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Collision lasers on atomic transitions

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Contents

***Abstract*. This paper reviews the research of cw collision lasers on transitions of atoms and atomic ions and presents characteristics of systems that are considered to be collision lasers. Literature data on ‘relaxation’ and ‘mixing’ of levels by collisions with heavy particles are discussed, with particular attention to the relaxation of metastable levels. The major problems in the development of efficient cw collision lasers are analysed, including difficulties in realising discharges suitable for pumping collision lasers. The possibility of further advances in collision laser development is discussed.**

Keywords: *gas discharge lasers, transitions of atoms and atomic ions, relaxation of levels by heavy-particle collisions.*

1. Introduction

Among the many types of gas lasers, only a few have found practical application. These are mostly lasers offering high efficiency, such as CO_2, CO, excimer, exciplex and atomic copper vapour lasers. Unfortunately, the great majority of gas lasers have efficiencies below 1 %, which severely limits their application field. Moreover, in recent years they have faced strong competition from efficient solid-state and fibre lasers in combination with a variety of frequency conversion techniques.

The collision laser concept was proposed very early in gas discharge laser development and held promise for high efficiency and high-power continuous operation. This paper reviews studies concerned with collision lasers and demonstrates that their performance is still far from expected. An analysis is presented of the factors that limit their efficiency and power output, and an attempt is made to ascertain whether all the possibilities for designing efficient collision lasers have been utilised or further progress in this area is possible.

2. Collision laser concept

The collision laser concept was formulated in 1965 by Bennett [1] and Gould [2]. It emerged very early in gas laser development in the context of the search for high-efficiency systems. As a matter of fact, in developing this concept the main purpose was to achieve high efficiency. Recall that the problem in question is that of creating a cw gas discharge laser. To achieve a steady-state population inversion and lasing, the following condition should be met [1]:

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$$R_1 > (g_2/g_1)(A_{21} + R_2F_1/F_2).$$

Here, subscripts 1 and 2 refer to the lower and upper laser levels; R_1, R_2, and F_1, F_2 are the level depopulation and population rates, respectively; A_{21} is the transition probability; and g_1 and g_2 are the statistical weights of the lower and upper laser levels. Continuous operation has some advantages over pulsed regime because the energy input for plasma generation is needed only once, and then the optimal inversion conditions can in principle be maintained for a long time, with only a low power needed to maintain a steady state of the plasma.

A systematic analysis [1, 2] showed that, in almost all the gas discharge lasers known at the time, the lower laser level was depopulated through spontaneous emission on a strongly allowed transition. In such a situation, an intermediate level between the lower laser level and the ground state is needed because spontaneous decay to the ground state is impeded by resonance radiation trapping. Moreover, the rate of electron-impact excitation on an allowed transition is, as a rule, rather high, leading to an undesirable increase in the lower laser level population. If there is an intermediate level with allowed transitions to the lower laser level and ground state, raising the pump power causes the power output to saturate and then decline.

On the other hand, due to the presence of an intermediate level and because high spontaneous decay rates are typical of short-wavelength transitions, the laser transition occurs between high-lying levels, reducing the laser efficiency.

Gould [2] used the following relation for laser efficiency:

$$\eta \approx f\frac{h\nu_{\mathrm{L}}}{E_{\mathrm{u}} - E_{\mathrm{t}}},$$

where $h\nu_{\mathrm{L}}$ is the emitted photon energy, E_{u} is the energy of the upper laser level, E_{t} is the energy of the 'reservoir' level utilised for pumping, and f is the fraction of atoms that are excited from the reservoir level and contribute to the laser transition. The presence of an intermediate level reduces both the energy ratio in the above formula and f because near and below the upper laser level there is a significant number of other levels, which 'intercept' the excitation energy.

Examining a molecular laser, Gould [2] concluded that, in such a case, the large number of levels in molecules would almost inevitably lead to a low value of f and that, therefore, the density of molecules in an efficient gas discharge laser should not be high.

He considered a variety of relaxation mechanisms for the lower laser level and identified the fastest processes: spontaneous emission and collisions with heavy particles (collisional relaxation). Since, as mentioned above, relaxation through spontaneous emission leads to efficiency limitations, he concluded that an efficient gas discharge laser should utilise heavy-particle collisions to relax the lower laser level. In such a case, use can be made of transitions between relatively low lying levels and higher efficiency can be expected.

The basic principle of an efficient gas discharge laser, referred to as a collision laser, emerged from the analysis presented in [1, 2]. It utilises steady-state discharges with a large difference between the electron temperature T_{e} and the gas temperature T_{g}, such that the population of some levels is determined primarily by the electron temperature, and that of other levels, by the gas temperature. In addition, it was assumed that $T_{\mathrm{e}} \gg T_{\mathrm{g}}$. The following recommendations regarding the selection of collision laser systems were made by Gould [2]:

The primary excitation of the atoms would be by collisions with free electrons, while inelastic collisions between atoms would both enhance the upper level population and relax the lower. The ideal characteristics of such a system are summarized as follows:

(a) The laser transition is slow (partially forbidden).

(b) The upper level is not depleted by inelastic atom–atom collisions ($E_{\mathrm{u}} - E_{\mathrm{l}} \gg kT_{\mathrm{g}}$).

(c) All active levels have low energy to insure high population and high electron excitation rates.

(d) The cross-section is large for collisional transfer between the laser levels and nearby levels (spacing $\Delta E \sim kT_{\mathrm{g}}$).

(e) All levels with significant population facilitate funneling the atoms through the laser transition.

(f) The total energy defect, $\Sigma\Delta E$, spanning a series of inelastic transfers suffices to make $N_{\mathrm{l}} < N_{\mathrm{u}}$. A necessary condition is

$$\mathrm{e}^{-\Sigma\Delta E/kT_{\mathrm{g}}} < \mathrm{e}^{-(E_{\mathrm{u}}-E_{\mathrm{t}})/kT_{\mathrm{e}}}.$$

(Here, E_{u} and E_{l} are the energies of the upper and lower laser levels, E_{t} is the energy of the reservoir level utilised for pumping, T_{g} is the gas temperature and T_{e} is the electron temperature.)

(g) The gas density is low enough to allow $T_{\mathrm{e}} \gg T_{\mathrm{g}}$, but large enough to insure the above inequalities. Therefore $N \sim 3 \times 10^{17}$ cm^{-3}.

(h) Excitation of the upper level is indirect, via collisions of the second kind with a more populous atomic species having an isolated level (thus dominating wasteful electron excitation of the lower levels).

