discuss the phenomena that are specifically important in gases and to contrast the properties of gases that are relevant in connection with negative absorption with similar properties of solids.

The simple appearance of the helium-neon laser is misleading. The physical phenomena that take place in a gaseous laser are complicated, and laser oscillations are achieved only if these phenomena interact in a rather sophisticated manner calculated to produce population inversion. The reproduction of a gas laser requires only a moderate skill in optics and in vacuum technology; the difficult problem is to discover what transitions among the energy levels of a gas may be used for laser oscillations and what the optimum conditions for the excitation of these oscillations are.

First we note that the spectroscopic aspects of the problem are simpler in gases than in solids. The energy-level schemes of the free atoms involved are simple compared with those of the atoms embedded in a crystal lattice. The selection rules are generally observed; nonradiative transitions are less prevalent than in solids and, in fact, in gases they consist of energy conversion in collisions. The absence of absorption bands is a simplification for the theorist, but it is no help to the experimentalist bent on producing a condition of negative absorption. It means that he cannot use an ordinary black-body type of source for excitation because the gas absorbs only in isolated lines. This limits optical excitation to irradiation by high-intensity spectral sources, one of whose emission lines coincides with an absorption
line of the gas. Fortunately, there are other means of excitation in a gas that are absent in a solid: excitation by electron impact and transfer of excitation between colliding atoms. Naturally one must consider not only the processes that lead to an increase of atomic energy but also the processes by means of which an excited atom may lower its energy, since the removal of electrons from the terminal level is important for the success of the fluorescence cycle.

As the atoms excited by one means or another cascade down the energy scale, a certain stationary nonequilibrium situation will be established in which the number of atoms in each energy state remains constant. This situation requires that the rate at which the atoms arrive in a state due to all causes be equal to the rate at which they leave that state. The equations governing this process are similar to those already introduced in Section II.2. The number of atoms in each state adjusts itself to establish a balance. Those states from which escape is slow will accumulate a large number of atoms. In particular, crowding will occur in the so-called metastable states, which are higher in energy than the ground state but from which radiative transitions to lower levels are forbidden by the selection rules of quantum mechanics.

Whether population inversion is achieved depends on the rate of excitation and on the decay rates of all levels involved in the cascade process. Several processes contribute to decay of a single level: radiative processes, collisions with electrons, and collisions with other atoms including those which constitute the walls of the vessel containing the gas. In addition to these phenomena one must consider the possibility of the resonance trapping of radiation, i.e., excitation of an atom to an occupied state by radiation emitted from another atom of the gas. This phenomenon is clearly dependent on the pressure of the gas and on the geometrical configuration. It slows down the rate at which the atoms return to the ground state at the end of their cascade process.

In principle the simplest method of making a gas laser is that of optical excitation. It can be accomplished by making use of the fortuitous coincidence of the emission line of one element with a suitable absorption line of another.

The classical and best-known example of the coincidence of an absorption line of an element with a strong emission line of another is the 3888-Å line common to helium and cesium. In 1930 Boeckner demonstrated that the fluorescence of cesium can be excited by irradiation with the 3888-Å helium line, and one of the early attempts made at extending the maser art to the optical region involved the optical pumping of cesium by means of this radiation. In a series of experiments performed in 1957 Butayeva and Fabrikant [1] searched for population inversions of cesium connected with
transitions in the visible region. The results were inconclusive. The rather complicated structure of the fluorescence of cesium was studied in detail by Townes and his students at Columbia University [2]. They concluded that conditions were correct for obtaining laser oscillations at 3.20 and 7.18 μ. After overcoming serious technical difficulties connected with the confinement of the highly reactive Cs vapor at a temperature of 175°C, the irradiation with a powerful He discharge, and the alignment of the optical system, Rabinowitz, Jacobs, and Gould [3] observed laser action at the predicted frequencies. The energy levels and transitions involved in this laser action are shown in Fig. 71.

Excitation by electron collision can be most conveniently accomplished in an electric discharge. When an electric discharge takes place in a gas, ions and free electrons are formed. These current carriers are accelerated by the field that creates the discharge and they acquire a kinetic energy at the expense of the power fed into it. The motion of the ions is of little consequence; the electrons are the agents that acquire energy from the

![Fig. 71. Energy levels and transitions of cesium participating in the fluorescence cycle of the optically excited lasers.](image-url)
power source and distribute it in their encounters with atoms. In discharges that take place well below atmospheric pressure—a few millimeters of mercury or below—the average kinetic energy of the electrons usually greatly exceeds that of the atoms and ions present in the discharge. In a steady discharge within a short time of the order of a millisecond the electrons establish an equilibrium distribution among themselves. This is essentially a Maxwell-Boltzmann distribution characterized by an electron temperature $T_e$ that is proportional to the mean kinetic energy of the electrons. The average kinetic energy of the atoms is considerably lower; its value corresponds in equilibrium to a much lower temperature.

Inelastic collision in which the atom either gains or loses energy may occur between atoms and electrons, but only the amounts of energy consistent with its energy-level scheme can be transferred to or from the atom. If no other processes were present, these collisions would cause the atoms to distribute themselves among the energy levels according to Boltzmann’s law with the temperature $T_a$. Then $N_i$ would be $N_1 e^{-\frac{E_i}{kT_a}}$. Actually the number of excited atoms in state $i$ will be much smaller for two reasons. (1) As a consequence of atomic collisions, some excitation energy will be transformed into kinetic energy. This process will “pull” the atoms toward a distribution, according to the lower temperature $T_a$. (2) Emission of radiation will take place, and as a result atoms will pass from high-energy states to lower-energy ones. As a rule, this process is more significant, since the transitions are more probable.

When more than one gas is present in a discharge, excitation may be exchanged between colliding atoms of different kinds, provided that they possess a pair of energy levels near one another. The probability of such an exchange is proportional to $e^{-\frac{\Delta E}{kT}}$, where $\Delta E$ is the energy difference of the participating levels and $T$ is the temperature of the gas mixture. This exchange process is called resonant transfer of energy. It is particularly interesting when it involves a metastable level of one atomic species and an ordinary level of the other. The second species provides a path of escape from the crowded metastable level. Also the transfer of excitation to atoms of the second kind may create such a distortion of their distribution that population inversion may occur.

Population inversion can be established in a single gas by means of electronic excitation, but it is easier to succeed if one makes use of two gases and exploits the energy-transfer phenomenon. The operation of the helium-neon laser described in Chapter II is based on energy transfer from helium atoms to neon atoms. The functioning of this laser in the original range around 1.1µ is explained with the aid of the partial energy-level diagram of Fig. 72, which shows some of the lowest energy levels of helium and of neon. The $2^3S$ state of He is metastable; a direct radiative
transition to singlet ground state is forbidden, but a helium atom can arrive in this state by way of an electron collision process. When helium atoms in the $2^3S$ state collide with neon atoms in the ground state, the excitation may be transferred to the neon atoms, which then end up in one of the $2s$ states, the highest of which lies only about 300 cm$^{-1}$ below the $2^3S$ level of helium. Radiative transitions may then take place from the four $2s$ levels to the ten $2p$ levels. The $2p$ levels may be less populated than the $2s$ levels because there is no direct transfer to them from a helium level. Whether an inversion will actually take place depends on the relative abundance of the He and Ne atoms in the mixture and on the electron temperature. The $2p$ levels fortunately do not get overcrowded because they are readily drained by transitions to the lower $1s$ levels. The success of this scheme naturally depends on the achievement of the correct ratios of excitation and decay rates, which are determined by the gas pressures, the power expended in the discharge, and the radius of the discharge tube which enters into the matter because of the collision of excited atoms with the walls.
Another fortuitous near coincidence of a metastable helium level with a group of neon levels is responsible for the visible, 6328-Å laser. The cycle involved is similar to that of the infrared 1.1-μ lasers. (See Section V.4.)

The selective transfer of excitation from helium to neon atoms makes the achievement of negative absorption in neon easier, but it is not absolutely necessary. Negative absorption can be achieved in pure neon as well as in other pure noble gases provided that the electron density in the discharge is maintained at a proper level and provided gas pressure and tube dimensions are chosen so as to prevent the overpopulation of the terminal levels. The addition of helium may aid the process even in noble gases that have no common energy levels with helium. The reason for this beneficial effect of helium is believed to be that the presence of helium increases the electron density in the discharge through ionizing collisions between metastable atoms. One must not draw the conclusion that the addition of helium is universally beneficial for the functioning of any neon laser. Figures 73 and 74 illustrate the dependence of two stimulated emission lines of neon on gas composition and pressure [4]. The 1.96-μ line is most intense in a mixture containing approximately 5 per cent neon, with the maximum radiative output attained at a total pressure of 0.35 torr. In contrast to this the 2.10-μ line reaches its maximum in pure neon at 0.11 torr pressure.* Figures 73 and 74 should be viewed for the sake of comparison only, since laser outputs vary with the geometry of the discharge (tube radius) and also with the power level of the discharge. There is generally a power level at which the laser output is maximal; further increase of r.f. power decreases the laser output. The gain, or negative absorption coefficient, in neon is approximately inversely proportional to the diameter of the discharge tube. This relationship was found experimentally to hold for tube diameters ranging from a few millimeters to at least 1.5 cm. The explanation for an increase of gain with a decrease in tube diameter involves the role of the 1s levels to which the terminal levels of laser transitions decay. The 1s levels have long lifetimes, but they can be destroyed by collisions with the walls. When the rate of their destruction is too slow the overcrowding at this level will eventually create an overcrowding at the parent 2p levels and population inversion becomes more difficult to attain.

