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PULSED GAS-DISCHARGE LASERS

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The present review of the investigations of pulsed gas-discharge lasers is based mainly on the literature published up to the middle of 1970. Lasers utilizing transitions in neutral and ionized atoms and electronic transitions in molecules are discussed. The attention is concentrated on the systems whose efficiency is likely to be improved. The factors which determine the efficiency and the peak output power are considered. The choice of transitions likely to give high efficiencies and high peak powers is justified. A summary is given of the available information on pulsed gas-discharge lasers of two types: the lasers utilizing transitions from resonance to metastable levels in neutral and ionized atoms and the lasers using electronic transitions in diatomic molecules. The lasers using thallium, copper, and lead vapors and those utilizing transitions in nitrogen molecules are discussed in detail. Practical difficulties encountered in the construction of high-efficiency pulsed lasers are analyzed. A list is given of the transitions in other neutral and ionized atoms and molecules which can be expected to provide high-power pulsed emission. The prospects of utilization of the transitions from a resonance electronic level to the ground state of a molecule are stressed. The possibility of advance into the still unmastered vacuum ultraviolet region is discussed. Possible effects of increasing the density of the active gas are considered. Ways of going over from pulsed to continuous-wave emission in collision lasers are analyzed. It is concluded that pulsed gas-discharge lasers offer means for generating high-efficiency (up to 10%) and high-peak-power coherent pulses in a very wide range of wavelengths. It should also be possible to achieve high-efficiency continuous-wave emission in collision lasers operating at short wavelengths.

CONTENTS

1. INTRODUCTION

PULSED gas-discharge lasers represent one of the most important and numerous classes of lasers. Pulsed emission has been achieved for several hundreds of lines extending over a very wide range from vacuum ultraviolet (about 1600 Å) to submillimeter wavelengths (0.79 mm). Laser action has been obtained utilizing transitions in neutral and ionized atoms and in molecules. A variety of methods is used to establish population inversion. Considerable peak output powers (of the order of megawatts) and very high gains (up to 600 dB/m) have been attained. Some applications have been found for these lasers.

It must be stressed that pulsed laser action can be attained in a much wider range of active media, transitions, and wavelengths than is possible in continuous-wave (cw) emission. This situation arises because the conditions which are difficult or impossible to achieve in cw emission (high pumping powers, efficient heat removal, etc.) can be realized relatively easily under pulsed conditions. Consequently, investigations of pulsed laser action frequently provide conditions under which physical processes in the active medium can be followed more easily and the possibility of achieving population inversion can be determined more readily. This is why in many systems (for example, ion lasers) the laser action has been achieved first under pulsed conditions. Moreover, population inversion in pulsed lasers can be attained even when steady-state inversion is quite impossible.

It is known that in order to maintain a steady-state inversion corresponding to a transition $i \rightarrow k$ not only the upper laser level must be pumped selectively but also the probability of the laser transition A_{ik} must be less than the total probability of relaxation of the lower level A_k. This requirement imposes serious restrictions in the selection of transitions suitable for the maintenance of steady-state inversion. In practically all the cw gas lasers operating in the visible, ultra-

violet, and near infrared regions the relaxation of the lower laser level is the result of spontaneous emission. An analysis reported in[1,2] shows that the spontaneous-emission relaxation of the lower level ensures the necessary relaxation rate but imposes serious limitations on the output power and the efficiency. These limitations due to the fact that the laser transition involving spontaneous-emission relaxation of the lower level must necessarily occur between relatively high-lying levels. These difficulties can be overcome in the "collision" lasers in which the lower level relaxes as a result of collisions with heavy particles.[1,2] Such lasers have been developed only for the middle infrared range and--as predicted--they are characterized by high efficiencies and high output powers. The best example of a collision system is the well-known CO_2 laser.[3-5]

The relaxation of the lower level is unnecessary if the pulsed laser action takes place at the beginning of an excitation pulse. This extends considerably the range of suitable laser transitions and opens up a possibility of development of high-performance lasers (in particular, high-efficiency and high-peak-power lasers) operating in a wide range of wavelengths.

A very large number of papers has been published on the development and properties of pulsed gas-discharge lasers. The accumulated extensive experimental data have revealed the basic features of the physical processes leading to population inversion in the majority of promising classes of pulsed lasers. It therefore seems timely to make a systematic analysis of the results obtained and to establish the main tendencies and prospects in the development of pulsed gas-discharge lasers. No reviews of this subject have yet been published in the Soviet or foreign literature. The original literature on pulsed gas lasers is so extensive that it is unrealistic to attempt a comprehensive review in a single paper. The most urgent problem facing investigators in gas lasers is the improvement of the efficiency, particularly at short wavelengths. In view of this the present review concentrates mainly on systems showing a promise of higher efficiency and on ways and prospects of development of high-efficiency lasers. For reasons of space, the properties and physical mechanisms active in such lasers are discussed only briefly but the literature for each system is listed as fully as possible. The review is based mainly on investigations which appeared up to the middle of 1970. The most important results of later papers are included in tables and are discussed briefly in a note which follows conclusions.

2. LIMITING CHARACTERISTICS OF PULSED GAS-DISCHARGE LASERS

The efficiency of a cw laser[2] can be written in the form $\eta = f_p(h\nu_L/E_1)$, where $h\nu_L$ is the energy of a photon generated in the laser transition and E_1 is the energy of the other laser level, which will be denoted by 1. The ratio $h\nu_L/E_1$ is the fraction of the energy of the upper laser level which is utilized in the laser action. The quantity f_p represents that fraction of the pumping energy which is utilized in the excitation of the upper laser level. Since relatively high-lying levels (E_1 of the order of 15—30 eV) are utilized in the majority of cw gas lasers, the ratio $h\nu_L/E_1$ rarely exceeds 0.1. Moreover, the use of such high levels reduces also the value of f_p. Most of the energy evolved under typical conditions in a gas-discharge plasma is lost in the excitation of the lowest levels and in ionization processes. Only about 1% of the pumping energy is utilized in the excitation of the higher laser level. Consequently, a typical efficiency of a cw gas-discharge laser is 10^{-3}--10^{-4}. High values of the cw efficiency have been achieved only for lasers utilizing low-lying vibrational levels.

In atomic systems the major part of the discharge energy is normally expended in the excitation of the first resonance level. For example, about 60% of the total energy consumed in a steady-state discharge in mercury is used, under typical conditions, in the excitation of the resonance level.[6,7] The first resonance level usually has the largest electron-impact excitation cross section. Therefore, it would be desirable to use the first resonance level as the upper laser level. In atomic systems (consisting of neutral and ionized atoms) only a metastable level lying below the first resonance can act as the lower laser level. The utilization of a metastable level as the lower laser should make it possible to attain efficient population inversion under pulsed conditions because the electron-impact excitation cross sections of the forbidden transitions are usually much smaller than those of the allowed transitions. Thus, in a typical transition from a resonance to a metastable level we may expect to achieve efficient pulsed population inversion at the beginning of the excitation pulse. The selection of transitions suitable for efficient pulsed laser action is considered in[8].

Pulsed laser action involving transitions terminating in metastable levels is frequently called self-terminating (self-terminating transitions) because its duration is limited by the properties of the transition itself. The expression for the efficiency of such lasers is somewhat different. We must make allowance for the fact that the stimulated emission lasts only to the moment at which the populations of the two laser levels become equal. Thus, part of the population of the upper level is unused: this part depends on the ratio of the statistical weights of the upper g_1 and the lower g_2 levels. An allowance for this factor gives

$$\eta = f_p \frac{h\nu_L}{E_1} \frac{g_2}{g_2+g_1} = f_p \eta_{lim}.$$

The quantity η_{lim} will be called the limiting efficiency of the transition, i.e., the efficiency which can be attained when all the energy is used to excite the upper level. The factor $g_2/(g_2 + g_1)$ is always of the order of unity: usually it ranges from 2/3 to 1/4. However, the factor $h\nu_L/E_1$ can be considerably higher than under steady-state conditions. In the case of atoms with low-lying lower levels this factor can reach 0.5—0.7. Although the value of η_{lim} can always be calculated, the factor f_p is difficult to estimate because it depends strongly on the experimental conditions. For resonance levels the value of f_p may be of the order of 0.5. If we multiply all the factors in the above expression, we find that the efficiency may reach $\eta \approx 25\%$. The possibility of achieving such a high efficiency is of

very considerable interest. However, in order to realize this efficiency in a pulsed laser, we must satisfy additional conditions. The duration of the excitation pulse must be of the order of the inversion lifetime. Moreover, a considerable energy is needed to generate active plasma in a pulsed laser. Rough estimates show that the energy consumed in the ionization and the heating of the electrons in a plasma may be comparable with the energy required for the excitation of the laser level. In view of these difficulties and those encountered in the selection of atoms with an energy-level scheme ideal for population inversion, we may realistically expect a laser utilizing transitions from a resonance to a metastable level to have an efficiency of up to 10%.

If the pumping rate is well above the threshold value, the output power of laser is determined by the rate of pumping of the upper laser level. In the case of electron-impact excitation from the ground state the pumping rate is $Q = N_0\langle\sigma v_e\rangle n_e$, where N_0 is the population of the ground state, $\langle\sigma v_e\rangle$ is the product of the cross section and the electron velocity averaged out over this velocity, and n_e is the electron density. If we assume that $\sigma \approx 10^{-16}$ cm^2, $v_e = 10^8$ cm/sec, $n_e \approx 10^{15}$ cm^{-3}, $N_0 \approx 10^{17}$ cm^{-3}, we find that the rate of excitation is of the order of 10^{24} events-sec^{-1} cm^{-3}. This corresponds to a power density of 10^5 W/cm^2 in the case of laser emission in the visible region. The parameters just quoted represent conditions which are easily realizable in a pulsed discharge. It may be possible to increase the value of N_0 in future. Our estimate shows that high peak powers may be expected from pulsed gas lasers. Thus, a laser with an active medium of 1000 cm^3 volume should be capable of generating peak powers of the order of 100 MW.

Theoretical calculations of the pulsed inversion and of the saturated output power of systems with self-terminating transitions are reported in[9-17]. The method for calculating the saturation power, suggested in[9], is particularly important in analysis of pulsed operation of lasers. This method is based on the assumption of full saturation of the population inversion and of equality of the populations of the laser levels during an output pulse. This makes it possible to simplify considerably the mathematical treatment. In spite of this simplification the precision of the method is satisfactory for practical applications because the results obtained are in good agreement with those obtained by a more rigorous method[10] and with the experimental results. The simplified method has been used to calculate the saturation power of ultraviolet radiation resulting from transitions in nitrogen molecules,[9,10,16] of infrared radiation resulting from transitions in the same molecule,[13] and of radiation resulting from transitions in copper[11] and thallium.[15] Calculations showed that the duration of inversion and emission is of the order of the lifetime of the upper level when the electron density is reasonably high. When the electron density is such that the probability of transitions as a result of collisions with electrons become comparable with the probability of spontaneous transitions, the output pulse becomes shorter. In practice the duration of the output pulse is always much shorter than the lifetime of the upper level and it frequently amounts to 10^{-9}—10^{-8} sec. The durations of output pulses generated by specific laser systems will be given later.