Gould assumed that, in the limit, the upper and lower laser level populations might approach those at equilibrium at temperatures T_{e} and T_{g}, respectively. The laser efficiency may then be fairly high, approaching the efficiency of an ideal heat engine [2], $\eta = (T_{\mathrm{e}} - T_{\mathrm{g}})/T_{\mathrm{e}}$. In addition, a high power output was expected. Such prospects aroused great interest, and a variety of systems were proposed for collision laser development, in particular by Bennett [1] and Gould [2].

Note that the feasibility of creating a collision laser needs no proof: the well-known CO_2 laser relies on this principle and does offer high efficiency and high output power in the mid-IR. By the irony of fate, it operates on molecular transitions, whereas according to Gould its active medium should contain as few molecules as possible. At the same time, the development of collision lasers on molecular transitions in the near-IR and visible spectral regions faces severe difficulties. Here we will address the problem of creating a collision laser on transitions of atoms and atomic ions, desirably at short wavelengths.

Many attempts have been made to design such lasers on transitions in atomic systems. There have been a number of reports on the creation of collision lasers. These issues will be addressed below in greater detail. Ongoing attempts to create collision lasers based on the concept proposed in [1, 2] have been the subject of much discussion at a number of conferences and seminars. It can be said with certainty, however, that the creation of efficient high-power cw gas discharge lasers operating on transitions of atoms and atomic ions continues to be a challenge. Also, little progress has been made in visible and UV collision laser development.

3. Systems considered to be collision lasers

Bennett [1] and Gould [2] focused primarily on three-level lasers operating in a continuous regime, with brief discussion of more complex systems. Under ordinary conditions, when the electron energy distribution differs only slightly from the Maxwellian distribution, population inversion cannot be achieved using only electron impact excita-tion. A selective process is needed capable of reducing the lower level population and/or raising the upper level population.

In most later studies, transitions from resonance levels to metastable levels (R – M transitions) were considered promising. Indeed, R levels are as a rule best excited by discharge electrons, whereas electron excitation and deexcitation of M levels were expected to be weak. Accordingly, M levels were assumed to be relatively easy to relax by collisions with heavy particles. Typically, the simple energy level diagram presented in Fig. 1 was considered. The energy of the M level should considerably exceed kT_g. To raise its relaxation rate, it is desirable to have intermediate levels between levels 0 and M (shown schematically in Fig. 1).

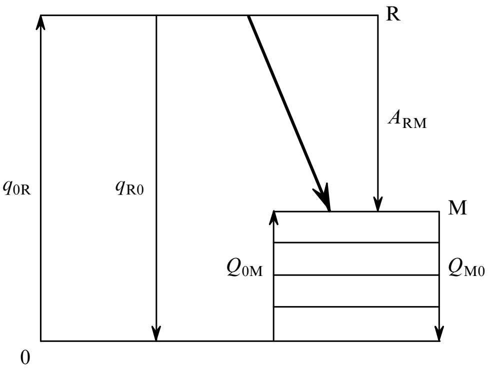

Figure 1. Energy level diagram of a collision laser on an R – M transition. The slanted arrow represents the expected laser transition. Q_{0M} and Q_{M0} are the rate constants of excitation and depopulation by heavy-particle collisions; q_{0R} and q_{R0} are the analogous rate constants for electron impacts; A_{RM} is the transition probability.

3.1 Lasers on Ca and Sr transitions

These are currently the only lasers that have been made to operate in a continuous regime. The energy level diagram and transitions of the Ca atom are presented in Fig. 2, where the laser transition at 5.547 μm is represented by a heavy arrow. Continuous operation was typically obtained when small amounts of hydrogen were added [3 – 9]. In this system, the lower laser level is depopulated by 'chemical cleaning': the removal of atoms from the lower laser level $^1D_2$ via the chemical reaction

$$\mathrm{Ca}(^1D_2) + H_2 \rightarrow \mathrm{CaH} + \mathrm{H} + \Delta E.$$

The rate of this reaction must be relatively high because it has a resonance character. ΔE is then comparable to kT_g.

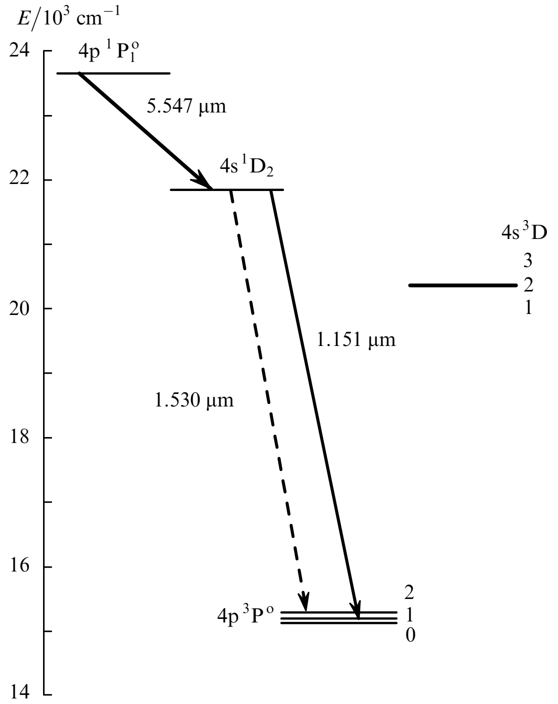

Figure 2. Partial energy level diagram and transitions of the Ca atom.

CW lasing was obtained at hydrogen pressures in the range 1 – 3 Torr. With the other parameters optimised, the output power was ~0.1 W, and the estimated efficiency was ~10^{-2}. This efficiency can be considered rather high because the ratio of the laser transition energy to the upper level energy is ~7.6 %. Under such conditions, the lasing lasted 10 – 30 min because the calcium pieces converted to calcium

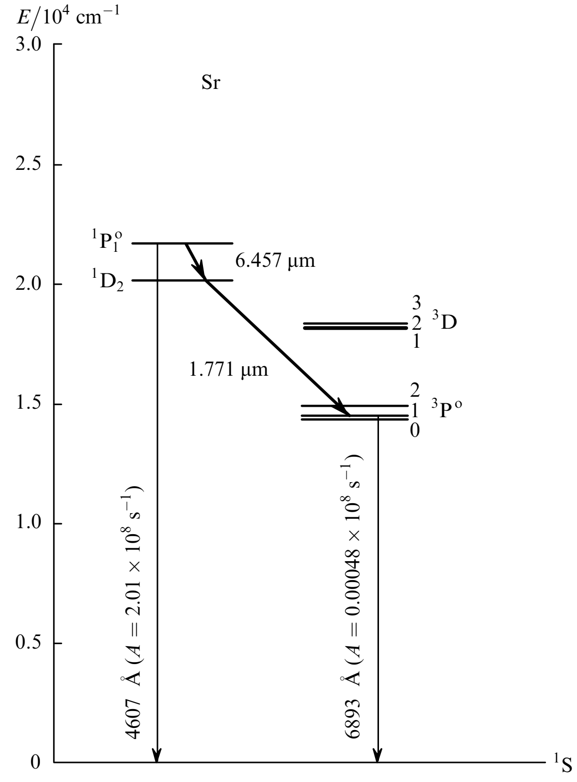

Figure 3. Energy level diagram and transitions of the Sr atom.

hydride powder over that time. At lower hydrogen pressures, lasing lasted several hours at a power of ~ 0.07 W.