In summary, we may say that the noble gas lasers work by excitation through electron collision; this process is greatly enhanced in certain special cases by the transfer of excitation from metastable helium atoms. Lasers based on noble gas atoms generally work in continuous operation; the radiation emitted is in the infrared. The majority of lines observed so far lies between 1 and 3.5 μ. A notable exception is the popular 6328-Å

* The relatively new unit 1 torr is the pressure of 1 mm Hg.
helium-neon laser, which is visible. The construction and operation of some of these lasers are discussed in Sections V.4 and V.5.

Ions of the noble gases have also been observed to emit stimulated radiation. Their laser emission was noted in the wavelength region 2600 to 8000 Å. The ionic lasers are excited by electron collisions; it probably takes two consecutive collisions to produce an excited Ne\(^+\) ion and three to produce an excited Ne\(^{++}\). Although the atomic laser lines of the noble gases are conveniently excited with low power (20 to 80 watts), the ionic lines require a dense discharge which dissipates several thousand watts. Consequently it is much easier to obtain ionic lines in pulsed discharges with a relatively low duty cycle than in continuous discharges which require heavy current sources and well-designed heat sinks. Some laser lines require pulse excitation because the relevant population inversion cannot be established as a stationary process, the lifetimes of the decay processes

![Figure 73](image_url)

**Fig. 73.** Variation of the intensity of the 1.9574-µ (3\(p_4 \rightarrow 2s_5\)) neon line with gas pressure for various percentages of neon in the He-Ne mixture.
being such that eventually an accumulation of population occurs at the lower laser level.

Charge transfer between colliding ions and atoms or molecules may also serve as a source of population inversion. It is possible that some of the observed laser lines may be explained by a charge transfer process [5].

Transfer of excitation between colliding atoms has already been mentioned as a process that is instrumental in producing population inversion among some neon levels. It is interesting that a similar collision between excited atoms and diatomic or polyatomic molecules may lead to population inversion in one of the atoms of the molecule by first causing the dissociation of the molecule. Such a process has produced laser action in oxygen, nitrogen, carbon, sulfur, and bromine. The atoms of helium, neon, or argon were used as carriers of the energy. They in turn were excited in an electric discharge of the same type that is used with the noble gas lasers.

Fig. 74. Variation of the intensity of the 2.1041-μ (3p₁ → 2s₂) neon line with gas pressure for various percentages of neon in the He-Ne mixture.
discharges and they are known to depend on nonstationary population inversions [6]. Some of these newer lasers are discussed briefly in Section V.6.

REFERENCES


2. SPECTROSCOPY OF NOBLE GASES

The common characteristic of the electronic structure of the noble gases neon, argon, krypton, and xenon is that the highest p-shells are filled and there are no electrons outside of these shells when the atom is in its ground state. Since the p-shells hold six electrons, the symbols of the electron configurations of these elements end with 2p⁶, 3p⁶, 4p⁶, and 5p⁶, respectively. The total angular momentum \( J \), the orbital angular momentum \( L \), and the spin angular momentum of such a closed shell configuration are all zero. When excitation takes place one of the electrons moves out of this closed shell, leaving a core of five p-electrons behind. Thus, in the case of Ne excited configurations of the type 2p⁶3s, 2p⁶3p, 2p⁶3d, 2p⁶4s, etc. arise. The symbols of the completed shells 1s²2s², which logically precede the 2p⁶ symbol, were omitted.

The outer electron of an excited noble gas atom is not coupled to the core electrons according to the rules of the L-S, or Russell-Saunders, coupling discussed in Section I.6. The interactions are such that in good approximation the orbital angular momentum \( L \) of the outer electron is coupled to the total angular momentum \( J_c \) of the core.* The resultant

* Bold face letters are used to distinguish a vector (\( \mathbf{I} \)) from its absolute value (\( I \)).
vector $K = J_c + l$ is then coupled to the spin of the outer electron to give a total angular momentum whose absolute value is $K \pm \frac{1}{2}$. Such coupling is called *pair coupling*. The terms of an atom in which pair coupling prevails are designated by Racah’s symbols, which consist of the symbol of the outer electron configuration followed by $[K]$. These symbols are exemplified by the first excited states of neon: $3s\left[\frac{3}{2}\right]$ and $3s'\left[\frac{1}{2}\right]$. They are arrived at as follows: The orbital angular momentum of the core must be 1 and the spin $\frac{1}{2}$; they combine either parallel or antiparallel, giving a $J_c = \frac{3}{2}$ or $\frac{1}{2}$.

![Fig. 75. Chart of the lowest excited states of Ne atoms. Paschen symbols on right, electron configuration of the Racah symbols on left, number of terms in center. (* indicates that some terms in this group have special Paschen symbols.)*](image)

Antiparallel combination of core orbital momentum and spin is indicated by the prime. Since the angular momentum of the $3s$ electron is 0, we must have $K = \frac{3}{2}$ in the first case and $K = \frac{1}{2}$ in the second. The total angular momentum $J$ of the atom is obtained by adding (vectorially) the spin of the $3s$ electron to $K$. Thus for $3s\left[\frac{3}{2}\right]$ the possibilities are $J = 2$ or 1; for $K = \frac{1}{2}$, on the other hand, $J = 1$ or 0. These values of $J$ are frequently written as subscripts; thus the symbols of the four lowest excited states of neon are $3s\left[\frac{3}{2}\right]$, $3s\left[\frac{1}{2}\right]$, $3s'\left[\frac{1}{2}\right]$, and $3s'\left[\frac{1}{2}\right]$. The reader can convince himself that the $3p\left[\frac{1}{2}\right]$ configurations ($J_c = \frac{3}{2}$) of Ne are possible with $K = \frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$, whereas the $3p'\left[\frac{1}{2}\right]$ configurations ($J_c = \frac{1}{2}$) permit only $K = \frac{1}{2}$ and $\frac{3}{2}$. 
Since the spin of the outer electron permits two orientations for each orbit, we obtain a total of ten states of the $3p$ type.

Racah’s notation is admittedly complicated, but it describes the physical situation or at least it provides a model. Unfortunately, the most commonly used notation, at least for Ne, is the Paschen notation, which is simply a system of shorthand symbols. Although the letters $s$, $p$, and $d$ are used, we cannot safely infer that a Paschen symbol with the letter $s$ always refers to an outer electron in an $s$ orbit. Paschen’s symbols must be treated as

![Diagram of excited states](image)

**Fig. 76.** Chart of the lowest excited states of Ar atoms. Paschen symbols on right, electron configuration of the Racah symbols on left, number of states in center. (* indicates miscellaneous or improvised symbols.)

arbitrary names given to levels. To find out what quantum-mechanical model represents the level, we must consult a list of these symbols which relates these to the model. Moore’s atomic-energy-level tables [1] contain the necessary information.

For the sake of orientation we include a chart of the lowest excited levels of Ne (Fig. 75) and a similar chart for Ar which is representative of the heavier noble gases (Fig. 76). The change in the angular momentum of the core from $J_c = \frac{1}{2}$ to $\frac{3}{2}$ affects the energy of the neon levels by only relatively small amounts. For this reason the levels which differ by only
the value of $J_c$ are not shown separately on the chart of neon, and the primes of the Racah symbols are also omitted. The separation of the energy levels with differing values of $J_c$ progressively increases with increasing atomic number. It is already conspicuous in the case of argon, and the grouping of the levels according to the value of $J_c$ is shown on the argon chart.

Each column of the charts contains infinitely many energy levels; their Racah configuration numbers are $ns$, $ns'$, $np$, $np'$, etc. All unprimed sequences tend to the same limit, the ionization energy required to produce a singly ionized noble gas atom in the $^2P_{3/2}^0$ state; all primed sequences tend to another limit, which represents the ionization energy required to produce an ion in the $^2P_{1/2}^0$ state. The latter energy is greater (in accordance with Hund’s rules), and the energy difference increases with increasing atomic number.

Table V.1 at the end of this section contains the numerical values of 44 levels of Ne including the groups $3s$, $4s$, $5s$, $3p$, $4p$, and $3d$ (Racah). Transitions among these levels account for the majority of observed Ne laser lines. The ground level is assigned the energy 0. Frequently tables are constructed from the ionization level $E_\infty$ down. The entries of such tables are $E_\infty - E_n$; they are called term values. For Ne, the value of $E_\infty$ is 173,931.7 cm$^{-1}$. Neon levels included in our short table represent but a fraction of the Ne levels contained in Moore’s tables [1]. It is interesting to note that the lowest excited level of Ne is over 134,000 cm$^{-1}$ above ground level and to compare this value with the first excited levels of ions which play a role in solid-state lasers.

Some conclusions can be drawn about the probabilities of radiative transitions between the levels listed in the tables from the selection rules (p. 36) and the quantum numbers listed in the tables. The actual calculation of the transition rates of permitted transitions is a very complicated task. It requires an approximate knowledge of the wave functions characterizing the states. The calculation of the transition rates, line strengths, and related properties of noble gases is described in the literature [2, 3]. Closely related to the transition rates are the decay rates of the states. These rates can be determined experimentally under conditions similar to those found in the laser. Although they represent engineering-type data, they have a certain physical meaning. The decay rate of any one state represents the sum of transition rates, radiative and otherwise, to all lower levels. Bennett [4] measured and compiled decay rates of the lowest Ne states. These are quite useful in laser calculations; therefore they were included in the table whose last column is the lifetime $\tau$ in nanoseconds. The decay rate is $\tau^{-1}$. (See p. 31.)