This discussion shows that pulsed gas lasers are capable of high-efficiency and high-peak-power generation of coherent radiation in a wide range of wavelengths. The practical attainment of such high performance characteristics depends on the extent to which the existing systems can be made to approach the theoretical limits outlined above. The first task is to select the most favorable active media and transitions. Moreover, we must consider the problem of practical realization of the excitation conditions necessary to achieve high performance characteristics. These problems and the descriptions of working systems will be discussed in subsequent sections.

3. PULSED LASERS UTILIZING TRANSITIONS IN NEUTRAL ATOMS

We have explained already that most efficient laser action can be achieved if we utilize transitions from the first resonance level. Then, the lower laser level of an atom can only be a metastable level. A considerable number of transitions of this type is known to occur in atoms, as confirmed experimentally by the attainment of laser emission or of superradiance. Table I lists the characteristics of these transitions. The transitions characterized by the highest limiting efficiency are given in Table II. This table includes the level parameters and the principal experimentally determined characteristics of laser action. Unfortunately, no data are available on the electron-excitation cross sections of the resonance levels of atoms of interest to us. Therefore, Table II gives the values of the maximum excitation cross sections of the upper laser level, σ_m, calculated using an approximate formula which is valid for allowed transitions:[18] $\sigma_m \approx (2f/\epsilon^2)\pi a_0^2$; here, f is the oscillator strength of the transition, ϵ is the transition energy in rydbergs, and πa_0^2 is the atomic cross-section unit. The listed values of the cross sections can be used to estimate approximately the pumping rates of the upper laser level. We can see from Table II that the highest limiting efficiency $\eta_{lim} = 47\%$ is exhibited by the green line of thallium. A somewhat lower value of η_{lim} is exhibited by some transitions in lead and copper atoms. The highest peak power of 40 kW has been achieved for the green line of copper.[22,24] The highest practical efficiency (1.2%) has also been achieved for copper.[22,24] The experimental results for the green line of neon are included in Table II for the sake of comparison. In spite of the fact that η_{lim} for this line is considerably lower than the corresponding efficiencies for the lines of thallium, lead, and copper, the peak power achieved for the neon line is considerably higher (190 kW).[33] The reason for this discrepancy will be discussed later.

We shall now consider in greater detail the energy level schemes and the characteristics of laser action in the three atomic systems characterized by highest values of the limiting efficiencies.

a) Thallium

Figure 1 shows the lower energy levels of the thallium atom. Thallium has two resonance levels: $7^2S_{1/2}$

Table I. Experimentally observed laser action in transitions from resonance to metastable levels in atoms and ions

λ, Å	Atom	Transition	Reference	η_{lim}, %	T, °C
3639	Pb I	$6p7s^3P_1^0 \rightarrow 6p^2\,^3P_1$	19	39	1000
3984	Hg II	$6p^2P_{3/2}^0 \rightarrow 6s^2\,^2D_{5/2}$	20	25 (I)*	70
4057	Pb I	$6p7s^3P_2^0 \rightarrow 6p^2\,^3P_2$	19	44	1000
4062	Pb I	$6p6d^3D_1^0 \rightarrow 6p^2\,^1D_2$	19	33	1000
5105	Cu I	$4p^2P_{3/2}^0 \rightarrow 4s^2\,^2D_{5/2}$	8, 11, 21–24	38	1500
5341	Mn I	$y^6P_{7/2}^0 \rightarrow a^6D_{9/2}$	8, 25	29	1200
5350	Tl I	$7^2S_{1/2} \rightarrow 6^2P_{3/2}$	15, 26, 30	47	800
5420	Mn I	$y^6P_{5/2}^0 \rightarrow a^6D_{7/2}$	8, 25	30	1200
5470	Mn I	$y^6P_{5/2}^0 \rightarrow a^6D_{5/2}$	8, 25	25	1200
5517	Mn I	$y^6P_{3/2}^0 \rightarrow a^6D_{3/2}$	8, 25	25	1200
5538	Mn I	$y^6P_{3/2}^0 \rightarrow a^6D_{1/2}$	8, 25	17	1200
5782	Cu I	$4p^2P_{1/2}^0 \rightarrow 4s^2\,^2D_{3/2}$	8, 21–24	38	1500
6278	Au I	$6p^2P_{1/2}^0 \rightarrow 6s^2\,^2D_{3/2}$	24	29	1500
7229	Pb I	$6p7s^3P_1^0 \rightarrow 6p^2\,^1D_2$	19, 27–29	24	1000
8542	Ca II	$4p^2P_{3/2}^0 \rightarrow 3d^2D_{5/2}$	8	28 (I)	700
8662	Ca II	$4p^2P_{1/2}^0 \rightarrow 3d^2D_{3/2}$	8	31 (I)	700
10330	Sr II	$5p^2P_{3/2}^0 \rightarrow 4d^2D_{5/2}$	31	25 (I)	500
10918	Sr II	$5p^2P_{1/2}^0 \rightarrow 4d^2D_{3/2}$	31	27 (I)	500
12900	Mn I	$z^6P_{7/2}^0 \rightarrow a^6D_{9/2}$	8, 25	14	1200
13294	Mn I	$z^6P_{7/2}^0 \rightarrow a^6D_{7/2}$	8, 25	15	1200
13319	Mn I	$z^6P_{5/2} \rightarrow a^6D_{7/2}$	8, 25	17	1200
13627	Mn I	$z^6P_{5/2} \rightarrow a^6D_{5/2}$	8, 25	15	1200
13864	Mn I	$z^6P_{3/2}^0 \rightarrow a^6D_{3/2}$	8, 25	14	1200
13997	Mn I	$z^6P_{3/2}^0 \rightarrow a^6D_{1/2}$	8, 25	9	1200
20583	He I	$2^1P_1 \rightarrow 2^1S_0$	26	0,7	20
55460	Ca I	$4p^1P_1^0 \rightarrow 3d^1D_2$	31	4,8	600
64560	Sr I	$5p^1P_1^0 \rightarrow 4d^1D_2$	31	4,5	500

*I denotes that η_{lim} was calculated for the excitation from the ground state of the ion.

Table II. Atomic and ionic systems with highest limiting efficiency

Atom	Transition	λ, Å	σ_m, 10^{-16} cm²	τ_1, nsec	τ_L, nsec	η_{lim}	Experiment			
							P_{peak}, W	P_{peak}/N, W/cm³	Δt, nsec	T, °C
Tl I	$7^2S_{1/2} \rightarrow 6^2P_{3/2}$	5350	4.0	7.6	15	0.47	300	300	1—3	800
Pb I	$6p7s^3P_1^0 \rightarrow 6p^2\,^3P_2$	4057	3.7	6	9	0.44			< 10	1000
	$6p7s^3P_1^0 \rightarrow 6p^2\,^3P_1$	3639	3.7	6	31	0.39			< 10	1000
	$6p6d^3D_1^0 \rightarrow 6p^2\,^1D_2$	4062	3.8	4	9*)	0.33			< 10	1000
	$6p7s^3P_1^0 \rightarrow 6p^2\,^1D_2$	7229	3.7	6	250*)	0.24	$2 \cdot 10^3$	250	15	1030
Cu I	$4p^2P_{3/2}^0 \rightarrow 4s^2\,^2D_{5/2}$	5105	9.7	7.2	770	0.38	$40 \cdot 10^3$	25	16	1500
	$4p^2P_{1/2}^0 \rightarrow 4s^2\,^2D_{3/2}$	5782	4.5	7.2	370	0.38				
Mn I	$y^6P_{5/2}^0 \rightarrow a^6D_{7/2}$	5420	2.1*	9*)	435*)	0.30	100	1	20	1200
	$y^6P_{7/2}^0 \rightarrow a^6D_{9/2}$	5341	2.0*	9.6*)	300*)	0.29				
Ca II	$4p^2P_{1/2}^0 \rightarrow 3d^2D_{3/2}$	8662	1.5	6.7	770	0,31 (I)**	30		30	700
	$4p^2P_{3/2}^0 \rightarrow 3d^2D_{5/2}$	8542	1.5	6.7	830	0.28 (I)				
Ne I	$2p_1 \rightarrow 1s_4$	5400		14.4	1430	0.09	$190 \cdot 10^3$		1,5	20

*The transition probabilities were taken from [38].
**I denotes that η_{lim} was calculated for the excitation from the ground state of the ion.

and $6^2D_{3/2}$. The oscillator strengths of the transitions from these levels are 0.13 and 0.30, respectively.[34] No published data are available on the electron-excitation cross sections of the thallium levels. If we use the aforementioned approximate formula for the maximum cross sections, we find that the excitation cross sections of both resonance lines of thallium are approximately equal. Therefore, we may expect the rates of excitation of these two resonance levels to be comparable. The transition from the $6^2D_{3/2}$ level to the metastable level $6^2P_{3/2}$ produces a relatively weak line at 3529 Å with f = 0.036, whereas the 5350 Å line resulting from the transition from the $7^2S_{1/2}$ level to the same metastable level has an oscillator strength f = 0.14. Thus, we may expect preferential laser action at the 5350 Å wavelength since the gain at this wavelength should be considerably higher than at 3529 Å. The metastable thallium level is located 7792 cm^{-1} above the ground state. At a temperature of about 800°C the population of the metastable level is about 6×10^{11} cm^{-3}. At this temperature the upper laser level is practically empty. The relatively high values of the population of the lower level means that population inversion can be established only if we excite the minimum rate of excitation of the upper level sufficient for the establishment of a population of the order of 6×10^{11} cm^{-3} at this level in a time interval of the order of its lifetime. The lifetime of the $7^2S_{1/2}$ level

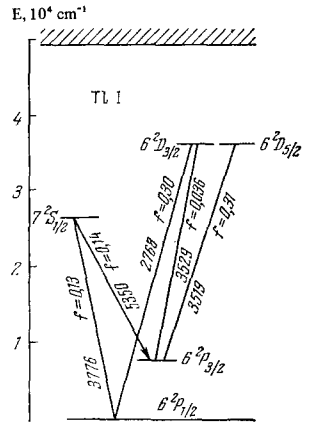

FIG. 1. Energy levels and transitions in the thallium atom.