CW lasing on a similar strontium transition was reported by Cahuzae [9], but he did not specify whether or not hydrogen had been added. The energy level diagram and transitions of the Sr atom are presented in Fig. 3.

Note that the laser transitions of atomic Ca and Sr are, strictly speaking, not R – M transitions because there are intercombination transitions from the lower laser level $^1D_2$ to the $^3P^o_{1,2}$ states. For one such transition of the Sr atom ($\lambda = 1.771$ μm, Fig. 3), even pulsed lasing was obtained, attesting to a sufficiently high transition probability. It is this spontaneous decay channel which was thought to account for the cw lasing [9]. Unfortunately, no reliable data on the probabilities of laser transitions or downward transitions from the lower laser level have been found for Ca or Sr. The effect of spontaneous decay of the lower laser level remains unclear. We note only that these probabilities must be lower for Ca compared to Sr because intercombination transitions are more likely for heavier atoms.

3.2 Helium – europium laser

This laser, first reported in 1973 [10], was of particular importance in collision laser development. It was the first to demonstrate a transition from pulsed to quasi-continuous operation, which sparked great interest in designing collision lasers and raised great hopes that the problem would be resolved. This laser has been the subject of numerous reports [8, 10 – 25]. Here we will outline the main results.

The energy level diagram (LS coupling scheme) and transitions of the europium ion are presented in Fig. 4. Note that, according to recent NIST data, the multiplets correspond to another coupling scheme and differ slightly from those in Fig. 4. Table 1 compares the designations used by

Table 1. Designations of Eu^+ levels in the LS coupling scheme (Fig. 4) and according to NIST.

NIST designation	J	E/cm^{-1}	LS coupling
$4f^7(^8S^o_{7/2})6p_{1/2}$ (7/2,1/2)	3	23774.28	$4f^{7\,9}P_3$
$4f^7(^8S^o_{7/2})6p_{1/2}$ (7/2,1/2)	4	24207.86	$4f^7\,{}^9P_4$
$4f^7(^8S^o_{7/2})6p_{3/2}$ (7/2,3/2)	5	26172.83	$4f^7\,{}^9P_5$
$4f^7(^8S^o_{7/2})6p_{3/2}$ (7/2,3/2)	4	26838.50	$4f^7\,{}^7P_4$
$4f^7(^8S^o_{7/2})6p_{3/2}$ (7/2,3/2)	3	27104.07	$4f^7\,{}^7P_3$
$4f^7(^8S^o_{7/2})6p_{3/2}$ (7/2,3/2)	2	27256.35	$4f^7\,{}^7P_2$

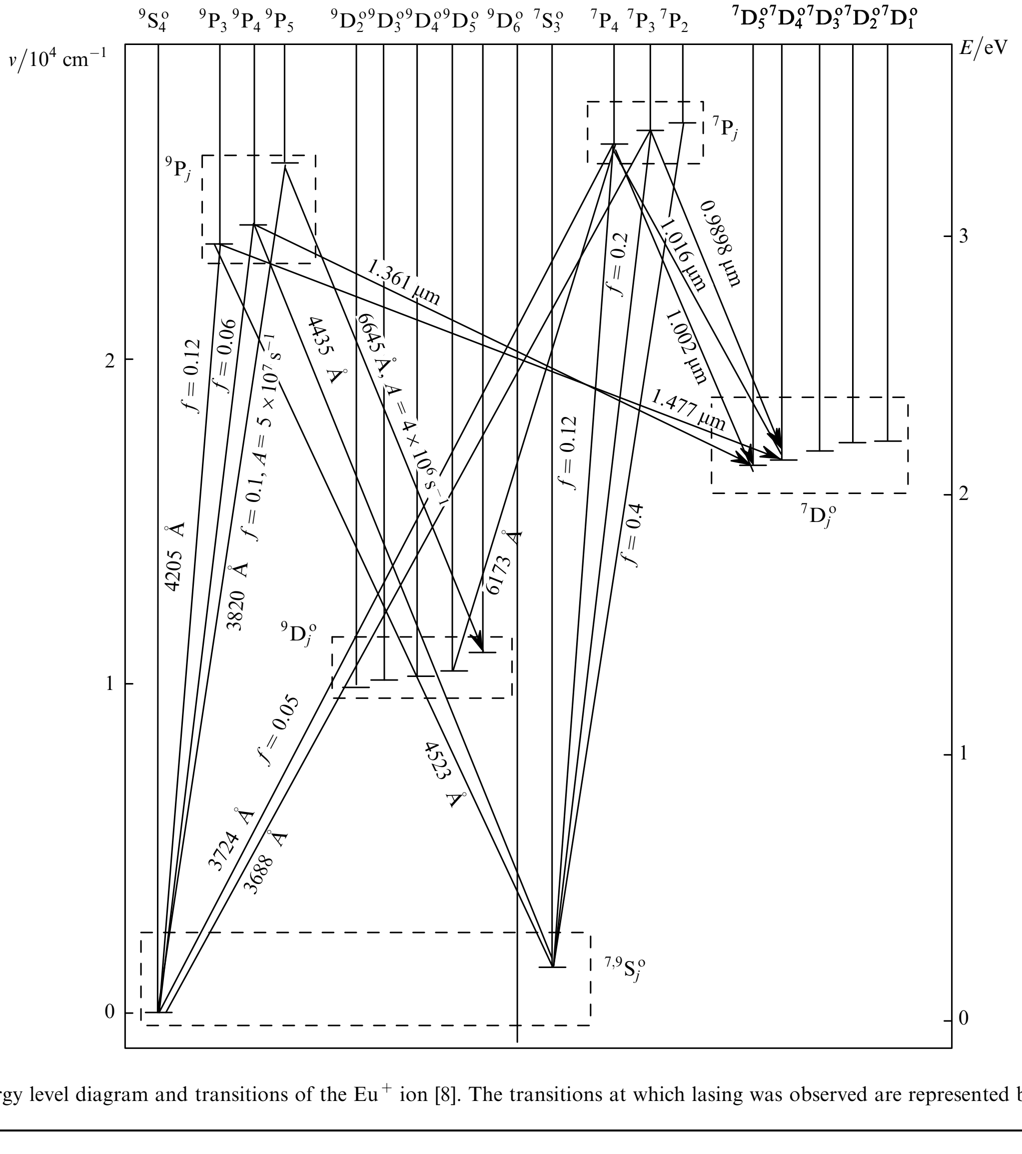

Figure 4. Energy level diagram and transitions of the Eu^+ ion [8]. The transitions at which lasing was observed are represented by arrows.