* The superscript 0 indicates an odd term. See p. 36.
The spectrum of an ion is quite different from the spectrum of the neutral atom from which the ion derives. Similarities exist however among the spectra of atoms and ions which contain the same number of electrons; such structures are called isoelectronic. Oxygen atom, singly ionized fluorine, and doubly ionized neon, for example, form an isoelectronic sequence. It is customary in spectroscopy to distinguish between the spectra of an atom and its various ions in the following manner: The spectrum of the neutral atom is provided with a Roman numeral I, that of the singly ionized atom by II, and so on. Thus we have so far discussed the energy-level structures of NeI, ArI, etc.

The principal features of the energy-level structure of singly ionized noble gases are illustrated on the example of NeII. The ground state configuration of NeII is 1s^22s^22p^5. Excited configurations are formed by promoting one of the 2p electrons to a higher orbit, say 3s, 3p, 3d, etc., or, exceptionally, by promoting one of the 2s electrons to obtain the configuration 1s^22s^2p^6. Leaving aside the exceptional situation, NeII will consists of a core with a 1s^22s^22p^4 configuration and an external electron which may be located in a variety of orbits with a principal quantum number at least 3. In the case of ArII, the core has a 1s^22s^22p^63s^23p^4 configuration and the external electron may be on either a 3d orbit or any orbit whose principal quantum number is at least 4. The halogens are isoelectronic with the singly ionized noble gases; therefore what was said about NeII and ArII also applies to FI and ClI, respectively.

In these atoms two electrons are missing from the core, so that the spin of the core is 0 or 1. When this spin is combined with the spin of the outer electron, the resultant spin coordinate S has the value ½ or ¾. The energy-level scheme will therefore consist of doublets $S = \frac{1}{2}$, and of quartets $S = \frac{3}{2}$. (Transitions between terms of different spin are not completely absent because the Russell-Saunders scheme is not completely applicable.) Removal of one of the p electrons may leave the core itself in several different states which are either singlets or triplets. Application of Pauli’s principle leads to the conclusion that the possible terms are $^3P$, $^1D$, and $^1S$. The lowest energy is associated with the $^3P$ core. A term of NeII with a lowest-energy core and with the outside electron in the lowest s orbit is 3s$^4P$, another with spin of the outside electron reversed is 3s$^2P$. The first term permits the J values $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{1}{2}$; the second only $\frac{3}{2}$ and $\frac{1}{2}$. When the core is in its next lowest state ($^1D$) the electronic symbol is provided with a prime; thus a term designated by 3s$^2D$ may be obtained. Double primes indicate a $^1S$ core.

In attempting to organize the energy levels of atoms we must keep in mind that the Russell-Saunders coupling scheme, as well as other coupling schemes, represents only approximations which do not have universal
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<td></td>
</tr>
<tr>
<td>$3p_8$</td>
<td>2</td>
<td>162 901.1</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>1</td>
<td>163 014.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$3p_6$</td>
<td>2</td>
<td>163 040.3</td>
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<td></td>
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<tr>
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<td>163 403.3</td>
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<td></td>
</tr>
<tr>
<td>$3p_4$</td>
<td>$4p[3]^p$</td>
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<td>163 659.2</td>
<td></td>
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<td>163 710.6</td>
<td></td>
</tr>
<tr>
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<td>164 287.9</td>
<td></td>
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<tr>
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<td>$5s[3]^p$</td>
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<td>165 830.1</td>
<td></td>
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<tr>
<td>$3s_4$</td>
<td>$5s[3]^p$</td>
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<td>165 914.8</td>
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<tr>
<td>$3s_3$</td>
<td>$5s[4]^p$</td>
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<td>166 608.3</td>
<td></td>
</tr>
<tr>
<td>$3s_2$</td>
<td>$5s[4]^p$</td>
<td>1</td>
<td>166 658.5</td>
<td></td>
</tr>
</tbody>
</table>
validity. As we progress through the elements He, Ne, Ar, Kr, Xe, we find that the forces responsible for Russell-Saunders coupling decrease in comparison with other forces. Even within one element the ratios of these forces are different for electrons that pass near the nucleus and for those whose orbits are farther removed. For heavier elements and for large quantum numbers \( n \) and \( l \) of the outer electron, the coupling approaches pair coupling.

Spectral lines, whether derived from spontaneous or from stimulated emission, can be given a transition assignment among the terms tabulated in the atomic energy tables insofar as the tables are complete. With the extension of stimulated emission spectroscopy into the infrared, lines are occasionally observed which result from transitions among levels not yet tabulated.

REFERENCES

3. CONDITIONS FOR LASER OSCILLATION IN GASES

The complete fluorescence cycle in a gas laser usually involves at least four energy levels. Let us denote the ground level by 1, and the levels of laser transition by indices 2 and 3, realizing that there may be levels above 3 and below 2 participating in the cycle. As before, let \( N_i \) designate the number of atoms per unit volume at level \( i \), and \( g_i \) the multiplicity \((2J + 1)\) of that level.

Population inversion (negative absorption) will be achieved when

\[
\frac{N_3}{g_3} - \frac{N_2}{g_2} > 0. \quad (3.1)
\]

A necessary, but by no means sufficient, condition for population inversion in a stationary state may be obtained from (3.1). The population of state 2 is increased by radiative transitions from state 3 and by other processes. It is diminished by a variety of processes, resulting in a total effective decay rate of \( R_3 \). Then

\[
\frac{dN_2}{dt} \geq N_3 A_{32} - N_2 R_3. \quad (3.2)
\]
Therefore in a stationary state \( N_3 A_{32} \leq N_2 R_2 \); hence
\[
\frac{N_3 g_2}{N_2 g_3} \leq \frac{R_2 g_2}{A_{32} g_3}.
\]

Population inversion takes place when the left side of (3.3) exceeds 1. This can occur only when
\[
A_{32} g_3 < R_2.
\]

The left side of this inequality contains atomic constants only; the right side is under the control of the experimenter to some extent. He must arrange circumstances so that the total rate of decay from the terminal level 2 exceeds the minimum set by (3.4).

The threshold of oscillation will be reached when the left side of (3.1) is not only positive, but large enough to compensate for the losses of the instrument. According to equation (2.8) of Chapter II this will occur when
\[
\left(\frac{N_3}{g_3} - \frac{N_2}{g_2}\right) = \frac{\gamma}{L P g_2 K},
\]
where \( \gamma \) is the loss coefficient, \( L \) is the length of the laser, and \( K \) is the integrated absorption coefficient of the line given by (6.9) of Chapter I. The shape of the absorption line in gases under low pressure is determined by the Doppler effect; the peak value, \( P \), of the curve is given by (6.14) of Chapter I. Combining these equations, we obtain the threshold condition for a gas laser:
\[
\left(\frac{N_3}{g_3} - \frac{N_2}{g_2}\right) = \frac{8\pi}{A_{32} g_3 \lambda^3} \sqrt{\frac{2\pi R T_2}{M L}} \gamma.
\]

Here \( M \) is the molecular weight of the gas \( R = Nk = 8.317 \times 10^7 \) erg/mole deg., and \( T_2 \) is the temperature of the gas. The formula shows that, other things being equal, it is easier to fulfill the threshold condition for longer wavelengths.

We have already noted that several processes of excitation and de-excitation are operative in gases. All processes are subject to the principle of detailed balance, which relates the probability of a process to that of the inverse process. Let us consider a large reservoir \( R \) of interacting particles in thermal equilibrium with each other at a temperature \( T \), and an experimental system \( E \) capable of interaction with the particles of \( R \) by means of a variety of physical processes. The principle of detailed balance asserts that each process acting separately will ultimately produce the same statistical
distribution of energy in $E$, namely a Boltzmann distribution with a temperature $T$. In the situation of interest here, $R$ may be represented by the electrons in the discharge, whose energy distribution is at least approximately that required for the validity of the argument. A similar result is obtained if we consider the interaction of the atomic system with a radiation field whose energy distribution is given by Planck’s radiation law.

Since all such processes tend to produce thermal equilibrium if acting alone, the question arises of how one can ever obtain a steady-state population inversion. The answer is that the rates at which these processes proceed toward equilibrium are all different. Consequently, we may play one of the mechanisms against the others and operate the system in an intermediate steady-state condition where no one process dominates and where no single temperature characterizes the population of all atomic levels. The situation is helped in multilevel systems if strong departures can be obtained from a Maxwellian (equilibrium) distribution of the interacting particles or if a highly monochromatic radiation field is employed in place of black-body radiation. There is, however, for each excitation process an inverse process which tends to produce thermal equilibrium, and the production of population inversion involves minimizing the effects of the inverse process at some stage. This may be accomplished, for example, by allowing light from some levels to escape through the walls, or by allowing metastable atoms to diffuse out of the discharge. Like the thermodynamic engines, the lasers must have a sink. This sink however may be one or more steps removed from the lower laser level.

The following energy exchange processes are important in a discharge that takes place in a single gas:

1. Electron collision of the first kind in which an atom gains energy from an electron.
2. Electron collision of the second kind in which an excited atom loses energy to an electron.
3. Spontaneous emission of radiation from an excited atom.
4. Absorption of radiation by an atom.
5. Stimulated emission of radiation by an atom.

The rates at which these processes take place are determined by the numbers of atoms available in the proper states and by the probabilities that these phenomena will occur in the unit time for an individual particle. These probabilities are stated as reciprocal lifetimes and they relate to atoms in given states.