is 7.4 nsec[34-36] However, in cases of practical importance the radiation resulting from the transition to the ground state is re-absorbed ("resonance trapping") and therefore the upper level can only relax to the metastable level and its lifetime is about 15 nsec. An effective population inversion can be established only if a significant population of the upper laser level is achieved in a time interval not exceeding the lifetime of this level. Therefore, the relevant population inversion can be attained if the excitation pulses have sufficiently steep leading edges. It is desirable to ensure that the leading edge is of the order of 10 nsec or less and that the total pulse duration is of the same order of magnitude. It is quite difficult to construct a power supply capable of producing steeply rising pumping pulses of sufficient power. Nevertheless, laser emission and superradiance corresponding to the green line of thallium at 5350 Å have been achieved experimentally.[26] The laser emission was observed at temperatures from 600 to 900°C and the best results were obtained at about 800°C (this corresponds to a thallium pressure of about 1 torr and an atomic concentration of 9×10^{15} cm^{-3}). The discharge was excited by pulsed cable transformer.[37] The gain of the 5350 Å line was found to be so high that superradiance was achieved easily in a single pass along the active zone which was only 20 cm long. Superradiance was observed for four components corresponding to the two isotopes of thallium Tl^{205} and Tl^{207}, each of which has a nuclear moment of 1/2. Only very rough estimate of the superradiance power and pulse duration were made in[26] because the temporal resolution of the recording system was not sufficiently good. Later measurements showed that light pulses were considerably shorter than was thought initially—their duration was about 1—3 nsec—and the peak output power was of the order of 300 W. These results were obtained for a tube of 3 mm internal diameter and 20 cm active length. Further improvements in the power supply system would be necessary before one could establish the ultimate capabilities of the laser systems producing the green line of thallium.

b) Copper

The energy level scheme of the copper atom is shown in Fig. 2. Copper has two closely spaced resonance levels. The excitation cross sections of these resonance levels, estimated by the aforementioned approximate formula, are 9.7×10^{-16} cm^2 for the $^2P^0_{3/2}$ level and 4.5×10^{-16} cm^2 for the $^2P^0_{1/2}$ level. These cross sections are larger than those for any other transition listed in Table II. Population inversion may be expected for three transitions to metastable levels $^2D_{3/2}$ and $^2D_{5/2}$ but, because of competition, laser emission should be observed for the two lines denoted by arrows in Fig. 2. At the working temperature (1500°C) the population of the $^2D_{5/2}$ level is about 5×10^{11} cm^{-3}. When the resonance radiation is trapped, the lifetimes of the upper levels are 770 and 370 nsec, i.e., they are considerably longer than in the case of thallium. Consequently, the requirements that the pumping pulses must satisfy are much less rigorous.

Laser emission corresponding to the transitions shown in Fig. 2 was achieved at a temperature of about 1500°C (this corresponds to a copper vapor pressure of 0.4 torr and a copper atom concentration of 2×10^{15} cm^{-3}).[22,24] It was difficult to find a suitable material for the construction of a discharge tube which could withstand this temperature. An alumina (Al_2O_3) tube[32] was used in[21,22,24]. Fairly high values of the peak and average output power and a relatively high efficiency were achieved for the transitions in copper. In a tube of 5 cm inner diameter and of 80 cm active length the peak power of the 5106 Å line exceeded 40 kW for pulses of 16 nsec duration and the corresponding efficiency exceeded 1%.[22,24] When the pulse repetition frequency was about 1 kHz, the average power was about 0.5 W. It is worth noting particularly the high efficiency achieved for the green line of copper. At present no other gas laser operating in the visible range is capable of efficiency in excess of 1%. It is suggested in[24] that all the characteristics of the copper vapor laser can be improved by about one order of magnitude and, in particular, the efficiency may be raised by better matching of the durations of the pumping and the emission pulses. It is forecast that it should be possible to reach an efficiency of 5—10%. The difficulties encountered in the construction of the copper vapor laser are mainly due to the very high working temperatures.

c) Lead

The energy level scheme of the lead atom is shown in Fig. 3. We can see that two resonance levels can be pumped efficiently. The electron-excitation cross sec-

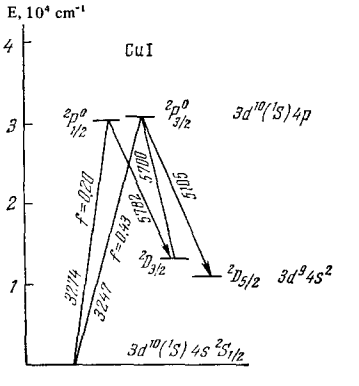

FIG. 2. Energy levels and transitions in the copper atom.

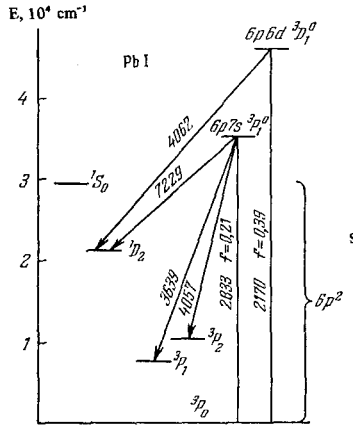

FIG. 3. Energy levels and transitions in the lead atom.

tions of these levels, calculated as usual by means of the approximate formula, are almost equal and they amount to 3.7×10^{-16} cm^2, i.e., they are about half as large as the cross section of the $^2P^0_{3/2}$ level of copper and they are close to the cross sections of thallium. The special feature of lead is the presence of several transitions from resonance to metastable levels which—in principle—can be utilized for laser emission.

The laser action was first achieved for resonance-to-metastable level transitions corresponding to the 7229 Å line.[27] A record gain of 600 dB/m was obtained for this line.[28] Pulses of 2 kW peak power and 15 nsec duration were generated in a tube of 10 cm active length and 10 mm diameter.[28] Long emission times were achieved only for the 7229 Å line. However, if it is assumed that the metastable levels are weakly populated by direct electron excitation, it is found that laser emission should occur also at other wavelengths because their gains are not lower than the gain of the 7229 Å line. We may assume that the absence of laser emission at other wavelengths is not due to direct population of the metastable levels by electrons but due to insufficiently steep leading edges of the excitation pulses. The lifetimes of the resonance levels of lead are of the order of 10 nsec even in the case of complete re-absorption of the radiation resulting from transitions to the ground state. This is due to the presence of several transitions to the metastable levels which means that an effective population inversion can be achieved for most of the postulated transitions only if the excitation is sufficiently rapid. On the other hand, population inversion corresponding to the 7229 Å line may be achieved even if the pumping is slower provided that the populations of the other metastable levels are raised initially by spontaneous emission to such an extent that the radiation associated with these levels is also re-absorbed. In fact, excitation by means of a pulsed cable transformer resulted in the generation of laser radiation and superradiance corresponding to three additional transitions in lead[19] represented by the 4057, 3639, and 4062 Å lines. Under these excitation conditions the laser emission frequently appeared first at the shorter wavelengths and the 7229 Å line was emitted only when the temperature rose sufficiently. These experiments provided further evidence of the importance of a sufficiently steep leading edge of the pulses used to excite such systems.

It follows from Tables I and II and from the above description of the laser emission in thallium, copper, and lead vapors that the results achieved so far are still far from the best possible. It is also clear that further improvement in the characteristics of lasers utilizing transitions from resonance to metastable levels in atoms will meet with considerable difficulties. These difficulties are primarily due to two factors. In all the experiments the excitation pulses were considerably longer than the emission pulses. Moreover, the leading edges of the excitation pulses were not sufficiently steep. Therefore, the efficiencies achieved could not approach the limiting value. The peak powers attained were also limited by the power supply systems.

The second factor likely to cause difficulties is the high working temperature which is needed in all the atomic systems characterized by high limiting efficiencies. High working temperatures complicate the experimental conditions and are likely to cause problems in commercial production of such lasers. In particular, temperatures of about 1500°C are needed for copper vapor lasers for which the best results have been achieved. It is quite likely that the optimal conditions for laser action will require even higher temperatures. At such temperatures very great difficulties are encountered in the construction of the discharge tube, the selection of suitable materials, etc.

The experimental conditions are much easier at temperatures below 1000°C because vitreous silica can be used. Among lasers operating below 1000°C the most promising is that utilizing the green line of thallium. The working temperature for this line is about 800°C and the limiting efficiency is 47%. However, efficient emission in thallium lasers can be achieved only if the power supply is capable of generating short (10 nsec) but powerful excitation pulses.

The two difficulties described above are mutually related. We shall show later that efficient high-power pulsed excitation systems can be achieved by utilizing transverse discharges. However, technical difficulties are encountered in constructing transverse-discharge systems operating at high temperatures.

We can summarize our discussion by stating that all the aforementioned lasers utilizing transitions from resonance to metastable levels in atoms have not yet been investigated sufficiently thoroughly for a final ıal judgment to be made about their ultimate practical capabilities. Further and more detailed investigations are needed. However, it is already clear that the potential capabilities of these lasers can be utilized only if several technical problems are solved. Most important of them is the development of power supply systems capable of generating excitation pulses of duration comparable to that of emission pulses and the construction of discharge cavities which can work at high temperatures.

The question naturally arises whether alternative systems have been fully considered and whether the difficulties outlined above can be avoided by selecting atoms which can be used under more practical conditions.

Inert (noble) gases are most suitable for practical applications. Among them only helium has a transition

from a resonance to a metastable level. Laser action corresponding to this transition was observed experimentally[26] (see Table I). The metastable level of helium lies very high on the energy scale and therefore the limiting efficiency of the corresponding transition is not very high. The other inert gases have similar energy level schemes. Laser emission or superradiance resulting from transitions terminating on metastable 1s-levels (Paschen's notation) has been observed for all these gases. The upper levels in these transitions are of the 2p-type (Paschen's notation) and are not connected to the ground state by allowed transitions. A brief list of the data on the laser transitions in inert gases is given in Table III. Only the laser transitions in neon have been investigated in great detail. Laser emission in pure neon is observed for four lines in the visible part of the spectrum. There are three different points of view on the mechanism of population inversion responsible for these four lines. It is suggested in[39,41] that the inversion responsible for the 2p—1s transitions in neon results from cascade decay from the 2s levels excited by stimulated radiation. A multi-stage excitation of the 2p levels by electrons, via an intermediate resonance level $1s_2$, is proposed in[43]. A similar point of view is put forward in[56] on the basis of an analysis of the lifetimes and excitation cross sections of the neon levels. Finally, it is suggested in[42,46] that the 2p levels are excited by direct electron impact with ground-state atoms. An experimental study of the simultaneous laser action involving the 2p—1s and 2s—2p transitions indicates that the most likely mechanism is the multi-stage excitation via the $1s_2$ resonance level.[49]

The limiting efficiency of the laser action involving the aforementioned transitions in inert gas atoms is considerably lower than the corresponding efficiency for transitions from resonance to metastable levels in thallium, copper, and lead atoms. Moreover, the pumping of the upper level in inert gas atoms is less efficient.