NIST and those in Fig. 4. Lasing was demonstrated on many Eu^+ lines (represented by arrows in the diagram), but particular attention was given to the 1.002-μm line, which typically ensured quasi-cw lasing.

As shown by Bokhan et al. [10], the output power increases as the He pressure is raised to ~600 Torr, and the lasing duration points to quasi-continuous operation. The $He-Eu^+$ laser was studied later in detail in both quasi-continuous and pulsed regimes. Collisional energy transfer between the laser levels and other levels was investigated by modulating the output power and examining its effect on the population of individual levels. The measured relaxation times of the lower laser levels turned out to be abnormally short [17]. The relaxation time of the lower laser level $^7D_5^o$ at an He pressure of 1 atm was estimated at 4 ns. Several studies were concerned with pulsed lasing at He pressures of up to 5 atm [21, 23, 24].

In early reports [12, 17] quasi-cw lasing was assumed to be due to the lower level depopulation by collisions with He atoms (hence, $\Delta E \approx 9kT_g$). This led to the conclusion that large depopulation cross sections of metastable levels were also possible for $\Delta E \gg kT_g$.

The results of the first studies of the $He-Eu^+$ laser, particularly the conclusion that sufficiently rapid depopulation of metastable levels by collisions with heavy particles is possible for $\Delta E \gg kT_g$, raised great hopes and generated considerable interest in designing collision lasers on atomic transitions.

Later, another mechanism of lower laser level relaxation was proposed, which assumed the fast relaxation of the $^7D_j^o$ levels to be due to recombination via autoionising states of the atom. This mechanism was probably first examined by Klimkin et al. [15] and was analysed in greatest detail in Klimkin's doctoral dissertation [8], where he concluded that the $He-Eu^+$ should be considered a new type of laser, based on ionisation/recombination processes.

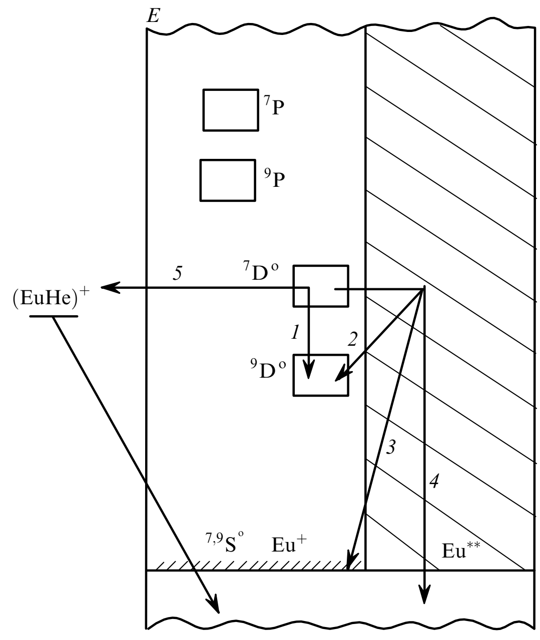

Figure 5. Relaxation scheme for the lower laser levels of Eu^+ [9]. Eu^{**} designates the autoionising levels of the Eu atom. The arrows represent (*1*) quenching, (*2*, *3*) trapping/ionisation, (*4*) recombination and (*5*) the formation of a molecular ion [8].

In our opinion, the latter mechanism – relaxation via autoionising levels of Eu – is supported by more conclusive evidence than the direct depopulation of the lower laser level by collisions with helium atoms at $\Delta E \gg kT_g$. An important point is that the relaxation via autoionising levels of the Eu atom occurs at low ΔE values.

The possible relaxation channels in this mechanism are illustrated in Fig. 5.

Assuming that the lower laser levels are depopulated by the recombination mechanism outlined above, we have to dismiss the statement that the $He-Eu^+$ lasing indicates that metastable levels can be effectively depopulated by collisions with He atoms for $\Delta E \gg kT_g$.

3.3 Laser on a tin transition

This laser utilises the $^1D_2-{}^3P_2$ transition ($\lambda = 1.9244$ μm) between two metastable states of the ground state configuration $5p^2$ of the Sn atom. The feasibility of lasing on transitions of np^2 configurations was discussed by Klimkin et al. [26]. CW 'collision' lasing in an $He-{}^{120}Sn$ mixture was reported by Bokhan [27]. The discharge was initiated in a tube 0.5 cm in diameter and 50 cm in length by applying an alternating current of ~0.6 A at a wall temperature of ~1400 °C. Because of the low transition probability, the laser gain was low, estimated at $\sim 2 \times 10^{-4}$ m^{-1}. In our opinion, the results reported in [27] provide no conclusive evidence of quasi-cw lasing because the lasing duration was much shorter than the estimated lifetime of the upper laser level.

3.4 Laser on a thulium transition

Several recent papers [28–30] have reported the creation of a laser on a Tm transition ($\lambda = 1.069$ μm) as the first metal vapour collision laser meeting Gould's criteria. The only evidence presented is that the output power increases with helium pressure, whereas lasing on other lines occurs at pressures below that at which the 1.069-μm lasing begins. This interpretation relies on the assumption that the lower laser level decays through collisions with He atoms at $\Delta E/kT_g \sim 13$, which appears unlikely. Note also that the process obviously had a pulsed character: the full width at half maximum of the pulse (~50 ns) was well below the current pulse width. No evidence of quasi-cw lasing was presented. Recall that Gould's criterion refers to cw collision lasers. The conclusion drawn in [28 30] that collision lasing on a Tm transition was achieved appears unfounded.

3.5 Other systems considered to be collision lasers

According to Bokhan [31], the barium vapour laser is a collision laser and can generate long laser pulses owing to the depopulation of the metastable state by collisions with ground-state Ba atoms and with the wall of the laser tube. He presented measured rate constants for deexcitation of the metastable Ba state ($^1D_2$) by electrons ($k_e = 0.7 \times 10^{-7}$ cm^3 s^{-1}) and ground-state barium atoms ($k_M = 1.6 \times 10^{-11}$ cm^3 s^{-1}) and determined the probability of the $^1P_1^o-{}^1D_2$ laser transition: $A = (0.9 \pm 0.1) \times 10^4$ s^{-1}.