Let $\theta_{ij}$ denote the lifetime of the transition of an atom from level $i$ to level $j$ when the atom is subjected only to collisions with electrons of a given density in equilibrium among themselves at the absolute
temperature $T$. If no other processes than these collisions were to take place, the rate of change of the number of atoms at level $i$ would be given by

$$
\frac{dN_i}{dt} = \sum_j \left( \frac{N_j}{\theta_{ji}} - \frac{N_i}{\theta_{ij}} \right).
$$

(3.7)

We shall now show that $\theta_{ij}$ and $\theta_{ji}$ are simply related. Let us assume that a thermodynamic equilibrium is established at temperature $T$; the number of atoms in each level will then be stationary. Let these stationary, or equilibrium, values be denoted by $N_i^*$. Then

$$
\sum_j \left( \frac{N_j^*}{\theta_{ji}} - \frac{N_i^*}{\theta_{ij}} \right) = 0
$$

(3.8)

for every value $i$. Moreover, the principle of detailed balance requires that the exchanges between each pair of energy levels balance out. This means not only that the sum in (3.8) must vanish but that

$$
\frac{N_j^*}{\theta_{ji}} - \frac{N_i^*}{\theta_{ij}} = 0
$$

(3.9)

must hold for every $i$ and $j$. Therefore

$$
\frac{\theta_{ij}}{\theta_{ji}} = \frac{N_j^*}{N_i^*}.
$$

(3.10)

Since $N_i^*$ and $N_j^*$ are occupation numbers of the levels in thermal equilibrium at temperature $T$, it follows from Boltzmann's law that

$$
\frac{\theta_{ij}}{\theta_{ji}} = \frac{g_i}{g_j} e^{- (E_i - E_j)/kT}.
$$

(3.11)

When atomic transitions are the result not only of electron collisions but of other processes as well, the distribution will not be given by

$$
\frac{N_i}{N_j} = \frac{g_i}{g_j} e^{- (E_i - E_j)/kT}.
$$

(3.12)

This fact, however, does not affect the validity of (3.11), which is a relation between probabilities. The stationary distribution can be obtained by analysis of the change due to all causes in the population of a given level. In the simplest case of two levels, with a spontaneous radiative transition with lifetime $\tau_2$ from levels 2 to 1, the rate equation is

$$
\frac{dN_2}{dt} = \frac{N_1}{\tau_{12}} - \frac{N_2}{\tau_{21}} - \frac{N_2}{\tau_2}.
$$

(3.13)

* Actually, the symbol $T_e$ would be preferable to indicate "electron temperature," but we omit the $e$ in view of the overabundance of subscripts.
Conditions for Laser Oscillation in Gases

In the stationary state

\[ \frac{N_2}{N_1} = \frac{1/\theta_{12}}{1/\theta_{21} + 1/\tau_2}. \]  

(3.14)

When the radiative process is comparatively fast, so that \( \tau_2 < \theta_{21} \), we have

\[ \frac{N_2}{N_1} \approx \frac{\tau_2}{\theta_{12}} = \frac{g_2\tau_2}{g_1\theta_{21}} e^{-(E_2 - E_1)/kT}. \]  

(3.15)

The ratio \( \tau_2/\theta_{21} \) is a measure of the departure from the Boltzmann equilibrium distribution. To an extent, this factor is under the control of the experimenter, since \( \tau_2 \) is fixed, but \( 1/\theta_{21} \) is proportional to the electron density in the discharge. However, \( N_2/N_1 \) cannot be increased arbitrarily by increasing the electron density because the validity of (3.15) is based on the assumption that \( \tau_2/\theta_{21} \) is much less than 1.

Let us now introduce an additional level above level 2, and let us assume no direct interaction between these levels (2 and 3). Under similar assumptions, we find

\[ \frac{N_3}{N_1} \approx \frac{g_3\tau_3}{g_1\theta_{31}} e^{-(E_3 - E_1)/kT}. \]  

(3.16)

From (3.15) and (3.16) it follows that

\[ \frac{g_2 N_3}{g_3 N_2} \approx \frac{\tau_3}{\tau_2} \frac{\theta_{21}}{\theta_{31}} e^{-(E_3 - E_2)/kT}. \]  

(3.17)

Even though the exponential factor is always less than 1, the factor in front of the exponential may be sufficiently greater than 1 for certain values of the parameters to cause \( N_3 g_2 \) to exceed \( g_3 N_2 \). This is the condition of population inversion or negative absorption. In order to achieve this condition by electron excitation alone, we must select materials and states so that \( \tau_3/\tau_2 \) is large without an adversely large ratio of \( \theta_{31}/\theta_{21} \). Actually, there are many complicating factors not yet considered. One is that levels other than those already listed become populated by electron collisions. If in their radiative decay they populate the lower level 2 in preference to level 3, they may spoil the narrow margin otherwise available for population inversion.

In order to satisfy the threshold condition for oscillation, not only must \( N_3 \) exceed \( N_2 \) but the difference \( N_3 - N_2 \) must exceed a certain minimum. Therefore, other conditions being equal, it is desirable to have a discharge of high density. At high densities the absorption of the photons produced by spontaneous emission in other atoms becomes important. Reabsorption of the spontaneously emitted radiation increases the lifetime in the excited level. This effect is useful when it increases the lifetime of the atom in the
upper level 3. It will take place when the optical transition between levels 1 and 3 is permitted by the selection rules. Under these conditions the system can operate at relatively high densities. However, when the ground level is connected to level 2 by a permitted transition, a high density is undesirable because it increases the relative population of level 2. This unfavorable situation prevails for the levels of helium already mentioned.

The quantum theory of transitions indicates that in the case of transitions allowed by the selection rules the ratio $\frac{\tau_i}{\theta_{ii}}$ is essentially the same for all levels. It can be seen from (3.17) that negative absorption cannot be obtained in such a case. At least one of the two levels in the system must therefore be such that optical transition between it and the ground state is forbidden. This level is excited by electron collisions; the excitation cross section for this process is generally smaller than for optically allowed transitions. In a favorable case the transition between the ground level and the lower state is forbidden, and the small excitation of this level favors the production of negative temperature. Such a situation was found for many transitions in noble gases but population inversion could be achieved by electron excitation alone only when provision was made to relieve the overpopulation of the terminal laser level resulting from transitions from higher levels to that level.

The first gaseous lasers contained a mixture of He and Ne in the proportion about 10 to 1. Population inversion was established between the 2s and 2p (Paschen) levels of neon with the assistance of resonant transfer of excitation from helium atoms to neon atoms. This transfer is not absolutely necessary, as experiments with pure neon and other noble gases have shown, but it is a process which under certain circumstances aids significantly the attainment of population inversion. The circumstances are described by the rates of the various energy exchange processes which take place between the electron gas in the discharge and the two components of the gaseous mixture.

The kinetics of this process has been developed by Basov and Krokhin [1, 2], whose work is reproduced here briefly. The two gases are distinguished by the letters $a$ and $b$. The atoms of the working gas $a$ have three relevant levels: $E_1 = 0$, the ground level, $E_2$, and $E_3$. The atoms of the auxiliary gas $b$ have two relevant levels, $E_1 = 0$ and $E_3$. The coincidence, or near coincidence, of level 3 is essential for the operation of the scheme because it is a condition for significant transfer of excitation between the two atomic species.

The rate of change of the number of atoms of gas $a$ in level $E_3$ is

$$\frac{dN_{3a}}{dt} = N_{1a}\left(\frac{1}{\theta_{13}} + \frac{1}{t_{ba}}\right) - N_{3a}\left(\frac{1}{\theta_3} + \frac{1}{t_{ab}} + \frac{1}{\tau_3}\right).$$ (3.18)
Here $1 / t_{ba}$ is the rate per ground state atom of gas $a$ of resonant transfer of energy in collision with atoms of gas $b$ on level 3, and $1 / t_{ab}$ is the rate per excited atom of gas $a$ for the inverse process; $1 / \theta_3$ is the total transition rate for gas $a$ from level 3 resulting from collisions of the second kind with electrons; that is,

$$\frac{1}{\theta_3} = \frac{1}{\theta_{31}} + \frac{1}{\theta_{32}}.$$  \hspace{1cm} (3.19)

The parameter $\tau_3$ is the radiative lifetime of level 3, including radiative transitions to both lower levels. Excitation from level 2 to level 3 is neglected in this analysis because a term describing the process is proportional to $N_2^a$, hence much smaller than the leading terms in (3.18).

The rate of change of the number of atoms $N_2^a$ on level $E_2$ is given by

$$\frac{dN_2^a}{dt} = N_1^a \left( \frac{1}{\tau_{12}} + \frac{1}{\tau_{32}} \right) - N_2^a \left( \frac{1}{\theta_{21}} + \frac{1}{\tau_{21}} \right),$$  \hspace{1cm} (3.20)

where the $\tau$'s indicate radiative lifetimes.