Table III. Laser-active 2p—1s transitions in inert gases for which pulsed emission was observed experimentally

λ, Å	Transition	η_{lim}, %	Remarks	Reference
	Ne I			
5400.56	$2p_1-1s_4$	9.0	P_{peak} = 190 kW	41-49
5944.8	$2p_4-1s_5$	5.5		39, 41, 43, 47, 49, 51
6143.06	$2p_6-1s_5$	5.4		39, 41, 43, 47, 49, 50, 51
6266.5	$2p_5-1s_3$	2.6		49
6304.79	$2p_6-1s_4$	4.0	With admixture of SF_6	54
6506.53	$2p_8-1s_4$	3.8	With admixture of SF_6	54
	Ar I			
6965.8	$2p_2-1s_5$	8.3		52
7067.3	$2p_3-1s_5$	6.6		52
7634.8	$2p_6-1s_5$	6.1		47
7723.6	$2p_7-1s_5$	7.8	Or $2p_2-1s_3$	47
7948.0	$2p_4-1s_3$	2.9		47
	Kr I			
8104.37	$2p_8-1s_5$	6.7		40, 53
	Xe I			
8409.19	$2p_7-1s_5$	9.4		53, 55
9045.4	$2p_9-1s_5$	7.1		40, 53
9799.7	$2p_{10}-1s_5$	8.3		40

In addition to the transitions terminating on levels of the 1s-type, it has been found that strong superradiance is emitted by inert gases as a result of other transitions from higher levels.[39,41,47,49,52,53,57-59] These transitions lie in the infrared part of the spectrum and do not belong to the class of transitions considered here. High efficiencies are even more difficult to achieve for these transitions.

We shall now consider other potential transitions which begin from the first resonance level. Apart from the atoms listed in Table I, there are many other atoms in which transitions can take place from resonance to metastable levels. For example, transitions of this type are exhibited by elements with partly filled p-shells and by several other elements. A list of such transitions is given in[8]. Laser action has been achieved for some of these transitions (they are included in Table I). There are several reasons why we cannot use all the elements which have suitable energy schemes. Many elements form vapors which consist of stable molecules. The use of molecules in laser emission is a separate topic and it will be considered later. Other elements form atomic vapors but extremely high temperatures are needed to achieve the vapor density necessary for laser action. We shall consider only the elements whose vapor pressures are of order of 0.1 torr at temperatures not exceeding 1600°C. We shall ignore radioactive elements. Some elements have a metastable level lying so close to the ground state that it is strongly populated at the working temperatures. If both these restrictions are borne in mind, we can make a list of elements (Table IV) which have transitions suitable for laser action. Table IV gives the transition, the wavelength, the limiting efficiency η_{lim}, and the temperature needed to reach a vapor pressure of 0.1 torr. This value of the pressure was selected, as in[8], because in most cases it is sufficient to observe the laser effect although optimal pressures may be higher. Moreover, Table IV lists the maximum values of the excitation cross section of the upper level from the ground state (calculated using the approximate formula given above), the values of $\tau_L = 1/A_L$ (A_L is the probability of the laser transition), and the populations of the lower level N_2 (at the temperature given in the last column) divided by the population of the ground state N_0. The transition probabilities and the oscillator strengths used in the calculation of the excitation cross sections are taken from[38] and they should be regarded as estimates. The vapor pressures of the elements are taken from[60].

We can see from Table IV that for most of the atoms having a suitable energy level scheme the working temperature exceeds considerably 1000°C so that the difficulties discussed earlier still remain. The only exceptions are barium and bismuth whose working temperatures are lower. These elements are also interesting because they have relatively large values of the excitation cross sections of the upper laser level.

It follows from Table IV that several of the silicon, germanium, tin, and gold lines have low-lying metastable levels and high limiting efficiencies (close to 50%). However, these are the lines for which it would be most difficult to achieve the laser action because the lower levels are strongly populated at the working temperature. The energy level schemes of the chrom-

Table IV. Potential laser-active transitions from resonance to metastable levels in atoms

Atom	Transition	λ, Å	σ_m, 10^{-6} cm²*	η_{lim}, %	τL, nsec	N_2/N_0	T, °C (0.1 torr)
Si I	$4s^3P_1^0 \rightarrow 3p^2\,^1S_0$	4102.94	2	15	—	$2\cdot10^{-5}$	1480
	$4s^1P_1^0 \rightarrow 3p^2\,^1S_0$	3905.52	—	15	34	$2\cdot10^{-5}$	1480
	$4s^3P_1^0 \rightarrow 3p^2\,^1D_2$	2987.65	2	52	50	$2\cdot10^{-2}$	1480
Ge I	$5s^3P_1^0 \rightarrow 4p^2\,^1S_0$	4685.84	4.2	14	200	$1.5\cdot10^{-7}$	1590
	$5s^3P_1^0 \rightarrow 4p^2\,^1D_2$	3269.49	—	50	15	$1.5\cdot10^{-2}$	1590
Sn I	$6s^3P_1^0 \rightarrow 5p^2\,^1S_0$	5631.71	3.5	13	700	10^{-5}	1410
	$6s^1P_1^0 \rightarrow 5p^2\,^1S_0$	4524.74	1.6	16	25	10^{-5}	1410
	$6s^3P_2^0 \rightarrow 5p^2\,^1D_2$	3801.02	3.5	47	15	$2.5\cdot10^{-3}$	1410
	$6s^1P_1^0 \rightarrow 5p^2\,^1D_2$	3262.34	1.6	49	2.5	$2.5\cdot10^{-3}$	1410
Ba I	$6p^1P_1^0 \rightarrow 5d^1D_2$	14999.9	19.5	26	—	$2\cdot10^{-7}$	720
	$6p^1P_1^0 \rightarrow 5d^3D_2$	11303.0	19.5	31	—	$5\cdot10^{-6}$	720
Au I	$6p^2p_{3/2}^0 \rightarrow 6s^2\,^2D_{5/2}$	3122.78	0.5	47	20	10^{-3}	1570
Bi I	$7s^4P_{1/2}^0 \rightarrow 6p^3\,^2D_{3/2}^0$	4722.19	10	43	110	$6\cdot10^{-9}$	690
Cr I	$y^7P_3^0 \rightarrow a^5S_2$	4942.50	5.5	30	2500	$5\cdot10^{-4}$	1510
	$y^7P_2^0 \rightarrow a^5S_2$	4964.93	5.5	36	3600	$5\cdot10^{-4}$	1510
Fe I	$z^5F_5^0 \rightarrow a^5F_5$	5012.07	1.5	37	1500	$4.5\cdot10^{-3}$	1590
	$z^5P_3^0 \rightarrow a^5P_3$	8688.63	1.8	20	530	$6\cdot10^{-7}$	1590
Co I	$x^4D_{7/2}^0 \rightarrow b^4P_{5/2}$	4086.31	3.2	26	36	$4\cdot10^{-7}$	1310
	$z^4F_{7/2}^0 \rightarrow b^4P_{5/2}$	7354.59	2.0	20	6700	$4\cdot10^{-7}$	1310
Ni I	$z^3D_3^0 \rightarrow b^1D_2$	6191.18	1.2	23	6000	10^{-6}	1260
	$z^3D_3^0 \rightarrow a^3P_2$	7110.90	1.2	20	5300	10^{-7}	1260

*Calculated using the simplified formula $\sigma_m \approx \pi a_0^2(2f/\epsilon^2)$.

ium, iron, cobalt, and nickel atoms are very complex and obviously other transitions may occur apart from those listed in Table IV. It is difficult to achieve high laser efficiences for these atoms because the excitation is spread over a large number of resonance lines. The same applies also the rare-earth atoms. For some of them (Eu, Gd, Sm, Tu, Yb) sufficient vapor pressures can be achieved at moderately high temperatures. Laser action is reported in[61,62] for many lines of thulium, samarium, and ytterbium in the near infrared. Some of them are due to transitions from resonance to metastable levels. The large number of levels in these atoms makes it difficult to achieve high efficiency and high power output for a single line.

A comparison of Tables II and IV shows that the achievement of laser action by means of the transitions listed in Table IV will hardly alter the situation. The potentialities of the laser transitions in thallium, copper, and lead atoms are not inferior to those of the transitions listed in Table IV. Moreover, the difficulties associated with the need to employ high temperatures are not avoided by the use of the transitions in Table IV. Thus, it is clear that efficient utilization of the transitions in neutral atoms can be achieved only if serious technical difficulties are overcome.

Till now we have considered only one method for achieving the necessary concentration of active atoms, i.e., the heating of the active substance. We shall now discuss other ways. It is reported in[23] that the laser effect can be achieved in copper vapor generated by electric explosion of copper wires. This method can be used to generate high-density vapors of many metals, including those which are difficult to volatilize. However, it can only be used to produce single pulses. Other methods for introduction of the active substance into a discharge are described in[63-65] for the laser transitions in some ions. It is not clear whether these methods would be suitable for neutral atoms of copper, thallium, and lead because considerably lower concentrations of active particles are employed in ion lasers.

Active atoms can also be introduced into a discharge from molecular compounds. Salts (mainly iodides) of some metals which have resonance lines in the appropriate part of the spectrum have been used for a long time in gas-discharge lamps for the improvement of their luminous efficiency and color characteristics.[6] When the temperature of the lamp wall is about 400°C, the vapor pressures of these salts are of the order of 1 torr. This wall temperature results from the heating by the discharge. Along the discharge axis the temperature is higher, the salt is decomposed, and the metal is strongly excited. The metal is not deposited in free

state on the colder walls because it forms molecules during the time taken to diffuse to the wall. It would be very interesting to introduce copper, thallium, and lead in sufficient concentrations into a discharge by the method just described because discharge tube walls could be kept at a much lower temperature and refractory glasses could be employed. Laser emission corresponding to the green line of thallium was achieved in Tl I vapor.[30] However, in this investigation the mechanism was different from that just described. A study of the discharge spectrum demonstrated that the most probable inversion mechanism under these conditions was the dissociation of molecules as a result of collisions with electrons. This resulted in the preferential formation of thallium atoms in an excited state corresponding to the $7^2S_{1/2}$ resonance level. This mechanism is of intrinsic interest and should be investigated further.

We can summarize our discussion by stating that a convenient method for introduction of active atoms into a discharge is still to be found. Further work is needed on this subject.

4. PULSED LASERS UTILIZING TRANSITIONS IN IONIZED ATOMS

Efficient pulsed lasers can be constructed utilizing transitions from resonance to metastable levels in ions, by analogy with similar transitions in atoms. However, we must bear in mind that the processes of excitation of energy levels in atoms and ions are not fully analogous. It is obvious that the electron-impact excitation in a laser using atomic transitions necessarily produces a considerable number of ions. Therefore, levels of neutral atoms and ions are excited in a discharge. However, the levels of ions are always located above the levels of the corresponding neutral atoms. Therefore, when the degree of ionization is low the energy of electrons in a discharge is utilized mainly in the excitation of atomic levels. Moreover, a high output power and a high efficiency can be achieved only if there is a sufficient number of particles in the state from which serves as the starting point of the pumping process. Therefore, if the pumping is to start from the ground state of an ion (as in the case of atoms), efficient laser emission due to a transition from resonance to a metastable level can be achieved only if the gas is almost fully ionized. However, even in this case the situation pertaining to ions is different. The difference is due to the fact that the initial ionization of a gas may include also direct excitation of the ionic levels starting from the levels of a neutral atom, in particular from the ground state of the atom. In this process the metastable level of the ion may be filled and this makes it more difficult to achieve laser action. Thus, in order to estimate the possibility of establishing population inversion for a transition in an ion, it is necessary to know distribution of the populations of the ionic levels resulting from the ionization process. In most cases this distribution is difficult to predict. The most favorable situation arises at a high pulse repetition frequency when a considerable number of active ions is available at the beginning of each excitation pulse. However, at this moment the population of the metastable level should decay sufficiently, i.e., the conditions should be such that the electron recombination time is considerably longer than the lifetime of the metastable level. It follows that it is more difficult to achieve efficient laser action by means of transitions in ions.