In another report [19], Bokhan stated that quasi-cw or cw collision lasing had been achieved in Ba, Eu, Eu^+, Pb and Sr^+ vapours. Note that those reports did not contain any details of how collision lasing had been obtained or any of its characteristics (duration, power, discharge conditions or others). It was just mentioned that thin (0.3–0.4 cm in diameter) discharge tubes had been used. For a Ba vapour laser with a compact cavity in a 0.4-cm-diameter tube, the lasing duration was reported to reach hundreds of microseconds [31]. According to the later report [19], long laser pulses, occasionally up to hundreds of microseconds, were obtained in gas-discharge-pumped barium and europium vapour lasers in tubes of diameter 0.3 – 0.4 cm. Like in the case of the europium ion laser, the lasing duration is limited by the drop in electron temperature in the discharge development process.

We think that the above results should be considered only tentative because they lack any solid evidence of collision lasing. To our knowledge, there are no reports in which the characteristics of the lasers considered by Bokhan [19, 31] have been analysed as thoroughly as is necessary in the scientific literature.

The conclusions drawn by Bokhan came under criticism by Batenin et al. [32]. They proposed criteria for differentiating between self-terminating and quasi-cw lasing. Based on those criteria, they concluded that there were no grounds to regard the metal vapour lasing reported by Bokhan [19, 31] as collision lasing. To account for the quasi-cw lasing, a different inversion mechanism was proposed, which involves excitation of metastable states to ionised states [33 – 38]. This mechanism is however beyond the scope of this review because it differs markedly from the one commonly believed to underlie collision laser operation.

3.6 Collision laser with energy exchange between upper levels

In addition to collision lasers on R – M transitions, another laser scheme, based on energy transfer from one atomic species (donor) to another (acceptor), has been analysed theoretically. The donor level from which energy transfer occurs is assumed to be effectively excited by electrons, and its population is determined by T_e. The acceptor level to which energy is transferred is situated somewhat lower, is excited by electrons only weakly and has a downward transition with a moderate probability. Population inversion in such a system results from efficient population of the donor level and predominant population of the acceptor level through atomic collisions or energy transfer by collisions with the buffer gas or another gas added for this purpose. In the limit, such collisions must lead to the equilibrium populations of these levels at temperature T_g. In an ideal case, the laser efficiency in this scheme must approach the efficiency of a heat engine, η.

Most effort has been focused on a system of two two-level atoms. Population inversion in such a system results from the transition of the acceptor to its ground state. To our knowledge, this scheme was first analysed by Zalesskii [39], who considered energy transfer from an oxygen molecule in the $O_2(^1\Delta_g)$ state to an iodine atom in the $I(5^2P_{1/2})$ metastable state ($\Delta E = 279$ cm^{-1}). According to his calculations with a number of simplifying assumptions, population inversion in this scheme is possible. However, in the experiments described in his report no lasing was obtained.

Later [40], it was proposed to use this scheme with a donor atom possessing a resonance level that is easy to excite by electrons and enables energy transfer to another atom's level which is excited by electrons only weakly and has a half-forbidden downward transition. The spontaneous decay of the donor's resonance level was assumed to be strongly inhibited by resonance radiation trapping. The approximation of two two-level systems was used to calculate the saturated power with no assumptions regarding the relative rates of the processes involved. The results indicated that, in that case, population inversion was also possible. The inversion conditions were identified, and criteria for selecting donor and acceptor atoms were formulated.

As an example, calculations were made for energy transfer from Na resonance levels to the $Ca(^3P_1^o)$ level. The results indicate that, with the process parameters chosen, population inversion exists in rather wide ranges of electron and calcium concentrations.

Zalesskii [41] calculated the unsaturated gain in the scheme considered in [40] and proposed, in addition to Na – Ca, seven systems: Rb – Bi, K – Bi, Yb – Ca, Na – Bi, Yb – Ca (another Ca transition), Ca – Mg and Yb – Mg. The expected lasing wavelengths in these systems range from 457 to 875 nm. Specific calculations were made for the Rb – Bi and Ca – Mg systems under glow discharge conditions. The estimated gain coefficients are 4.9×10^{-4} and 16×10^{-4} cm^{-1}, respectively (see Ref. [41] for more details).

The excitation scheme under consideration differs from that on R – M transitions in that the energy separation between the donor level and the upper laser level is not very large (a few times kN_g). Accordingly, they are close in excitation threshold. Below the threshold energies, electrons cause little excitation of both levels. For a donor acceptor pair in which the electron excitation rate constant of the resonance level far exceeds that of the upper laser level, this will be the case at almost any electron energy. Note that, with this scheme, one can vary the ratio of the donor and acceptor concentrations, which makes population inversion easier to achieve.

The above-mentioned studies [39 – 41] dealt with two two-level systems. To bring our analysis closer to realistic conditions, we must also consider ionisation of atoms and the case when other atomic levels may play a significant role. Ionisation and transitions to higher levels may impede achievement of population inversion. A group of closely spaced levels, either involved in energy transfer between excited states or situated near the ground state of the

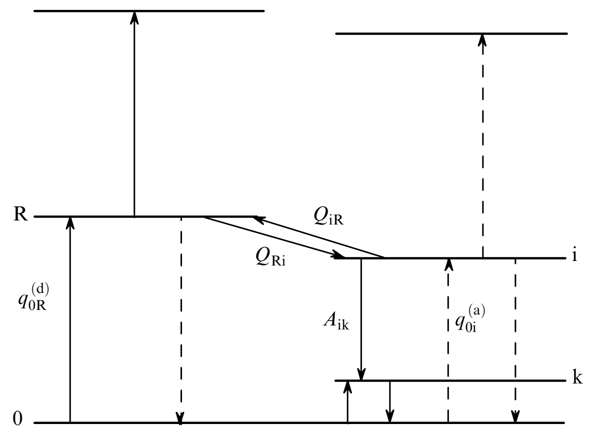

Figure 6. Energy level diagram of a collision laser with energy exchange between upper levels and collisional relaxation of the lower laser level; $q_{0R}^{(d)}$ and $q_{0i}^{(a)}$ are the rate constants of donor and acceptor level excitation by collisions with electrons.

acceptor, may improve conditions for creating population inversion. Therefore, one may consider a 'hybrid' scheme in which there is energy exchange between upper levels and the laser transition is not terminated at the ground level, so that the lower laser level is also depopulated through collisions with heavy particles, as was assumed in the case of R – M transitions. Figure 6 presents a simplified scheme of this excitation process.

To conclude this survey of systems that may be considered collision lasers, it is worth pointing out that, in the four decades since the collision laser concept was proposed, there has been only modest progress in designing such lasers. CW lasing has only been obtained in the mid-IR, with lasers that do not exactly conform to the original concept. The strategic goal to create efficient, high-power cw lasers on atomic transitions has not been achieved.