In the stationary case the derivatives vanish. The condition of negative absorption between the levels $E_3$ and $E_2$ of gas $a$ is given by

$$N_3^a > N_2^a.$$  \hspace{1cm} (3.21)

When the left sides of (3.18) and (3.20) are set equal to zero, the ratio $N_3^a / N_2^a$ can be calculated in terms of the lifetimes occurring in these equations. The condition (3.21) becomes an inequality involving the combinations of these lifetimes. We then make use of (3.11) as well as of the fact that according to collision theory

$$\frac{t_{ba}}{t_{ab}} = \frac{N_1^b}{N_3^b}.$$  \hspace{1cm} (3.22)

This ratio is determined by the effective excitation temperature for the atoms of gas $b$. It may be considered an external parameter as far as the distribution of the atoms of gas $a$ is concerned. The condition (3.21) then becomes

$$\frac{1}{t_{ba}} \left[ 1 - \frac{N_1^b}{N_3^a} e^{-E_2/kT} + \theta_{21} \left( \frac{1}{\tau_{21}} - \frac{1}{\tau_{32}} - \frac{1}{\theta_{32}} \right) \right]$$

$$> \theta_{31} \left( \frac{1}{\theta_3} + \frac{1}{\tau_3} \right) e^{-E_3/kT} - 1 + \theta_{21} \left( \frac{1}{\tau_{32}} + \frac{1}{\theta_{32}} - \frac{1}{\tau_{21}} \right).$$  \hspace{1cm} (3.23)

The quantities $\theta_{31}/\theta_3$ and $\theta_{31}/\theta_{32}$ do not depend on the density of the electrons in the discharge but are determined by the cross sections of the corresponding processes and by the temperature of the electrons.
Inequality (3.23) has a different meaning in different regions of the variables \( \theta_{21}(1/\tau_{21} - 1/\tau_{32}) \) and \( \theta_{31}/\tau_3 \). The range of these variables is divided by two curves into four regions. The equations of the curves are obtained by setting the expression in brackets and the right side equal to zero. To the first order of approximation the rate \( 1/\theta_{32} \) may be neglected in comparison with \( 1/\tau_{32} \) and the assumption may be made that \( \theta_{31}/\theta_3 \approx 1 \); the curves mentioned above then become straight lines described by the equations

\[
\frac{\theta_{21}}{\tau_3} \left( \frac{1}{\tau_{21}} - \frac{1}{\tau_{32}} \right) = \frac{N_1^b}{N_3^b} e^{-E_3/kT} - 1, \tag{3.24}
\]

\[
\frac{\theta_{31}}{\tau_3} = \frac{\theta_{21}(1/\tau_{21} - 1/\tau_{32}) + 1 - F(T)}{F(T)}, \tag{3.25}
\]

where \( F(T) = e^{(E_3 - E_2)/kT} \).

Clearly, to the right of the vertical line in Fig. 77, whose equation is (3.24), the condition (3.23) is satisfied for any value of \( t_{b0} \) when the right side is negative. This situation corresponds to region I in the figure. In this case \( t_{b0} = \infty \) also leads to population inversion, and gas \( b \) is unnecessary. In region II population inversion occurs because of the presence of gas \( b \). In region III gas \( b \) impedes the formation of a population inversion. The occurrence of population inversion in region IV is impossible.

Unfortunately the variables involved in equations (3.24) and (3.25) are generally not known to a degree of precision necessary to make predictions from this theory in specific cases. The value of the theory lies mostly in disclosing the possibilities and in specifying the variables which affect the phenomena.
REFERENCES


4. COMMON HELIUM-NEON LASERS

The neon lasers whose operation is based on the transfer of excitation from helium to neon atoms form the most important group of gaseous lasers. They were the first to be discovered and studied and are the easiest to construct and to operate. Most of the knowledge and experience available in 1964 on gaseous lasers was obtained on such He-Ne lasers. Spectroscopic data pertaining to these lasers are summarized in Table V.2. Laser oscillations in other lines of neon are discussed in Section V.5 together with lasers of other noble gases.

Javan, Bennett, and Herriott [1] made the original discovery in 1960, observing stimulated emission on five nearby lines in the near infrared, lines 2 to 6 in Table V.2. The strongest oscillation occurs in line 3 at 1.1523 μ. The radiation arises from stimulated transitions from the 2s to the 2p levels of Ne; the population of the former group is enhanced by transfer from the 23S state of He. These transitions are shown on the energy-level diagram of Fig. 78; many other transitions have subsequently been observed in stimulated emission.

TABLE V.2. SELECTED LASER LINES OF NEON*

<table>
<thead>
<tr>
<th>Line Number</th>
<th>λair, Microns</th>
<th>σr, cm⁻¹</th>
<th>Transition</th>
<th>Gain, cm⁻¹</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>0.6328</td>
<td>15798.0</td>
<td>3s₂→2p₄</td>
<td>5 × 10⁻⁴</td>
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<tr>
<td>2</td>
<td>1.1177</td>
<td>8944.07</td>
<td>2s₂→2p₉</td>
<td>12 × 10⁻⁴</td>
</tr>
<tr>
<td>3</td>
<td>1.1523</td>
<td>8676.10</td>
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</tr>
<tr>
<td>4</td>
<td>1.1614</td>
<td>8607.87</td>
<td>2s₃→2p₅</td>
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<tr>
<td>5</td>
<td>1.1985</td>
<td>8341.53</td>
<td>2s₃→2p₂</td>
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</tr>
<tr>
<td>6</td>
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<td>8285.25</td>
<td>2sₙ→2p₀</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>1.5231</td>
<td>6563.87</td>
<td>2s₂→2p₁</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>3.3913</td>
<td>2947.90</td>
<td>3s₂→3p₄</td>
<td>&gt; 4 × 10⁻²</td>
</tr>
</tbody>
</table>

* For a complete table of Ne lines observed in stimulated emission see Table A.2 in the Appendix.

White and Rigden [2] discovered the visible He-Ne laser (line 1) in 1962. The radiation arises from the 3s₂→2p₄ transition; the population of the upper level is enhanced by the transfer of excitation from the 2¹S state.
of He. This laser is the most convenient one to use for demonstration and alignment purposes; several companies market such lasers for general use.

It is somewhat inaccurate to refer to the 6328-Å laser as the visible Ne laser, since laser oscillations can be produced in other visible Ne lines. Once the 3s₂ level of Ne is heavily populated, one can force laser transitions into a number of the 2p levels other than 2p₄. The gain in these transitions is less than that in the 3s₂-2p₄ transition; therefore longer discharge tubes are used with dispersive prisms incorporated into the path of the beam within the mirrors to separate the desired wavelength and enhance its oscillations, suppressing all others [3]. These unusual visible Ne laser lines are included in Table A.2 in the Appendix.

Shortly after the discovery of the visible He-Ne laser Bloom, Bell, and Rempel [4] observed that infrared radiation of over 3-μ wavelength
frequently accompanies the emission of the visible line and that the emission of this infrared radiation interferes with the operation of the visible laser. The transition responsible for the infrared radiation originates at the $3s_2$ level, the starting level of the visible radiation. It terminates at the $3p_4$ level, yielding a radiation of $3.3913 \mu m$ in wavelength (line 8). The operation of this $3.39-\mu m$ laser not only depletes the $3s_2$ level—to the detriment of the population inversion required for the visible laser—it overpopulates the $3p_4$ level and thereby offers the possibility of enhancing laser action originating at the latter level. Such a cascade effect was observed at the end of 1963 by American [5], French [6], and German [7] investigators simultaneously. The main line of this cascade proceeds from $3p_4$ to $2s_2$ with emission of $2.3951-\mu m$ radiation, then to $2p_4$ with the emission of the $1.1523-\mu m$ line. Several other branches of such a cascade were also observed [7, 8]. The intensity of a cascade line depends on the intensity of the line preceding it in the cascade. The latter can be controlled by varying the regeneration of the laser in a frequency-dependent manner.

It is very easy to attribute to He a role that it does not entirely deserve, and this was frequently done in the early literature. It should be noted that the strongest He-Ne lines are obtainable in pure neon [9], and many Ne lines can be produced in stimulated emission whose upper level is not connected with an excited state of He. Even some of the so-called He-Ne cascades were eventually observed in pure Ne [10]. Therefore we ought to conclude that the transfer of excitation from He substantially enhances population inversion in certain levels of Ne without being the exclusive cause of such inversion. How important the effect of He can become practically may be judged by the results of Patel [9], who compared stimulated emission in pure Ne with emission in an optimized mixture of He and Ne. In a tube 2 meters long and 1.5 cm in diameter, the maximum output of the $1.1523-\mu m$ radiation was 0.2 mW in pure Ne, while in a 10 to 1 He-Ne mixture an output of 10 mW was obtained.

The general appearance of the early He-Ne lasers was shown in Fig. 16, p. 56. These lasers had plane mirrors inside their vacuum envelope. The fabrication of such a system and the adjustment of the mirrors was difficult. Subsequently the plane mirrors were replaced by spherical ones for the reasons discussed in Chapter II, and they were moved to the outside to facilitate fabrication, alignment, and exchange. The passage of radiation out of the vacuum envelope was accomplished with minimal reflections by the use of optical flats as end plates, and by orienting these at the Brewster angle. Figure 79 is the schematic diagram of such a laser; Fig. 80 is a photograph of a laser with external mirrors constructed at the Hughes Research Laboratories in 1962.

The optimal tube diameter seems to be around 6 to 8 mm. Although
Lasers 2 meters long are frequently used in spectroscopic studies, and one 8 meters long has been reported. A 70-cm tube is adequate to excite the lines listed in our table provided that high-quality dielectric mirrors are used, which ensure a reflection coefficient of about 99 per cent for the
weaker lines. The requirements are less stringent for the 1.15-\(\mu\) line, which has a high gain. Considerably shorter tubes (10 cm) are adequate for the 3.39-\(\mu\) line, and the reflectivity of the mirrors can also be reduced in this case.