Laser transitions from a resonance to a metastable level had been reported for calcium[8], strontium,[31] and mercury[20] ions. The characteristics of the corresponding ion lasers are listed in Tables I and II. The laser emission by calcium ions appears much later than the corresponding emission of copper atoms. The delay is due to the fact that at a certain time is necessary for the population of the ground state of calcium ions and this time must be sufficient for the re-absorption (trapping) of the resonance radiation. Unfortunately, little is known about laser transitions of this type in ions since they have been hardly investigated.

Obviously laser transitions of this type can be expected also for many other lines because there is a large number of ions which have suitable energy level schemes. Usually the level structure of a singly ionized atom is similar to the structure of the neutral atom in the preceding position in Mendeleev's periodic table. This analogy can be used to predict a large number of suitable transitions in ions. For example, the laser transition in Hg II is analogous to the laser transition in Au I (Table I). The structure of levels of Pb II is similar to the structure of levels of Tl I. There is a transition in Pb II which is analogous to the transition from the resonance to the metastable level in thallium.

Table V lists transitions from resonance to metastable levels for some ions which can be utilized, by analogy with atoms, for efficient laser action. The list of lines in Table V is far from complete. Many of the listed lines are of interest in spite of the low efficiency of the laser action because they suggest possible ways of generating laser radiation in the unmastered range of the spectrum corresponding to wavelengths shorter than 2300 Å.

Table V. Potential laser-active transitions from resonance to metastable levels in ions

Ion	Transition	λ, Å	η_{lim}(I), %	A_L, 10^8 sec^{-1}	Lower level, cm^{-1}	T, °C (0.1 torr)
Ba II	$6p^2P^0_{3/2} \rightarrow 5d^2D_{5/2}$	6142	44	0.095	5 675	720
Ba II	$6p^2P^0_{1/2} \rightarrow 5d^2D_{3/2}$	6497	51	0.075	4 873	720
Ba II	$6p^2P^0_{3/2} \rightarrow 5d^2D_{3/2}$	5854	39	0.012	4 873	720
Pb II	$7s^2S_{1/2} \rightarrow 6p^2P_{3/2}$	2203	51	2.8	14 081	837
Sn II	$6s^2S_{1/2} \rightarrow 5p^2P_{3/2}$	1900	61		4 251	1410
Cu II	$4p^1P^0_1 \rightarrow 4s^1D_2$	2112	40	1.2	26 265	1420
Cu II	$4p^1P^0_1 \rightarrow 4s^3D_1$	2015	33		23 998	1420
Cu II	$4p^1P^0_1 \rightarrow 4s^3D_2$	1970	43		22 847	1420
Ag II	$5p^1P^0_1 \rightarrow 5s^1D_2$	2280	31		46 046	1160
Ag II	$5p^1P^0_1 \rightarrow 5s^3D_1$	2166	25		43 739	1160
Ag II	$5p^1P^0_1 \rightarrow 5s^3D_2$	2034	34		40 741	1160
Sb II	$6s^3P^0_1 \rightarrow 5p^2\,^3P_1$	1504	48		3 055	400
Sb II	$6s^3P^0_1 \rightarrow 5p^2\,^3P_2$	1565	57		5 659	400
Sb II	$6s^3P^0_1 \rightarrow 5p^2\,^1D_2$	1762	51		12 790	400
Sb II	$5p^2\,^3D^0_1 \rightarrow 5p^2\,^1D_2$	1869	50		12 790	400
Bi II	$7s^3P^0_1 \rightarrow 6p^2\,^3P_1$	1777	40		13 324	690
Bi II	$7s^3P^0_1 \rightarrow 6p^2\,^3P_2$	1902	47		17 030	690
Bi II	$7s^3P^0_1 \rightarrow 6p^2\,^1D_2$	2803	32		33 936	690
Bi II	$6d^3D^0_1 \rightarrow 6p^2\,^1D_2$	2143	36		33 936	690

5. PULSED LASERS UTILIZING ELECTRONIC TRANSITIONS IN MOLECULES

We shall now consider the possibility of efficient laser action in which electronic transitions in molecules are utilized. The special feature of molecular systems is the complex structure of levels consisting of a large number of vibrational and rotational sublevels. Because of this complex structure the excited molecules are usually distributed over a large number of levels so that the population inversion and the gain of a given rotational transition are much smaller than in the case of atomic transitions. This makes it difficult to reach the laser threshold. When this threshold is attained, the laser action is usually observed in a large number of rotational lines and sometimes even in several bands. Nevertheless, if we consider the total output power of all the lines we find, as in the CO_2 laser, that this does not greatly impair the changes of achieving high efficiencies and high output powers. Other special features of the molecular systems are the additional selection rules governing transitions between molecular levels and the relatively fast energy transfer by collisions, particularly between the rotational levels. All this leads to new mechanisms for establishing population inversion which are not available in the case of atomic transitions. Consequently, the situation in molecules differs considerably from the situation in atoms. However, it is still desirable to use a resonance state as the upper laser level.

We shall follow these general remarks by a brief analysis of the properties of practical systems in which laser action has been achieved by the use of electronic transitions in molecules. A brief summary of the properties of molecular lasers is given in Table VI. This table gives the systems of bands and the vibrational transitions observed in the laser emission, the approximate wavelengths of the emission bands, and the experimentally determined characteristics such as the peak output power, the saturation value of the peak power density, and the duration of the emission pulses. The best results are listed for each system. Table VI includes also the values of the limiting efficiency and, whenever possible, the cross sections for the excitation of the upper vibrational laser levels by electron impact from the molecular ground state.

It is evident from Table VI that the laser emission involving electronic transitions has been achieved only for diatomic molecules in the infrared, visible, and ultraviolet regions. The laser effect has been achieved for a very limited number of molecules, basically those of nitrogen, hydrogen, and CO. The laser emission achieved for a single line of the NO molecule is a special case of little importance. Thus, the laser effect has been achieved only for the simplest and most convenient (from the experimental point of view) molecules. Apart from hydrogen and its isotopes, which have few suitable lines because of the large rotational constant, the laser emission is observed for many rotational lines and sometimes even several vibrational branches. The total number of laser emission lines in the bands listed in Table VI amounts to a few hundred.

The characteristics of laser action involving electronic transitions in molecules have been investigated quite thoroughly, with the exception of the transitions in nitrogen in the middle infrared region (3–8 μ) and the transition in NO. Therefore, some reliable conclusions can be drawn about the principal population inversion mechanism. A detailed analysis of the features of population inversion in different molecules and transitions is outside the scope of the present review. We shall restrict ourselves to the general statement that in all the most important cases the upper laser level is populated as a result of direct electron-impact excitation of the ground state of a molecule.[100] This is supported by the general nature of the laser emission spectra and by the good agreement between the experimentally obtained emission characteristics with the calculations based on the assumption of direct electron-impact excitation.[9,10,13] Different points of view on the population mechanism of the upper level have been put forward only in the case of the first positive system of the nitrogen molecule. However, later investigations[13, 84, 100] have shown that the direct excitation mechanism explains fully the behavior of the laser effect in this system. This has been demonstrated particularly clearly in[13] where a detailed analysis is given of the inversion mechanism in this nitrogen system and a quantitative agreement is obtained between the experimental results and the calculations based on the direct excitation process.

A characteristic feature of the laser emission resulting from electron transitions in molecules is the fact that the laser effect is observed in few of the large number of the spontaneous emission bands. This feature can be explained quite easily on the assumption of direct electron-impact excitation coupled with the Franck-Condon principle. It is shown in[100] that the application of the Frank-Condon principle to suitably distributed potential curves of the laser states ensures almost automatically that inversion is achieved for some transitions.

By way of example we shall consider the laser transitions in the nitrogen molecule. Figure 4 shows the potential curves of four states of this molecule. A vertical dashed arrow shows, in an arbitrary manner, the band in which the molecules should be excited from the ground state in accordance with the Franck-Condon principle. Downward arrows represent laser transitions (bands) in the first positive ($B^3\Pi_g \rightarrow A^3\Sigma_u^+$) and the second positive ($C^3\Pi_u \rightarrow B^3\Pi_g$) systems of nitrogen. The effect of the Franck-Condon principle can be

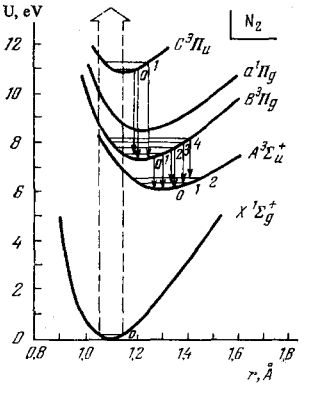

FIG. 4. Potential curves of the nitrogen molecule. The transitions shown in the figure are active in the laser emission.