In many regards, the lasers developed to date do not meet Gould's criteria. The lower laser level is relaxed mainly at a high value of ΔE, rather than through a series of intermediate levels. The upper laser level is populated mainly via direct electron impacts, rather than from a higher level or from donor atoms. In a number of cases, cw lasing was reported without conclusive evidence.

In the next section, an attempt is made to understand why the desired results have not been achieved and to analyse the problems encountered in efficient cw collision laser development.

4. Difficulties in creating efficient collision lasers on atomic transitions

Transitions in metal atoms are believed to hold considerable promise. In most instances, to achieve the necessary metal vapour density by conventional thermal heating of the metal, a fairly high temperature is needed. The gas temperature T_g must obviously be even higher than the temperature of the metal, located on the wall of the gas discharge tube (GDT) or in a separate compartment. The greater the power delivered to the discharge and the expected power output, the higher should be T_g. Since T_e must considerably exceed T_g, this implies that T_e cannot be low: typically it should be ~ 1 eV or above. As will be shown below, this puts certain constraints on the type of discharge.

The excitation cross section of metastable levels of copper has recently been found to be rather large [42], in contrast to what was believed earlier. This is due to their excitation through negative-ion states, which was left out of consideration in the early stage of R – M laser development. Reliable data on the excitation cross sections of M levels in other atoms are still missing, but studies of their decay between pulses in R – M lasers and lasing of such systems at high pulse repetition rates suggest that the electron excitation rate of M levels in other suitable atoms is also fairly high. It is therefore necessary to revise the approaches for designing collision lasers on atomic R – M transitions.

Note first of all that the electron excitation rate of the R level exceeds that of the M level only starting at a certain value of T_e; for copper, starting at $T_e \approx 1.3$ eV (with the statistical weights of the levels taken into account) (Fig. 7). The optimal electron temperature is 2 – 4 eV. This also imposes certain requirements on T_e. Most likely, T_e must exceed 2 eV.

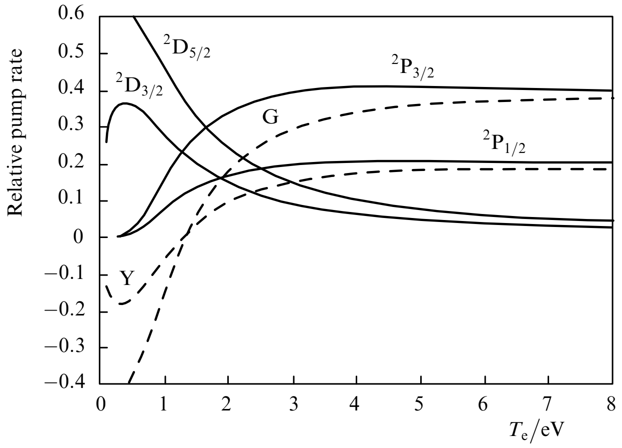

Figure 7. Relative electron pump rates of the laser levels of atomic copper from the ground state as functions of electron temperature. The dashed lines show the difference in the pump rate between the upper and lower laser levels (with allowance for their statistical weights) for the green (G) and yellow (Y) lines [43].

Another difficulty stemming from the large excitation cross section of metastable levels, σ_{0M}, which has not been considered previously, is the necessity to ensure the below inequality, desirable for creating an efficient collision laser:

$$Q_{M0}N_a = \langle \sigma^{a}_{M0} v_a \rangle N_a \gg \langle \sigma^{e}_{M0} v_e \rangle n_e = q_{M0} n_e .$$

Here, the subscripts 'e' and 'a' refer to electrons and heavy particles, angle brackets denote averaging over the velocity distribution, Q_{M0} and q_{M0} are the rate constants of metastable level depopulation by heavy particles and electrons, and v_a and v_e are the atom and electron velocities. Typically, $v_e/v_a \sim 10^4$, and σ^{e}_{M0} is hardly below σ^{a}_{M0}; therefore, N_a/n_e must far exceed 10^4. This relationship also influences the choice of the type of discharge.

5. Discharges suitable for collision lasers

The condition $T_e \gg T_g$ is of key importance in the collision laser concept. Neither Bennett [1] nor Gould [2] considered in detail which discharges were best suited for fulfilling this condition. They merely mentioned that it was fulfilled in some discharges. They seem to have meant the positive column of a glow discharge.

In the positive column of a steady-state glow discharge, T_e is determined by the relationship between the ionisation and ion loss rates [44]. If the ion loss process is governed by diffusion, as is usually the case, T_e can be found from the relation [44]

$$\left(\frac{kT_e}{I}\right)^{1/2} \exp\frac{I}{kT_e} = \mathrm{const}(pR)^2 ,$$

which represents the universal dependence of kT_e/I on cpR, where c is constant for a given gas, p is pressure, I is the ionisation potential, and R is the radius of the GDT.

This dependence [44] is illustrated in Fig. 8. T_e is seen to rapidly decrease with increasing cpR. In most of the cpR range under consideration, $T_e/I \approx 10^3$ K eV^{-1}, which corresponds to $T_e \approx 0.1I$. Since metal atoms with ionisation potentials below 10 eV are the main candidates for collision

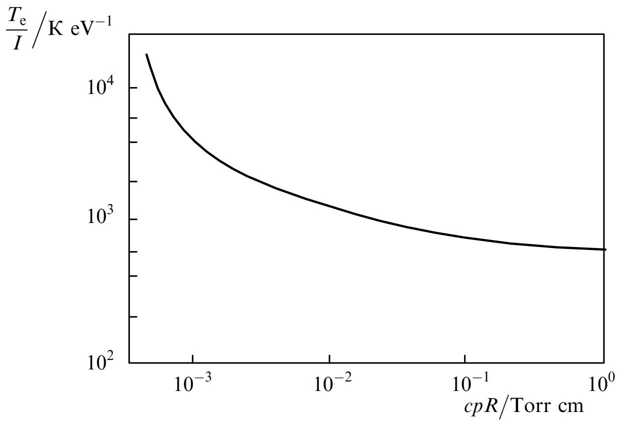

Figure 8. Universal curve for determining T_e as a function of I and cpR.

laser development, it would be expected that, in discharges produced in mixtures of metal vapours with buffer gases, where metal atoms are mainly ionised, $T_e < 1$ eV at not too low pressures. As pointed out above, collision lasing at such values of T_e is unlikely. Relatively high T_e would be expected only at low gas pressures in narrow tubes. Low pressures imply slow relaxation of the lower laser level by collisions with heavy particles, and narrow tubes limit the output power.

Thus, the positive column of a steady-state glow discharge, where the ion loss is determined by diffusion, is poorly suited for collision laser development.