It was shown earlier that the gain, or negative absorption, is proportional to the transition probability and to the factor

\[
p = \frac{N_2}{g_2} - \frac{N_1}{g_1}
\]

This factor, which describes the population inversion in the medium, depends on the operating conditions of the discharge and on the geometry as well as the gas composition. It generally varies from the center to the periphery of the discharge, since its value is the result of the balance of processes which include collisions with the walls. Because of the great variability of this factor, experimental values of gain have limited physical meaning; they have practical or engineering significance when applied to situations similar to those in which they were measured, and they serve as measures of comparison for different spectral lines excited simultaneously. The tabulated values of the gain (Table V.2) are those of Faust and McFarlane [11]; the corresponding (not tabulated) values for the other 2s\(-\)2p lines are of the order of 1 to 2\( \times \) 10\(^{-4}\) cm\(^{-1}\). Faust and McFarlane estimated that in an optimized He-Ne discharge with the gain values as stated, the values of \(p\) are as follows:

<table>
<thead>
<tr>
<th>Line</th>
<th>(6328 , \text{Å})</th>
<th>1.15 (\mu)</th>
<th>3.39 (\mu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(p)</td>
<td>(1.3 \times 10^9 , \text{cm}^{-3})</td>
<td>(1.5 \times 10^8 , \text{cm}^{-3})</td>
<td>(\geq 9 \times 10^8 , \text{cm}^{-3})</td>
</tr>
</tbody>
</table>

Considerable work has been done to determine experimentally the optimal characteristics of various lasers and the radiative outputs obtainable from them. The early results are summarized in the review article of Bennett [12], additional contributions were made by Gordon, White, and others [13, 14, 15]. Only a few outstanding facts are quoted here. The best operation of the 6328-Å and the 3.39-\(\mu\) lasers is attained for a 5-to-1 He-to-Ne ratio. The product of the total pressure and the tube diameter should lie between 2.9 and 3.6 torr-mm. For the 2s\(\rightarrow\)2p transitions the optimal composition is 1 torr He and 0.1 torr Ne with the tube diameter around 7 mm.

The discharge may be excited by means of an rf oscillator using external electrodes. A matching transformer of 10 to 15 turns is convenient for the adjustment of the discharge for optimal operation. Radio frequencies from 23 Mc to 40 Mc have been used with equal success; the 27- to 30-Mc region is preferred. The large fields under the external electrodes tend to drive He out of the tubes. This is no serious detriment when the tubes can
be readily refilled. Sealed-off tubes driven by rf excitation get exhausted after a few hundred hours or sooner. Sealed-off tubes of more permanent type have internal electrodes, usually with a heated cathode for d-c operation. Discharge currents between 25 and 100 mA are usually required.

The minimum tube length $L_t$ which produces oscillations is determined by the gain of the line and by the losses at the terminations. For each operating condition one may find an $L_t$. The power output of the laser of length $L$ is a nearly linear function of $L/L_t$ [14]. Experimental results pertaining to a 6328-Å laser with spherical mirrors and a 6-mm discharge tube are shown in Fig. 81. The minimum tube lengths varied from 61 to 88 cm, depending on the discharge current as shown in the insert. Thus an output of 3 to 8 mW can be obtained from visible He-Ne lasers of

![Graph](image_url)

Fig. 81. Output power of a visible neon laser as a function of the length of the discharge. ($L_t$ is the minimum length for oscillations; $i$ is the discharge current. After White, Gordon, and Rigden [14].)
convenient length. Similar results were found for short 1.15-\mu lasers. The minimum length could be made as short as 10 cm, and an output of 12 mW was obtainable from discharges 33 cm long [15].

The radiation pattern of the He-Ne laser depends on its construction, particularly on the mirrors and on the limiting aperture.

Precision measurements of Herriott [16] show that the far-field pattern of the original Bell Laboratories laser (with internal reflectors) consists of a beam whose width is 1 min of arc when the reflectors are adjusted to parallelism. The beam obtained from a laser with confocal external reflectors without a limiting aperture is somewhat fuzzy at the edges; the far-field pattern measured between half-power points is about 30 min. With a circular iris of 2-mm diameter placed 3 cm in front of the exit mirror of the confocal laser, both the near and the far fields become sharply limited, the power density increases, and the far field narrows to about 3 min of arc measured between half-power points. The beamwidth is determined by the angular beamwidth of the cavity mode, which is given by the Boyd-Gordon formula: equation (3.21) of Chapter II. This formula is valid whenever the aperture introduced is not so small that its diffraction pattern further limits the definition of the beam.

At low levels of excitation the output of the external reflector He-Ne laser is plane polarized as a consequence of the discriminatory action of the windows, which by their orientation favor one polarization. When the gain in the laser is considerably above the oscillation threshold, the other polarization will also appear. Similarly, a number of cavity modes may be excited simultaneously, and beat frequencies between these oscillations may be observed [18].

Oscillations occur at or near the peaks of those interferometer or cavity resonances that fall within the linewidth of the atomic resonance where appreciable gain is present. This is illustrated in Fig. 82, which shows the "natural" atomic linewidth, the actual linewidth as it appears in the gas due to Doppler broadening, and finally the fine comblike structure of the cavity resonances.

The most relevant modes of the confocal system are similar to those of the plane parallel system. (Section II.3.) In the simplest case of the axial modes, or TEM\(_{00n}\) modes of Fox and Li, for each interferometer resonance appearing at frequency \(\nu_n\), another resonance exists at the frequency \(\nu_n + 1 = \nu_n + \Delta \nu\), where \(\Delta \nu = c/2L\). Here \(L\) is the separation of the reflectors, and the index \(n\) refers to the number of modes in the standing wave pattern formed between the reflector plates. Thus, for 1 meter separation, \(\Delta \nu = 150\) Mc. Since the atomic linewidth (Doppler) is about 900 Mc, several of these modes may be excited simultaneously.* The oscillations do

* See Section VI.1 for further discussion of the modes.
not take place exactly at the peaks of the interferometer resonances; they are shifted a small amount toward the center frequency of the atomic resonance.

The beat notes between nearby spectral lines can be detected by means of a multiplier phototube. If it were only for the axial modes, the spectrum would consist of approximately equally spaced lines, 150 Mc apart for a reflector separation of 1 meter. There are off-axis, or unsymmetrical, modes as well, whose excitation depends critically on the adjustment of the reflectors. In the case of \( L = 1 \) meter, for example, there is an unsymmetric mode separated by 1.3 Mc from the symmetric mode. As a consequence, satellite lines appear in the beat frequency spectrum with 1.3 Mc separation on either side of the main lines. This is shown in Fig. 83, which is adapted from the work of Herriott [16].

The linewidths of the He-Ne laser oscillations are many orders of magnitude narrower than the resolution of the best spectrometers or interferometers; therefore standard optical techniques cannot be used to measure the linewidth. The linewidth may be determined by examining the beat notes between the different interferometer modes. These beat notes are obtained directly from the multiplier phototube on which the laser light is incident; since the phototube is a square law detector, its output contains the beat frequencies. Early measurements of Javan and Herriott [1, 16] indicated linewidths of 10 to 20 kc. Actually, the lines are considerably narrower, but their measurement is difficult because the most minute changes in the distance between the reflectors cause a shift in the lines.
Thus the system responds in the form of frequency modulation to minute thermal fluctuations and mechanical vibrations. Fortunately, two modes subject to similar frequency variations yield a beat frequency that to a large extent is compensated for mechanical variations of length. The degree of this compensation depends critically on the frequency of each of the two interferometer resonances with respect to the center frequency of the atomic resonance. It has been mentioned in Section II.3 that each interferometer mode is pulled toward the center of the atomic resonance line $\nu_c$. The extent of pulling is a nonlinear function of the deviation of the frequency of the interferometer mode from $\nu_c$; therefore a change in the interferometer length will affect the frequencies of the participating modes in a different way. Consequently, it will also affect the beat frequency unless the participating modes are chosen by tuning in such a manner that these frequency changes of the beat note caused by the pulling are compensated.

Javan and associates [18] produced a 50-kc beat note by mixing two perpendicularly polarized oscillations of a carefully stabilized neon laser operating at 1.15 $\mu$. They obtained a fluctuation of the beat note of only 2 cps over a period of several seconds and from this they concluded that the frequency stability of each oscillation was about 1 cps. Although the experiment was a remarkable one, the final conclusion would have been warranted only if the oscillations had been independent. As a matter of
fact, their fluctuations were correlated because all precautions were taken that fluctuations in laser parameters affect them in the same manner. One might say that after all device fluctuations are compensated, an additional 1 cps independent fluctuation remains.

More meaningful are the results of frequency stability investigations which involve the observation of the beat note obtained by mixing the outputs of two independently oscillating lasers. To observe this signal, it is essential not only that the same area of the photocathode of the multiplier phototube be exposed to the two laser beams but that the wavefronts of the two beams be parallel. This alignment is achieved by a mirror system shown in Fig. 84. In an experiment designed to achieve the greatest freedom from external fluctuations the lasers were supported on a massive shock-mounted table with resonant frequencies of many seconds and located in a cellar room of an isolated building. The difference signals of the two lasers operating at $2.6 \times 10^{14}$ cps ($1.15 \mu$) were observed around 1700 cps. Fluctuations of the beat note indicated a short-term frequency fluctuation of only 20 cps for the individual lasers, or 8 parts in $10^{14}$ [19].

REFERENCES

The observation of stimulated emission in noble gases requires a laser structure described in Section V.4 in connection with the He-Ne laser. Interchangeable mirrors are usually provided, each set peaked for reflection in a narrow spectral region. Such mirrors enable the experimenter to concentrate on a relatively narrow region of the spectrum and avoid the excitation of unwanted oscillations. The spectral selectivity of the apparatus may be further increased by incorporating a dispersive prism between the discharge tube and one of the reflectors, which must then be placed perpendicular to the rays deviated by the prism. The experiments generally require longer tubes than those used for the excitation of the common...
He-Ne lines because the gain obtainable is generally lower. Many lines were discovered with tubes 2.25 meters long. The occurrence of stimulated emission is usually inferred from the directional characteristics of the radiation emerging from the ends of the tube and from the effects on the output intensity of the blocking of the mirror opposite the output.