Table VI. Gas lasers utilizing electronic transitions in molecules

Molecule, transition	$v'-v''$	λ, Å	σ_m, 10^{-18} cm^2	η_{lim}, %	Experiment			Reference
					P_{peak}	$\frac{(P_{peak})_{sat}}{v}$	Δt, nsec	
N_2 1st positive $B^3\Pi_g \rightarrow A^3\Sigma_u^+$	4—2 3—1 2—0 2—1 1—0 0—0 0—1	7 500 7 600 7 750 8 700 8 900 10 480 12 312	14 16 19 19 17 11 11	10 10 10 9 9 8 7	55 kW [129]		200	66–73, 13, 100, 84, 129
2-я положит. $C^3\Pi_u \rightarrow B^3\Pi_g$	1—0 0—0 0—1	3 159 3 371 3 577	9 16 16	18 16.5 15.5	3 MW [33, 86]	40 kW/cm^3	4—7	9, 10, 16, 33, 69, 74–90, 100, 109, 125
N_2 $a^1\Pi_g \rightarrow a'^1\Sigma_u^-$	2—1 1—0 0—0	3.3μ 3.5μ 8.2μ		2 2 2			< 1000	91, 98
N_2 $w^1\Delta_u \rightarrow a^1\Pi_g$	0—0	3.6μ		2				93
N_2		5.2—6.3μ						
H_2 $E^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$	2—1 1—0 0—0 0—1 0—2	8 350 8 899 11 222 13 161 15 820		6 5.5 4.5 4	0.7 kW [13]		200	13, 91–100
D_2 $E^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$	2—0 1—0	8 278 9 530		6 5			200	13, 95–97
HD $E^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$	1—0	9 172		5.5			200	13, 95–96
CO Angstrom system $B^1\Sigma^+ \rightarrow A^1\Pi$	0—0 0—1 0—2 0—3 0—4 0—5	4 511 4 835 5 198 5 610 6 080 6 620	2.4	12.5 12 11 10 9 8	3 kW [130]		100	84, 100–106
NO $B'^2\Delta \rightarrow C'\Pi$	4—1	10 215						107, 108

seen most clearly in the first positive system of bands. We can see from Fig. 4 that direct electron-impact excitation results in a strong population of the $B^3\Pi_g$(v = 0—5) and $A^3\Sigma_u^+$(v = 6—11) levels. The most likely transitions starting from the $B^3\Pi_g$(v = 0—5) levels are those which begin from "turning points" on the right. These transitions terminate at the lower vibrational levels of the $A^3\Sigma_u^+$ state which—according to the Franck-Condon principle—are weakly populated by direct excitation from the ground state. Thus, even these simple considerations show that population inversion corresponding to these transitions should be established at the beginning of a discharge. It is clear from Fig. 4 that these are the transitions which exhibit the laser effect. A similar situation occurs also in the second positive system but it is not so pronounced because the potential curve of the upper state is not displaced so greatly relative to the ground state. A more rigorous analysis utilizing the Franck-Condon factors[110] supports the qualitative picture given above for both systems.

Such a separation of the excitation and the laser emission channels occurs whenever the minima of the potential curves of the laser-active states are shifted relative to the ground state of a molecule. Population inversion will occur in many molecules even if the relationship between the excitation cross sections of the upper and lower laser states is not favorable because the shifted potential curves are the rule rather than the exception and because the Franck-Condon factors can vary by several orders of magnitude. Thus, we may expect to find a large number of additional electronic transitions capable of laser emission under similar inversion conditions. The number of molecules, even just the diatomic molecules, is considerably larger than the number of atoms (elements). Obviously, many of them are unsuitable for our purpose, because some are chemically active and others need heating. Nevertheless, there must be a large number of stable diatomic molecules which are more or less suitable for use in gas lasers. However, it is difficult to select those molecules and transitions which could be used in

gas lasers. The difficulty is the lack of sufficient data for the various molecules. The potential curves are known only for some molecules and the transition probabilities or the lifetimes of electronic levels are known for very few substances. There is hardly any information on the electron-impact excitation cross sections. The Franck-Condon factors are known for few molecules. All this makes it difficult to analyze the population inversion processes involving electronic transitions in molecules or to estimate the capabilities of molecules in any comprehensive manner. Experimental investigations will play a decisive role in future searches for new laser transitions in molecules.

Let us now consider which transitions in molecules are suitable for efficient laser emission. Obviously, the transitions from resonance to low-lying metastable states are of great interest. However, such transitions are encountered quite rarely. Instead, we can use different transitions which are not available in atoms; we can employ molecular transitions from a resonance electronic level to the ground state in which the excitation and the emission channels are separate because of the Franck-Condon principle. This method of population inversion was proposed in 1965[111] for the hydrogen molecule. The potential curves of this molecule are shown in Fig. 5. The vertical dashed lines show, in an arbitrary manner, the band in which the excitation from the ground state of the molecule takes place. This excitation affects the $B^1\Sigma_u^+$ and $C^1\Pi_u$ resonance states which are the starting levels of the well-known Lyman and Werner bands. The potential curves of these states are shifted relative to the ground state in the direction of greater internuclear distances. Therefore, the vibrational levels of these resonance states, which are excited most strongly from the ground state, can act as the starting levels of transitions from the right-hand turning points to higher vibrational levels of the ground state. It is assumed that these vibrational levels are excited weakly from the ground vibrational state. Thus, population inversion should be achieved for these transitions right at the beginning of the excitation pulse. These transitions are indicated by arrows in Fig. 5. They lie in the vacuum ultraviolet near 1600 Å for the Lyman bands and in the 1025–1240 Å range for the Werner bands. In a recent paper[112] a calculation was given of the saturation power corresponding to the hydrogen bands excited under the conditions described in[33]. This calculation demonstrated that the saturation output power could reach 10^3 W/cm^2 and that many of the lines considered should exhibit superradiance. The main difficulty encountered in the utilization of these transitions for laser emission is the very short lifetime of the upper laser states. Calculations show that the inversion lifetime is about 1 nsec. We shall later see that this difficulty is typical of transitions corresponding to short wavelengths.

The method for achieving a population inversion described in the preceding paragraph is applicable to many molecules. It differs from the population inversion in the case of transitions from resonance to metastable levels in atoms in the utilization of higher vibrational levels of the ground state instead of the metastable level. It can be used if the potential curve of the upper laser state is shifted considerably relative to the corresponding curve of the ground state. The lifetimes of the lower laser levels are in practice determined by the rate of exchange between the vibrational levels and, at the usual pressures, they amount to 10^{-4}–10^{-5} sec. In most cases the corresponding probabilities of the laser transitions are higher and therefore we may assume that the laser transitions will be of the pulsed type even in the absence of any modulation.

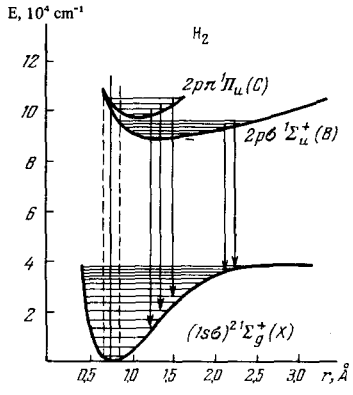

FIG. 5. Potential curves of the hydrogen molecule.

The excitation energy of electronic states is usually high compared with the separation between the vibrational levels and with the thermal energy. Therefore, the limiting efficiency of the laser emission based on transitions involving these states should be quite high. For example, the expected efficiency for the Werner lines is $\eta_{lim} = 42\%$ and that for the Lyman bands is 35%. The excitation cross sections of the resonance states of the molecules may, in favorable cases, amount to 10^{-16} cm^2, i.e., the rate of pumping to the upper state should be close to the rate employed in the pumping of atomic levels.

Apart from the transitions in the hydrogen molecule discussed in the preceding paragraphs, it is worth mentioning also some other transitions in molecules which can be achieved by the inversion method outlined above. In particular, the displaced potential curves are exhibited by the CO molecule. In this case we may expect inversion for the $A^I\Pi$ ($v' = 2$–4) → $X^1\Sigma^+$ ($v'' = 8$–12) transitions near $\lambda \approx 2000$ Å[100]. The lifetimes of the vibrational levels of the $A^I\Pi$ state are of the order of 19 nsec.[113]

An interesting situation arises in the iodine molecule.[100] In this molecule we may expect inversion corresponding to transitions from $B^3\Pi^+_{Ou}$ to the ground state. The potential curves of this molecule exhibit a large shift. Laser transitions should occur in the near infrared and in the visible parts of the spectrum. A detailed analysis is difficult because of the large number of the vibrational levels and because of the complex structure of the Franck-Condon factors.[114,115] The lifetime of the $B^3\Pi^+_{Ou}$ state is known (7×10^{-7} sec).[116] The low value of the rotational constant of the iodine molecule ($B_0 = 0.037$ cm^{-1}) makes it difficult to reach threshold inversion because the excitation is spread over a large number of rotational levels.

There are other transitions in various molecules which might satisfy the requirement for population inversion of the type described for the hydrogen mole-

cule. The lack of necessary data makes it difficult to determine whether laser transitions in such molecules are really possible.

So far we have considered the excitation of laser levels by electron impact in a discharge. However, since the relevant optical transitions undoubtedly obey the Franck-Condon principle, the considerations outlined above apply also to optical excitation. It is usual to ignore optical excitation of gas systems because it is not a very promising inversion method. This opinion is based on the fact that the absorption lines of gases are normally very narrow. Consequently, the coincidence of an absorption line of an atom with an emission line suitable for optical pumping is a very rare event. However, this is not true of molecules with their much more numerous levels. A good example is the iodine molecule. It is well known that a strong emission line coinciding with one of the absorption lines of iodine can be found quite easily.[117,118] Optical pumping is interesting because it provides a very selective means of excitation. Moreover, optical pumping perturbs the active medium much less than does an electric discharge. This is particularly important in the case of lasers which must emit a stable frequency. Optical pumping is also of interest from the point of view of enhancing the efficiency of gas lasers, which is the main subject of the present review. If a resonance line is chosen for the purposes of pumping, the efficiency of conversion of electric energy into resonance radiation may exceed 50%. Since optical pumping can be used to populate selectively a single molecular level, the efficiency of the whole system should be high. The ratio $h\nu_L/E_1$, which occurs in the expression for the limiting efficiency, is determined by the selected molecular transition and can be quite high. The factor f_p includes, apart from the excitation efficiency of a resonance level, the efficiency of utilization of resonance radiation in the system. A laser transition in a molecule can take place from a resonance state to the ground state with separate pumping and emission channels. This scheme is interesting because it makes it possible to utilize resonance levels of atoms which are sufficiently convenient for practical purposes and are yet optimal from the point of high efficiency of resonance emission. On the other hand, this approach is extremely selective and it avoids any difficulties associated with the direct population of the lower level. For example, if the iodine molecule is excited by short light pulses, we can be sure of pulsed inversion for some transitions terminating on high vibrational levels. Convenient pumping lines are the green line of mercury[117] and the resonance lines of sodium.[119]

Optical pumping can be used also without a separate pump lamp but simply by generating a discharge in a mixture of atoms emitting resonance radiation and of molecules which absorb this radiation. This system is definitely of interest because it makes it possible to use vacuum ultraviolet resonance lines for optical pumping even when no suitable transparent media are available. However, this approach is not very selective because molecular levels are also excited in this discharge. Since the optical and the electron-impact excitation mechanisms obey the Franck-Condon principle, we can select molecular transitions in such a way that both excitation processes tend to produce population inversion and the presence of impurity atoms simply increases the rate of excitation of the upper laser level. Practical systems of this type have not yet been constructed. Experimental realization of these systems would be of considerable interest because of the potentially high performance characteristics.