5.1 Collision laser utilising an electron-attachment-controlled discharge

Great hopes may be associated with the positive column in which ion losses are determined by rapid volume recombination. However, at the T_e values necessary for effectively creating population inversion, three-particle recombination is too slow. As a rule, it cannot compete even with ambipolar diffusion. A substantially higher ion loss rate can be ensured by dissociative recombination and electron attachment followed by ion–ion recombination. In the case under consideration, when inversion occurs on metal atom transitions, high concentrations of positive molecular ions are unlikely, as is therefore dissociative recombination, although in principle it cannot be totally ruled out. For the moment, we will not consider this possibility in the case of collision lasers.

Another possibility is electron attachment to electronegative species added for this purpose. The electron temperature in the positive column will then be determined by the relationship between the ionisation rate (which is typically a strong function of T_e) and the attachment rate (which is a weaker function of T_e). Most likely, it is electron-attachment-controlled discharges which are best suited for creating collision lasers on atomic transitions. (Note in passing that an electron-attachment-controlled discharge is used to pump cw CO_2 lasers [44].)

We believe that, in this case, the most attractive cw collision laser scheme is that in which ion–ion recombination enables selective population of an atomic level, in particular, the upper laser level or a higher lying level whose energy can be transferred to the upper laser level. One possible energy level diagram corresponding to this mechanism is shown in Fig. 9.

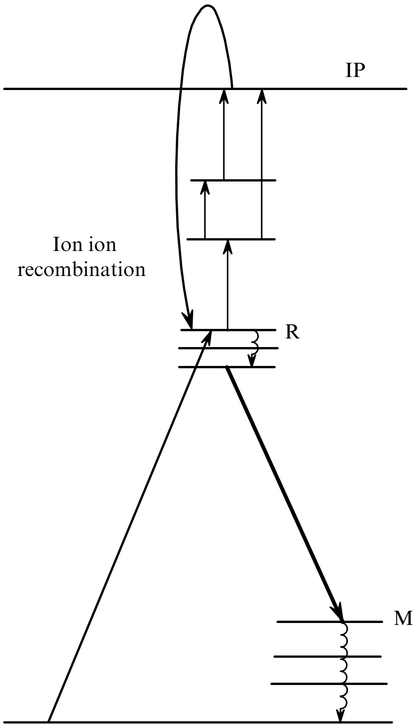

Figure 9. Possible energy level diagram of a collision laser that utilises a discharge controlled by electron attachment and ion–ion recombination. The upward-pointed arrows represent step-like excitation and ionisation by electrons, IP denotes the ionisation potential, the curved arrow represents transitions to the level during ion–ion recombination, the wavy arrows represent relaxation by collisions with heavy particles, and the slanted downward-pointed arrow represents the laser transition.

The ion–ion recombination rate constant is typically rather large: $\alpha_{ii} \sim 10^{-7}$ cm^3 s^{-1}. The recombination process can be 'directed' to selectively pump one or a few levels. In this scheme, pumping is also due to collisions between heavy particles (positive and negative ions). The released energy, typically $\Delta E \gg kT_g$, goes into pumping and partially converts to the kinetic energy of the atoms resulting from the recombination. The energy consumed for ionisation in the discharge does not go into recombination at the wall, in contrast to what occurs in a conventional glow discharge, but is directed via volume recombination to pump the desired level. This gives hope that the energy delivered to the discharge can be used more efficiently.

Since one would expect rather high T_e in discharges with volume recombination ($T_e \gg T_g$), the known schemes of collision lasers, in particular those considered above, may well be realised in combination with pumping in the course of ion–ion recombination. One possibility is energy transfer from a donor atom to an acceptor. The possible types of collision lasers that utilise ion–ion recombination will be considered in a separate paper.

6. Collisional mixing of levels and decay of lower laser levels

Knowledge of the energy transfer rate that can be achieved through collisions with heavy particles, and in particular of the rate of lower laser level depopulation by such collisions, is fundamental to the physics of collision lasers.

Energy transfer through heavy-particle collisions has been the subject of extensive studies (see, e.g., the reviews in [20, 45 – 49] and references therein). The results obtained in this area of research would require too much space to be described in detail. Here, our consideration will be restricted to the most general issues. Specific details can be found in the above-mentioned reviews.

In most cases, the effective cross section of the process

$$\mathrm{A}^{**} + \mathrm{B} \rightarrow \mathrm{A}^{*} + \mathrm{B} + \Delta E$$

(where A^{**} is an excited atom, A^{*} is the same atom in another excited state, B is another atom, which may in general also be in an excited state, and ΔE is the energy released or absorbed in this process, which increases or reduces the kinetic energy of the atoms) drops sharply with increasing ΔE. This is however not always the case. In some cases, the cross section of this process may increase with ΔE [47, 48]. An essential role is then played by the relative positions of the potential curves representing the interaction between colliding particles and between the products. If the potential curves are closely spaced at a point or there is a quasi-intersection, the process may have a large cross section.

When an A^{**} atom collides with a molecule, the internal degrees of freedom of the molecule come into play. The process may have a resonance character, with a large cross section, because some of the released energy goes into excitation of vibrations and rotations. Comparatively large cross sections are also typical of collisions resulting in chemical reactions.

6.1 Collisional relaxation of metastable levels

Since R – M transitions have been proposed for collision laser development in the vast majority of cases, data on collisional relaxation of metastable levels are of special interest. This issue has received considerable attention in the literature.

For example, Donovan and Husain [46] discussed possible relaxation mechanisms and presented a large amount of data on the relaxation of metastable levels of atomic C, Si, Ge, N, P, As, O, S, Se, Te, F, Cl, Br and I.

Since these species are not thought to be suitable for collision laser development, we will not present here data from that review. We note only that the relaxation of their metastable levels by collisions with inert-gas atoms has low efficiency. The relaxation rate, as a rule, increases in going from light to heavy inert gases and may be rather high for Xe. Relatively fast relaxation is typically ensured by collisions with molecules.

Table 2. Relaxation rate constants of the Cu $3d^9 4s^2 (^2D_{5/2})$ level for collisions with atoms and molecules [50].

Process	Rate constant/cm^3 s^{-1}	
	CuI	CuCl
Cu($^2D_{5/2}$) + He	below $(5.1 \pm 1.5) \times 10^{-16}$	–
+ Ar	below 1.2×10^{-16}	–
+ Xe	below $(4.7 \pm 1.1) \times 10^{-16}$	–
+ N_2	below $(4.9 \pm 1.1) \times 10^{-16}$	–
+ CO	below $(5.8 \pm 1.8) \times 10^{-16}$	–
+ CO_2	below $(1.1 \pm 1.3) \times 10^{-16}$	–
+ SF_6	$(6.4 \pm 1.1) \times 10^{-15}$	$(13 \pm 2) \times 10^{-15}$
+ O_2	$(3.5 \pm 0.3) \times 10^{-12}$	$(1.1 \pm 0.1) \times 10^{-12}$

Note. Metastable copper atoms were produced by pulsed photolysis of CuCl and CuI at $T \sim 600$ K, which approximates the operating conditions of copper halide lasers [50].