Noble gas lasers have He, Ne, Ar, Kr, and Xe atoms or ions as their working material. We shall discuss the operation of lasers based on radiation emitted from neutral atoms first. For a tabulation of these lines see Appendix A.2.

*Helium* was observed to emit a 2.0603-μ line in a $7^3D \rightarrow 4^3P$ transition [1, 2], and a 1.9543-μ line in a $4^3P \rightarrow 3^3D$ transition [3]. The conditions for the stimulated emission of these two lines are naturally different; in one case the population of the $4^3P$ level should be maximized, in the other minimized. The optimal pressure for the excitation of the 2.06-μ line is 8 torr, for that of the 1.95-μ line 0.3 torr.

*Neon* lines observed in stimulated emission number about 130. Some of these lines were reported to have been observed in He-Ne mixtures only, but generally the Ne lines which are found in these mixtures can be produced in pure Ne also, provided that longer tubes are used and precautions are taken to minimize losses. In principle any spectral line is a potential laser line provided that a practical way is found to create a population inversion for the relevant pair of levels. Consequently the task of making a list of "laser lines" is like the task of making a list of cities with parking problems. All cities can get on the list if drivers can be induced to congregate in them. In the case of neon the creation of population inversion is facilitated by certain near-coincidences between He and Ne levels. Some of these were already introduced in Section V.4. The pertinent numerical data are as follows:

<table>
<thead>
<tr>
<th>He level</th>
<th>Neon level group</th>
</tr>
</thead>
<tbody>
<tr>
<td>Symbol</td>
<td>Energy (cm⁻¹)</td>
</tr>
<tr>
<td>$2^3S$</td>
<td>159,850</td>
</tr>
<tr>
<td>$2^1S$</td>
<td>166,272</td>
</tr>
<tr>
<td>$2^1P$</td>
<td>171,129</td>
</tr>
</tbody>
</table>

Transfer of excitation from He materially aids the operation of lasers with transitions originating at some of the $2s$, $3s$, and $6p$ levels of Ne. The visible 6328-Å laser and the strong 3.39-μ laser belong in this group together with the fourteen near infrared laser lines resulting from $2s \rightarrow 2p$ transitions. In the far infrared region six lines were found between 35.6 and 37.4 μ which result from $6p \rightarrow 6d$ transitions. Stimulated emission in all these lines is aided by the admixture of He. On the other hand the presence of He tends to inhibit lines originating at $5p$ (Paschen) levels [4].
Neon laser lines between 1 and 20 \( \mu \) are shown graphically in Figs. 85 and 86. The visible lines lie outside the range of the graph, as do numerous lines between 20 and 133 \( \mu \). Most of the neon laser lines were observed by Faust, Garrett, McFarlane, and Patel of Bell Telephone Laboratories. Their publications contain a wealth of information about stimulated emission from all noble gases [4, 5, 6, 7].

The laser lines originating from complete atoms of Ar, Kr, and Xe are also shown in Figs. 85 and 86 with the single exception of the 26.944-\( \mu \) line of Ar. Many laser lines of the heavier gases originate at \( d \) levels, while in Ne the lower-lying \( s \) and \( p \) levels seem to be favored as starting levels.

![Diagram of Noble Gas Laser Lines](image)

**Fig. 85.** Noble gas laser lines in the near infrared (1.0 to 4.0 \( \mu \)). Number of closely spaced lines is indicated when they cannot be shown separately. (For He lasers see ext.)
Two Xe lasers are worth special notice because of their high gain. In a mixture of Xe (0.02 torr) and He (2 torr) contained in a 5- to 7-mm diameter tube a gain of 1 per cent per cm was obtained at 2.0262 \mu [8]. There is no transfer of excitation from He; the improvement caused by He is due solely to an increase in the electron density in the discharge. Under similar operating conditions a gain of 13 per cent per cm is obtained at 3.507 \mu [9].

Laser oscillations have been observed on many lines of the ionized noble gases. The noble gas ion lasers cover the entire visible spectrum and extend into the near ultraviolet. This copious treasure of visible lasers was discovered almost simultaneously by Bridges at Hughes Research Laboratories [10] and by Gordon and Labuda at Bell Telephone Laboratories. The joint publication of their results represents a notable instance of
scientific cooperation among competing laboratories [11]. Additional lines were found by Dana and Laures [12].

Excitation of ionic laser lines requires a discharge with a current density much larger than that necessary for the excitation of the atomic lasers. The higher density is necessary because in singly ionized gas lasers the upper laser level is probably populated by two successive electron collisions; the first produces an unexcited ion from the neutral atom, while the second

![Diagram of Noble Gas Laser Lines](image)

**Fig. 87.** Noble gas ionic laser lines [12, 14]. >: strong line. 2: two lines within 10 Å. (A 7993-Å line of Kr II is not shown.)
excites the ion to the upper laser level. This two-step process is deduced from the observed proportionality of the spontaneous emission from the upper level to the square of the discharge current.

The laser configuration is similar to the one used for neutral noble gases. Provision must be made, however, for the passage of very high currents through the discharge tube. This is accomplished by means of a d-c discharge with a heated cathode. Usually the operation is intermittent; the discharge can be powered using a capacitor charged to a potential of 2 to 10 kV. Maximum currents of 40 to 300 amperes were used with tubes around 4 mm in diameter [10, 13, 14]. Continuous operation is possible in many lines with smaller currents passing through narrower tubes [11].

Figure 87 shows the ionic noble gas lasers following the compilation of Bridges and Chester [14], whose paper contains a detailed analysis of the transitions believed responsible for the observed oscillations. The notations of the figure follow the standard spectroscopic practice of designating with Roman numerals II, III, and IV the singly, doubly, and triply ionized spectra, the numeral I being reserved for the spectrum of the neutral atom. The figure is to be taken as an illustration of the state of knowledge near the end of 1964. The rate of discovery of ionic laser lines is rapid and the identification of many lines is subject to revision. It is therefore impractical to print tables of ionic laser lines in a book to be published in 1965. For a tabulation and classification of such lines the reader is referred to the cited paper of Bridges and Chester as well as to their review article [15].

REFERENCES
6. MISCELLANEOUS GAS LASERS

Electric discharges in gases have produced a large variety of laser lines in many substances. The discovery of lasers outside the noble gas group began in 1963; during the year 1964 new materials and lines were reported every month. The knowledge available about these lasers varies over a wide range. At best not only is the line identified but a model is available which explains the occurrence of population inversion on the basis of the physical processes taking place in the discharge, taking into account the rates of the processes involved. In a less favorable situation the transition responsible for the line is identified and general conjectures are available concerning the processes that contribute to the inversion of population of the relevant levels. There are laser lines whose origin is in doubt, most frequently because the measurements of wavelength were not carried out with an accuracy sufficient to distinguish between two transitions of nearly equal energy difference. At worst, there is doubt even concerning the element from which the line originates. Very intensive work is in progress in this field and it seems too early to attempt a systematic organization of this subject in 1964. We shall therefore confine ourselves to a rather sketchy summary of the principal observations.

The material is divided into the following groups:

1. Laser lines in C, N, and O obtained on dissociation of diatomic and polyatomic molecules.
2. Laser lines in atoms and ions of halogens, S, and Hg; lines of ionized atoms. In these substances direct electron impact seems to be the essential factor.
3. Laser lines associated with molecular transitions.

Dissociative Excitation

The principle of dissociative excitation on collision with excited atoms has already been discussed in Section V.1. The best-explored systems in
which this process leads to population inversion and laser action are Ne-O₂ and Ar-O₂. The processes are different for the two noble gases; both are illustrated in Fig. 88. The energy levels of oxygen are shown in this figure as reckoned from the lowest bound state of the O₂ molecule, which is 5.080 eV below the ground state of the two O atoms removed from each other. The lowest excited (1s) states of Ne and Ar are also shown on the diagram; they are indicated as Ne* and Ar*. Transfer from the Ne* levels leads directly to an unstable molecular level from which the molecule dissociates with one of the oxygen atoms in the starting level (3p³P₂) of the laser. In the case of Ar the transfer takes place to two lower-lying levels: 2p¹D₂ and 2p¹S₀. These are metastable; radiative transition from these levels to the triplet ground level is forbidden by the selection rules. Because of their metastable nature, these levels become heavily populated, they become platforms from which the 3p³P₂ level can be reached by electron impact. The selection rules indicate that population inversion between the

Fig. 88. Energy levels of oxygen, neon, and argon participating in the Ne-O₂ and Ar-O₂ lasers. (Curves represent energies of a common electronic state.)
Miscellaneous Gas Lasers

$3p^3P_2$ and $3s^3S_1^o$ levels cannot be established by electron collision alone, i.e., without the assistance of a noble gas, because the transition from the ground $3p^3P_2$ level to the $3s^3S_1^o$ level is more likely than a transition to the $3p^3P_2$ level, the latter being forbidden by the parity rule. The selection rule is directly applicable to the so-called electric dipole transition probabilities only, but it can be shown that electron excitation probabilities are roughly proportional to probabilities of electric dipole transitions.