6. COMPARISON OF LASERS UTILIZING ATOMIC AND MOLECULAR TRANSITIONS

A comparison of Tables I, II, and IV shows that the performance characteristics of molecular lasers are likely to be poorer than those of atomic lasers. The excitation cross sections of the higher laser levels of the nitrogen molecule are about an order of magnitude smaller than the corresponding cross sections of the resonance lines of copper, thallium, and lead. The limiting efficiencies of molecular transitions are also lower. Nevertheless, the peak output power of the ultraviolet transitions in the nitrogen laser and, particularly, the specific peak power are considerably higher than the corresponding powers achieved in the atomic transitions (2.5 MW for nitrogen, 40 kW for copper; ~40 kW/cm^3 for nitrogen,* 0.25 kW/cm^3 for lead). This difference is very considerable and it is entirely due to the excitation method. The success achieved in the ultraviolet laser emission in nitrogen is due to utilization of transverse discharges. The output powers achieved in the nitrogen laser in conventional discharge tubes are comparable with those which can be obtained (under similar conditions) for the transitions from resonance levels in atoms. A similar conclusion on the importance of transverse discharges follows from a comparison of the characteristics of the copper, thallium, and lead lasers and of the neon laser with a transverse-discharge system (Table II). It seems that the specific output power of the lasers based on atomic transitions and utilizing transverse discharges should be higher than that achieved in the nitrogen laser.

The first transverse-discharge gas laser was described by Leonard.[75] He achieved a 200 kW peak output power for the ultraviolet emission from nitrogen. A transverse-discharge ultraviolet laser with a peak output of 100 kW and a repetition frequency 100 Hz is now commercially available from the Avco Everett Research Laboratories (USA).[79] Transverse-discharge systems with even more powerful excitation were developed later.[33,86] It is worth mentioning specially a system utilizing a transverse discharge and a traveling excitation wave.[33] The need to use a traveling excitation wave arises from the very short inversion lifetime (a few nanoseconds) in many lasers, including the nitrogen system. During this time the light can travel a distance comparable with the usual length of the active medium (of the order of 1 m). If the whole active medium is excited simultaneously, a part of the medium is traversed by the amplified light signal when the inversion is already exhausted and the medium starts absorbing. This means that a signal is amplified only in that part of the active medium whose length is

*Saturation value of the power per unit volume.

of the order of the distance traveled by light during the existence of the population inversion. This difficulty is avoided in[33] by ensuring that the breakdown along the active medium occurs successively and the delay is such that the breakdown wave travels along the active medium at a velocity close to that of light. In such a system a signal appears at one end of the active medium and travels along it being amplified continuously at moments close to the inversion maxima at all points. The light traveling in the opposite direction is amplified much less strongly. A system of this kind emits, even in the absence of mirrors, a power ten times higher in one direction than in the opposite direction. This technique is very important in the achievement of high power outputs in the case of short inversion lifetimes because it makes it possible to utilize a long active cell. The cell used in[33] was 2 m long and the pulses emitted by nitrogen were of 4 nsec duration. Other systems with steeply rising excitation pulses are also of interest: they cannot yield such high powers as transverse-discharge systems but they are much simpler and more convenient.[83,85,109]

A comparison of the parameters of laser emission utilizing transitions in atoms and molecules shows which directions of further development of these two types of laser should be pursued. In order to increase the output power and the efficiency of the lasers utilizing transitions from resonance to metastable levels in atoms it would be very desirable to use transverse-discharge excitation systems providing excitation pulses of duration comparable to the duration of laser emission pulses. This could be achieved by overcoming the technical difficulties associated with high working temperatures. Transverse-discharge systems are already used in molecular lasers. However, transitions optimal in respect of the efficiency and the rate of pumping of the upper level have not yet been found. Therefore, search should be continued for laser action in new molecules and transitions. It would be very interesting to study the laser transitions from a strongly excited resonance state of a molecule to a high vibrational level of the ground state.

7. PROSPECTS OF ADVANCE TO UNMASTERED SHORT WAVELENGTHS

The greatest hopes for mastering the short-wavelength part of the spectrum lie in the pulsed gas lasers. At present the unmastered part of the spectrum lies at wavelengths shorter than 2300 Å. This situation arises from the fact that the difficulties in achieving stimulated emission increase rapidly with decreasing wavelength. High-reflectivity mirrors are not yet available for wavelengths shorter than 2300 Å. Moreover, it is difficult to find transparent optical materials for the vacuum ultraviolet range. However, the greatest basic difficulty is the fall in the gain with decreasing wavelength. The gain at the center of a Doppler-broadened line is given by the expression

$$k_0=\frac{\lambda^2}{4}\frac{A_{ik}}{\Delta\omega_D}\Delta N\sim\frac{f_{ki}}{\Delta\omega_D}\Delta N;$$

here, A_{ik} is the probability of a laser transition; f_{ki} is the oscillator strength of this transition; $\Delta\omega_D$ is the Doppler line width expressed as the angular frequency; $\Delta N = N_i - (g_i/g_k)N_k$ is the population inversion. Under steady-state conditions in a favorable case (when the population of the lower laser level N_k can be ignored and the upper level is de-activated solely by spontaneous emission along the laser-active transition), we obtain

$$\Delta N=q_i/A_{ik},\quad k_0\sim\frac{\lambda^2}{\Delta\omega_D}\sim\lambda^3,$$

where q_i is the rate of pumping of the upper level.[120] Thus, the gain is independent of the probability of the laser transition and it falls with decreasing wavelength as λ^3.

Under pulsed conditions the situation is somewhat different. In principle, we can pump the upper level in an interval shorter than its lifetime. Then, the decay of the upper level can be ignored and ΔN is governed solely by the pumping rate. Consequently, a high gain can be achieved by selecting a transition characterized by a high probability A_{ik}. For example, if laser transitions with the same oscillator strengths are selected at different wavelengths, the gain depends on the wavelength only through the Doppler line width, i.e., $k_0 \propto \lambda$. However, we must bear in mind that the inversion lifetime under pulsed conditions is of the order of the lifetime of the upper level. This means that the inversion lifetime for transitions selected in this way decreases as λ^2 and it is necessary to increase strongly the pumping rate, i.e., to use high-power steeply rising excitation pulses. This simply alters the nature of the difficulties but does not simplify the problem. In fact, in the visible part of the spectrum a typical lifetime of the upper level of pulsed lasers is 10^{-8}—10^{-7} sec, whereas in the vacuum ultraviolet the lifetime of the same level is 10^{-9}—10^{-8} sec and the inversion lifetime is even shorter. Thus, it is necessary to utilize nanosecond pulses. Under these conditions resonators are practically useless because light can traverse a distance less than 1 m during the inversion lifetime. It means that one must consider using superradiance and traveling-wave excitation systems in order to utilize effectively active media of longer lengths.

One can also select laser transitions in a different way. For example, one may consider transitions of constant probability. In this case the inversion lifetime remains constant but $k_0 \propto \lambda^3$. For example, the gain decreases by a factor of 37 between 5000 Å in the visible range and 1500 Å in the vacuum ultraviolet.

We have mentioned earlier that the transitions favorable for the laser action at wavelengths shorter than 2300 Å are exhibited by various ions and molecules. Some of them are listed in Table V and in Sec. 5. Many promising transitions can be found in molecules, particularly among those from resonance to ground states, because numerous molecules exhibit resonance transitions corresponding to wavelengths shorter than 2300 Å.

8. INCREASE OF CONCENTRATION OF PARTICLES IN ACTIVE MEDIUM

Gas lasers have many advantages over other lasers (transparency of the medium over a wide range of wavelengths, high optical homogeneity, narrow emis-

sion lines, many inversion mechanisms). However, they have one common disadvantage: high output powers and energies have not yet been achieved in gas lasers and this applied particularly to the output power and energy per unit volume of the active medium. This disadvantage stems from the low concentration of the active particles. For example, in a solid laser a typical concentration of active ions is 10^{19} cm^{-3}, whereas in the majority of gas lasers the concentration of active atoms and molecules is of the order of 10^{16}–10^{17} cm^{-3}. Ionic lasers operate at concentrations which are lower by further two or three orders of magnitude. Hence, it is clear that in order to achieve high specific output powers and energies from gas lasers, one must increase the efficiency and also raise the concentration of active particles. The problem of increasing the concentration of active particles is one of the most important tasks in the future development of all the gas lasers, including pulsed systems. The pressure of the active gas must be raised up to atmospheric pressure in order to approach the concentration of active particles in solid lasers. The achievement of such concentrations presents serious difficulties.

It is clear from the preceding section that even at pressures of about 1 torr the atoms utilized in high-efficiency lasers (copper, lead thallium, etc.) must be heated to high temperatures. Further increase of the particle concentration by heating is not only technically inconvenient but also gives rise to a basic difficulty resulting from the rapid rise of the equilibrium population of the metastable levels with increasing temperature. This difficulty is particularly serious in the case of low metastable levels, i.e., in the case of transitions characterized by high efficiency.

In most gas-discharge lasers the optimal conditions are attained at some definite electron energy, i.e., at a definite value of E/P. For pulsed lasers the typical values of E/P are of the order of 100 V-cm^{-1}torr^{-1}. This would mean that a voltage of 8×10^6 V would have to be applied to a discharge tube 1 m long containing an active gas at atmospheric pressure. Operation at such high voltages is hardly practical. The situation becomes easier if transverse discharges are used. The utilization of such discharges makes it possible to use higher gas pressures and moderate voltages in laser cells whose active volumes are of the same oder as in longitudinal discharges.[33,75,86] This is achieved because of the short distance between the electrodes.

The need to operate at higher voltages is not the only difficulty preventing utilization of higher pressures. The properties of the discharge itself (filamentation and concentration of the discharge along the walls) change at higher pressures. At the atmospheric pressure the electric discharges are of the streamer type. All this makes it difficult to generate a homogeneous plasma and has an unfavorable effect on the laser emission. Very little information is available on the properties of high-pressure discharge plasmas suitable for population inversion.

It should also be pointed out that at atmospheric pressures the probabilities of transitions resulting from collisions with heavy particles may be comparable with radiative probabilities. Therefore, the mechanisms of level population may change with increasing pressure because of the participation of collisions with heavy particles. Moreover, an allowance must be made for the collision-induced line broadening. Thus, definite difficulties would be encountered at high pressures and intensive investigations of high-pressure operation are desirable.