Bennett, Faust, McFarlane, and Patel [1], who discovered the OI laser and analyzed the processes involved, obtained stimulated emission of the 8446-Å line from an rf discharge in a tube 2 meters long and 7 mm in diameter. The composition of the gas was O$_2$, 0.014 torr; Ne, 0.35 torr; or alternatively O$_2$, 0.036 torr; Ar, 1.3 torr. The output power was about 2 mW in each case. The same radiation is also obtainable upon the dissociation of CO, CO$_2$, NO, and N$_2$O in the presence of noble gases [2]. Laser radiation of the CI and NI lines may also be obtained from these compounds. The characteristics of the lines observed are as follows [2]:

<table>
<thead>
<tr>
<th>Spectrum</th>
<th>Wavelength</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>CI</td>
<td>1.0689 µm</td>
<td>$3p^3D_3 \rightarrow 3s^3P_2^o$</td>
</tr>
<tr>
<td>CI</td>
<td>1.4539 µm</td>
<td>$3p^3P_1 \rightarrow 3s^3P_1^o$</td>
</tr>
<tr>
<td>NI</td>
<td>1.3583 µm</td>
<td>$3p^2S_{1/2}^o \rightarrow 3s^2P_{3/2}^o$</td>
</tr>
<tr>
<td>NI</td>
<td>1.4544 µm</td>
<td>$4s^4P_{5/2} \rightarrow 3p^2D_{5/2}^o$</td>
</tr>
</tbody>
</table>

The details of the processes which generate these radiations are not as well explored as those which produce the 8446-Å oxygen line.

Miscellaneous Atomic Lasers

A large number of laser lines have been observed in discharges containing the mixture of an ordinary gas and a noble gas. In some instances an energy transfer from a noble gas to a molecule may be involved, but this is not established with any degree of certainty. It is quite possible that the beneficial effect of the noble gas is more indirect. We confine ourselves to a summary of the experimental results, noting that in most of the cases described the direct electron impact seems to play a major role in the creation of population inversion.

**Sulfur.** Two lines of S I were obtained from a mixture of SF$_6$ (0.03 torr) and He (2 torr), or with less efficiency from SF$_6$ alone [2]:

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0455 µm</td>
<td>$4p^3P_2 \rightarrow 4s^3S_1^o$</td>
</tr>
<tr>
<td>1.0628 µm</td>
<td>$4p^1F_3 \rightarrow 4s^1D_2^o$</td>
</tr>
</tbody>
</table>

**Halogens.** The structure of the complete halogen atoms is identical to that of the singly ionized noble gases; therefore the discussion of Section
V.2 is applicable to them. We confine the detailed discussion to the laser lines of complete halogen atoms and refer the reader interested in ionic lines to the review article of Bridges and Chester entitled "Spectroscopy of Ion Lasers."*

Paananen, Tang, and Horrigan [3] observed two laser lines in an rf-excited discharge in a chlorine-helium mixture. The observations were made in a 1.75-meter-long, 6-mm-diameter tube; the optimal pressures were chlorine 0.1 torr, helium 1.5 torr. Bockasten [4] classified the observed lines as follows:

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.9755 µ</td>
<td>$3d^4 D_{7/2} \rightarrow 4p^4 P_{5/2}$</td>
</tr>
<tr>
<td>2.0199 µ</td>
<td>$3d^4 D_{5/2} \rightarrow 4p^4 P_{3/2}$</td>
</tr>
</tbody>
</table>

McFarlane [5] obtained nine laser lines of CIII in a pulsed discharge operating a 1-meter-long tube with a peak current in excess of 500 amperes. The wavelengths extend from 4781 Å to 6095 Å and the radiation is attributed to a series of $4p \rightarrow 4s$ transitions.

The iodine laser lines observed by Rigden and White [6] were classified by Bockasten [4] transitions in I\(_2\):

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.236 µ</td>
<td>$5d(2)<em>{5/2} \rightarrow 6p[1]</em>{3/2}$</td>
</tr>
<tr>
<td>3.431 µ</td>
<td>$5d(4)<em>{7/2} \rightarrow 6p[3]</em>{5/2}$</td>
</tr>
</tbody>
</table>

Fowles and Jensen [7, 8] observed 12 laser lines from a pulsed discharge in an iodine-helium mixture. These lines extend from 4987 Å to 8804 Å and are attributed to I\(_2\).

In an rf discharge containing 0.09 torr Br and 1.8 torr Ar, laser action takes place at four very closely placed wavelengths between 8446 Å and 8447 Å. This may arise as a consequence of an excitation transfer from Ar to the Br\(_2\) molecule similar to the process encountered in O\(_2\)[2]. The laser transition is $5p^{4}D_{3/2} \rightarrow 5s^{4}P_{3/2}$.

**Mercury.** A variety of laser oscillations can be obtained from electric discharges containing mercury and a noble gas. The radiation is generated by stimulated transitions in complete mercury atoms and by similar transitions in singly or doubly ionized mercury. The discharges are produced in tubes similar to those described in connection with noble gas lasers; internal electrodes and d-c excitation are recommended, but rf excitation of HgI lines is possible. The stronger lines can be produced in

* Ref. 15, Section V.5, p. 207.
† The notation for the iodine levels is the one used in the most complete tables of Minnhagen (Ark. f. Fysik 21, 415, 1962).
tubes about 7 mm in diameter and 1 meter long; weaker lines require longer tubes.

The optimum pressure for HgI lasers is 0.2 to 0.3 torr Hg and about 1 torr noble gas; HgII and HgIII lasers operate best at about 0.001 torr Hg and 0.5 torr He or other noble gas. The mercury pressure is regulated by heating the tube to the required moderate temperature. The high current (10 to 50 amperes) required for the ionic lasers is usually produced by discharging a condenser through the tube. The different laser lines do not appear simultaneously; their evolution indicates that different processes progressing at different rates are responsible for the population inversions which produce them.

The first Hg lasers were described by Rigden and White [6]; the most accurate spectroscopic work in this field was done by groups of investigators at the Cie Générale de Télégraphie [9, 10], Raytheon Co. [11], and Spectra Physics [12]. The literature contains inaccurate data and erroneous conclusions concerning the transitions that may be involved. A critical examination of the published material and the application of spectroscopic selection and intensity rules leads to the following classification of the HgI lines observed in stimulated emission [13]:

<table>
<thead>
<tr>
<th>Wavelength*</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1177 μ</td>
<td>7p¹P₁⁰ → 7s³S₁</td>
</tr>
<tr>
<td>1.3674 μ</td>
<td>7p³P₁⁰ → 7s³S₁</td>
</tr>
<tr>
<td>1.5296 μ</td>
<td>6p'³P₂⁰ → 7s³S₁</td>
</tr>
<tr>
<td>1.6920 μ</td>
<td>5f¹F₃⁰ → 6d¹D₂</td>
</tr>
<tr>
<td>1.6942 μ</td>
<td>5f³F₂⁰ → 6d³D₁</td>
</tr>
<tr>
<td>1.7073 μ</td>
<td>5f³F₄⁰ → 6d³D₃</td>
</tr>
<tr>
<td>1.7110 μ</td>
<td>5f³F₅⁰ → 6d³D₂</td>
</tr>
<tr>
<td>1.7330 μ</td>
<td>7d¹D₂ → 7p¹P₁⁰</td>
</tr>
<tr>
<td>1.8130 μ</td>
<td>6p¹F₄⁰ → 6d³D₃</td>
</tr>
<tr>
<td>3.93 μ</td>
<td>5g G → 5f F⁰</td>
</tr>
<tr>
<td>5.86 μ</td>
<td>6p'¹P₁⁰ → 7d³D₂</td>
</tr>
<tr>
<td>6.49 μ</td>
<td>9s¹S₀ → 8p¹P₁⁰</td>
</tr>
</tbody>
</table>

A 3.34-μ line observed in a Hg-Kr mixture and erroneously attributed to HgI is probably the 3.341-μ KrI line.

* The values listed are wavelengths in air consistent with energy levels calculated from spontaneous emission spectroscopy. Exceptions are made for the tenth and twelfth lines of Hg. For these lines, whose classifications are uncertain, the best experimental values are listed.
At least ten laser lines were observed in pulsed mercury discharges which are definitely attributable to mercury ions.

**Ions of Atmospheric Gases.** About thirty laser lines have been obtained in high-current pulsed discharges in various constituents of air [14]. They were identified as OII, OIII, CII, CIV, NII, NIII, and NIV lines. Their wavelengths cover the entire visible spectrum.

**Transitions in Molecular Spectra**

Stimulated emission has been observed in the band spectra of N\(_2\), CO, and CO\(_2\). They are excited in high-current, pulsed discharges. The oscillations in N\(_2\) are very strong; they can be excited in tubes only 40 cm long with gas pressure over 1 torr. The N\(_2\) lines are parts of the band spectrum; they fall into six groups in the following wavelength regions: 7580–7620 Å, 7700–7750 Å, 8683–8710 Å, 8844–8910 Å, 10,450–10,505 Å, 12,300–12,350 Å. A peak power output of 100 watts was obtained from the first two groups of oscillations [15, 16].

Four groups of CO radiation were observed in stimulated emission in the following regions: 5186–5198 Å, 5590–5604 Å, 6063–6074 Å, 6595–6614 Å. Peak power output varied from group to group with a maximum of 8 watts in a group, a value which can probably be exceeded by optimizing experimental conditions [16, 17].

Similar laser action at 9.40 \(\mu\) and 10.41 \(\mu\) has been obtained from CO\(_2\) [18].

These molecular laser lines seem to have their origin in nonstationary population inversions. Their mechanisms are very actively investigated.

**REFERENCES**