Some progress toward higher pressures has been reported recently. Superradiance at 3371 Å was generated in nitrogen near atmospheric pressure.[83] High peak output powers were achieved under pulsed conditions at atmospheric pressures in a transverse-discharge system utilizing vibrational transitions in the CO_2 molecule.[121,122]

9. WAYS OF ACHIEVING CONTINUOUS-WAVE EMISSION IN COLLISION LASERS. MAXIMUM PULSE REPETITION FREQUENCY

The pulsed gas lasers discussed in the preceding sections are also very interesting as an intermediate stage in the development of continuously operating efficient collision lasers working in the ultraviolet, visible, and near infrared part of the spectrum. It would be very desirable to use an easily populated resonance level as the upper level in continuously operating collision lasers. On the other hand, since the lower laser level could still relax via collisions with heavy particles, this level can be metastable.* Thus, an efficient collision laser should utilize transitions of the type discussed in preceding sections. A pulsed laser utilizing transitions from a resonance to a metastable level can be converted to a cw collision laser by ensuring the necessary rate of collisional relaxation of the lower level. The cross sections for the de-excitation of levels by collisions with heavy particles are of the order of 10^{-16} cm^2 and under very favorable conditions they may amount to 10^{-15} cm^2. This means that the probability of de-excitation of a level is of the order of 10^5–10^6 sec^{-1}·torr^{-1} when the concentration of the quenching particles is 3×10^{16} cm^{-3} (this corresponds to a pressure of 1 torr at room temperature). We have mentioned earlier that difficulties are encountered when the working pressure is increased. At present one may expect to use quenching-gas pressures of the order of 10 torr, i.e., one may expect level de-excitation rates of the order of 10^6–19^7 sec^{-1}. This means that laser transitions suitable for cw collision lasers are those with a relatively low transition probability, which is of the order of 10^5–10^6 sec^{-1} or less. Among the transitions listed in Table I for which pulsed laser emission has been achieved experimentally suitable probabilities (of the order of 10^6 sec^{-1}) are exhibited by copper, manganese, helium, and ionized calcium. However, transitions in all these atoms have probabilities somewhat higher than 10^6 sec^{-1}. For example, $A = 1.3 \times 10^6$ and 2.7×10^6 sec^{-1} for the copper lines at 5106 and 5782 Å, respectively. In the case of manganese $A = 1.7 \times 10^6$ and 1.8×10^6 sec^{-1} for 5470 and 5538 Å, respectively.[38] The probabilities of the other transitions just mentioned are of the same order of magnitude. This means that continuous emission utilizing these

*Butaeva and Fabrikant [123] were the first to suggest collisional de-excitation of the lower laser level.

transitions is in principle possible but one must select a quenching gas characterized by a large collision cross section (of the order of 10^{-15} cm^2) and to operate at pressures of the order of tens of torr. The attainment of continuous operation in the known lasers utilizing transitions from resonance to metastable levels depends to a great extent on the selection of the quenching gas. This gas must de-excite selectively the metastable level but not the upper laser level and it should not impair greatly the pumping of the upper level because of a change in the conditions in the discharge plasma. Large energy-transfer cross sections are obtainable only in resonant processes. Thus, the quenching atom or molecule should have a level of energy close to the metastable level of the active atom. On the other hand, rapid relaxation to the ground state should be ensured by the availability of intermediate levels because the energy of the metastable level is usually too high (of the order of 1 eV) for a single-stage transition to the translational degrees of freedom. The quenching particles may be the active atoms or molecules. In this case the particles should have a set of closely spaced levels for multi-stage collisional transfer of energy from the lower laser level to the ground or some intermediate state. Manganese and lanthanum atoms have been suggested for collision lasers.[2] Pulsed emission utilizing many transitions in manganese was reported in[25] but the collision laser conditions were not achieved.

Energy level schemes suitable for collision lasers are found more easily in molecules. From the point of view of electronic transitions the closest approach to the ideal would be provided by a system with a laser transition from a resonance electronic level to higher vibrational levels of the ground state. We have said already that laser emission resulting from such a transition would have the advantages of high efficiency and high power output and population inversion in molecules with shifted potential energy curves would result from the Franck-Condon principle. In this case the lower laser level may relax by an almost resonant process involving energy transfer between neighboring vibrational levels. The rate of such a process is relatively high.[124] Some part of the vibrational energy of the ground state may be extracted in the form of stimulated radiation resulting from vibrational transitions. It is known that in some molecules these transitions are capable of highly efficient laser action when the upper laser levels are pumped by collisions with vibrationally excited molecules of a second substance. Thus, for example, in the event of achievement of laser emission by way of transitions from some upper electronic level to the ground electronic state of the nitrogen molecule it should be possible to transfer a considerable fraction of the vibrational energy of nitrogen to the CO_2 molecule and to extract this energy in the form of stimulated radiation resulting from transitions in CO_2.

The development of a laser utilizing electronic transitions in molecules at sufficiently high pressures of the active or a specially selected gas would yield a device whose operation could approach that of a four-level laser or a dye laser. A sufficiently rapid collisional exchange of energy between the vibrational levels of the upper and lower laser states would make the distribution of populations over the vibrational levels of these states approach the Boltzmann distribution with definite vibrational temperatures of the upper and lower states. In this case the laser action in molecules with displaced potential energy curves would be most likely due to a transition from a lower vibrational level of the upper electronic state to weakly populated upper vibrational levels of the lower electronic state. The operation of such a laser is shown schematically in Fig. 6.

The development of collision lasers emitting in the ultraviolet, visible, and near infrared regions would be extremely desirable. The continuous output power of such lasers is determined primarily by the rate of relaxation of the lower level. The rate of pumping of the upper level and the rate of relaxation of the lower level in collision lasers utilizing electronic transitions may be expected to be of the same order as in the CO_2 laser. This means that simply by increasing the energy of the laser quanta it should be possible to achieve an output power per unit volume an order of magnitude higher than that for the CO_2 laser. The efficiency of the electronic-transition lasers of this kind can be very high even if the transitions terminate at relatively high vibrational levels of the ground state. Utilization of high vibrational levels is desirable because it would make it possible to achieve laser action without significant reduction in the population inversion at higher gas temperatures. Thus, the development of collision lasers utilizing transitions from resonance to ground states in molecules would provide high-efficiency cw lasers emitting ultraviolet, visible, and near infrared wavlengths at power levels an order of magnitude higher than those achieved in the CO_2 laser.

The problem of development of short-wavelength collision lasers is closely associated with another pressing problem which is the increase in the repetition frequency and the average output power of pulsed lasers. The maximum repetition frequency is determined by the time necessary for the restoration of the conditions existing at the beginning of the preceding pulse. In particular, it is necessary to empty the lower laser level before the arrival of the next excitation pulse. In the case of systems with a metastable lower level the relaxation of this level results from collisions with heavy particles or from diffusion. The only difference between the pulsed and the cw collision lasers is that the excitation by electrons and the relaxation occur at different times in the pulsed case. The lower

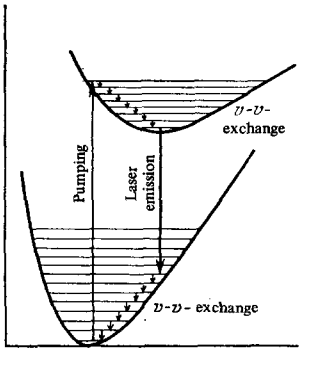

FIG. 6. Schematic representation of the operation of a collision laser which utilizes electronic transitions in a molecule in which relaxation takes place between vibrational levels.

level relaxes during the interval between pulses. The lasers working in this way are known as "cyclic lasers."[8] These lasers can be regarded as an intermediate type between pulsed and cw collision lasers. In view of this it would be very desirable to investigate maximum pulse repetition frequency and the output power as function of the experimental conditions, particularly the nature and the pressure of the gas added for the purpose of relaxation of the lower level. Such investigations would provide a convenient method for the determination of the operating conditions in cw collision lasers. Moreover, these investigations would be of intrinsic interest, especially from the point of view of practical applications. Very little work of this type has been done so far. Special investigations of the maximum repetition frequency have been carried out only for the laser action resulting from transitions in the nitrogen[69,90,126] and CO[102,104] molecules. Practically no work has been done on the influence of foreign gas admixtures.

10. CONCLUSIONS

Pulsed gas-discharge lasers hold out a strong promise of high efficiency, high peak power, and operation in a very wide range of wavelengths. Transitions from resonance levels are specially advantageous. Pulsed laser action utilizing transitions from resonance to metastable levels, which is promising from the point of view of efficiency and output power, has been achieved in many neutral and ionized atoms. Further development of atomic lasers requires overcoming of technical difficulties associated with the constructionn of discharge cavities capable of operation at high temperatures and of supply systems providing high-power short pulses. Pulsed laser emission utilizing electronic transitions in molecules has been achieved only for the simplest molecules. The peak output power of the molecular transitions is considerably higher than that of the atomic transitions although the limiting values of the pumping rate and the efficiency are lower in the molecular case. The high output power has been achieved in molecular lasers by the use of transverse discharges. However, the molecules and transitions with the best possible parameters are still to be found. The greatest promise is held out by transitions from a resonance electronic state to higher vibrational levels of the ground state.

The results achieved so far are far from optimal and can be improved in many respects. In particular, the peak output power can be raised considerably by increasing concentration of active particles.

Finally, pulsed gas-discharge lasers represent an intermediate stage in the development of efficient cw collision lasers operating in the visible, near infrared, and ultraviolet parts of the spectrum. These lasers should have efficiencies and specific output powers an order of magnitude higher than those achieved in the CO_2 laser.

Note. When the present review was completed, a report appeared of the experimental realization of the population inversion mechanism suggested in [111] (this mechanism is discussed in the second half of Sec. 5). Stimulated emission was achieved by means of transitions from resonance electronic states to higher vibrational levels of the ground electronic state in molecules.

Hodgson [126] reported achievement of stimulated emission in the Lyman bands of the hydrogen molecule (transitions from the vibrational levels $v' = 3$-7 of the resonance state $B^1\Sigma_u^+$ to the higher vibrational levels $v'' = 10$-13 of the ground electronic state of H_2). Stimulated emission was observed in 10 lines belonging to the P branches in the wavelength range 1581-1613Å. The experiments were carried out using a transverse-discharge cavity. Coherent light pulses of 1.5 kW peak power and about 2 nsec duration were obtained.

Waynant et al. [127] also observed stimulated emission at 10 wavelengths (1567-1613Å) in the Lyman bands of the hydrogen molecule. The spectral composition of the stimulated emission differed somewhat from that reported in [126]. A pulse-discharge system excited by currents of hundreds of kiloamperes in the form of pulses of 2.5 nsec edge was used. The discharge was of the transverse type and a traveling excitation wave was employed. The peak output power was 100 kW and the duration was about 1 nsec. As in [33], stimulated emission along the direction of propagation of the excitation wave was much more powerful than the emission in the opposite direction. This provided further confirmation of the high gain in the medium.

More recently, Hodgson [128] reported briefly that, in addition to the hydrogen molecule, he achieved stimulated emission in the Lyman bands of the molecules of parahydrogen, HD, and D_2 in the spectral range 1520-1615Å. These are the shortest wavelengths at which stimulated emission had ever been achieved. Hodgson also reported stimulated emission in the lines of the R and Q branches of the fourth positive system of the CO molecular bands in the spectral range 1800-2000Å. Utilization of these lines had been suggested earlier by the present author [100].

Further improvements in the performance characteristics have been reported for several lasers discussed in the present review. Kaslin, Knyazev and Petrash [129] used cooling of the gas to increase significantly the efficiency and to raise the peak output power to 55 kW for the first positive system of the nitrogen molecule. Under the same conditions they observed also high-peak-power superradiance in the same band system. The use of discharge tubes of larger diameter and cooling of the gas also increased strongly the peak output power (up to 3 kW) in the Ångstrom bands of the CO molecule. [130] The power obtained for the same bands in an earlier investigation was only about 20 W. [106]

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