Below, we present data on the relaxation of metastable levels of some atomic metals, mostly those which have demonstrated pulsed lasing on R – M transitions.

Relaxation of metastable levels of Cu. It seems likely that the laser on R – M transitions of copper would be of special interest for collision laser development because it demonstrates the highest efficiency and output power.

The relaxation rate constant of the $3d^9 4s^2$ ($^2D_{5/2}$) metastable level of the copper atom was measured by Trainor [50] for collisions with He, Ar, Xe, N_2, CO, CO_2, SF_6 and O_2. The results are summarised in Table 2.

Hao-Lin Chen and Ebert [51] measured the relaxation and collisional mixing cross sections for the Cu laser levels ($^2P_{3/2}$, $^2P_{1/2}$, $^2D_{5/2}$, $^2D_{3/2}$) with He, Ne, Ar, H_2, D_2, N_2, CO and Cu as collision partners. Atoms were excited optically on resonance transitions by the frequency-doubled radiation from a dye laser. The results are presented in Table 3.

Table 3. Relaxation and collisional mixing cross sections (cm^2) for copper levels ($T_g = 1600$ K) [51].

Gas	$^2D_{5/2} \rightarrow {}^2S_{1/2}$	$^2P_{1/2} \rightarrow {}^2P_{3/2}$	$^2D_{3/2} \rightarrow {}^2D_{5/2}$
He	$\leqslant 5.3 \times 10^{-22}$	4.0×10^{-16}	$\leqslant 5.7 \times 10^{-20}$
Ne	$\leqslant 7.3 \times 10^{-22}$	3.8×10^{-17}	$\leqslant 2.8 \times 10^{-20}$
Ar	$\leqslant 1.2 \times 10^{-21}$	3.4×10^{-16}	$< 10^{-20}$
H_2	6.8×10^{-21}	1.7×10^{-15}	3.6×10^{-18}
D_2	6.9×10^{-21}	1.1×10^{-15}	2.3×10^{-18}
N_2	8.3×10^{-21}	2.4×10^{-15}	2.0×10^{-18}
CO	–	–	1.7×10^{-17}
Cu	4.8×10^{-17}	8.7×10^{-15}	1.7×10^{-15}

For the convenience of analysis, recall the energy positions of the levels in question:

Level	E/cm^{-1}	ΔE/cm^{-1}
$^2D_{5/2}$	11203	$\Delta E(^2D_{3/2} - {}^2D_{5/2}) = 2042$
$^2D_{3/2}$	13245	
$^2P_{1/2}$	30535	$\Delta E(^2P_{3/2} - {}^2P_{1/2}) = 248$
$^2P_{3/2}$	30783	

In addition, Hao-Lin Chen and Ebert [51] discussed the possible mixing and relaxation mechanisms and estimated the pressures of various gases necessary for achieving the equilibrium populations of the levels involved and for their relaxation in a given time interval. The estimates are listed in Table 4.

Based on these results, they concluded that the gases studied were incapable of effectively depopulating the metastable (lower laser) levels of copper. They pointed out that, at high atomic copper concentrations (pressures above 0.7 Torr), the metastable level depopulation by collisions with copper atoms could ensure a pulse repetition rate of 20 kHz. However, copper vapour lasers typically

Table 4. Gas pressure (Torr) necessary for achieving the equilibrium population of copper levels and for their relaxation through the specified channels in the specified time interval [51].

Gas	$^2P_{1/2} \rightarrow {}^2P_{3/2}$ (50 ns)	$^2D_{3/2} \rightarrow {}^2D_{5/2}$ (50 ns)	$^2P_{1/2} \rightarrow {}^2P_{3/2}$ (100 ns)	$^2D_{3/2} \rightarrow {}^2D_{5/2}$ (100 ns)	$^2D_{5/2} \rightarrow {}^2S_{1/2}$ (50 μs)
He	40	2.8×10^5	20	1.4×10^5	$\sim 2 \times 10^4$
Ne	640	8.6×10^5	320	4.5×10^5	3×10^4
Ar	95	–	47	–	$\sim 2 \times 10^4$
H_2	5	2.4×10^3	3	1.2×10^3	1×10^3
D_2	15	6.9×10^3	8	3.4×10^3	2×10^3
N_2	11	1.3×10^4	5	6.6×10^3	$\sim 3 \times 10^3$
Cu	4	22	2	10	0.7

operate at substantially lower copper concentrations, and relaxation of the lower level is controlled by collisions with cooling electrons.

Hao-Lin Chen and Ebert [51] pointed out that the mixing of the $^2P_{3/2}$, $^2P_{1/2}$ upper laser levels was a rather fast process. For this process to have a significant effect on the relative intensities of the green and yellow lines, the buffer gas and copper vapour pressures must considerably exceed those commonly used in the copper vapour laser.

The relaxation of metastable levels of copper was also investigated by Bokhan [19, 20] using resonance fluorescence measurements in the afterglow plasma of a pulsed discharge. The relaxation cross section of the Cu($^2D_{5/2}$) level for collisions with He was within 10^{-20} cm^2, and that for collisions with ground-state copper atoms was 10^{-17} cm^2.

Relaxation of metastable levels of Pb. Collisional relaxation of metastable levels of atomic Pb was studied in [52–55]. Without going into the details of those studies, we present only their results in Table 5, which lists the relaxation rate constants obtained. Metastable atoms were produced by pulsed photolysis of tetraethyllead [52, 53] and tetramethyllead [54, 55].

Trainor and Ewing [55] measured the relaxation rate constants of the $^3P_1$ and $^3P_2$ levels at temperatures from 300 to 600 K. The rate constants were found to increase with temperature, but only gradually.

As can be seen from Tables 2 and 5, the relaxation rate constants of both Pb and Cu metastables for collisions with inert-gas atoms are rather small (in the order of 10^{-16} to 10^{-15} cm^3 s^{-1}). The rate constants for collisions with molecular species are considerably larger, especially in the case of O_2: in the order of 10^{-11} to 10^{-10} cm^3 s^{-1}. The higher rate of relaxation by collisions with molecular species is mostly due to E–V exchange processes and in some cases to chemical reactions.

Bokhan [19, 20] reported measured relaxation cross sections and rate constants for the Pb $^1D_2$ and $^1S_0$ levels. For both levels, the cross sections of relaxation by collisions with He were within 2×10^{-20} cm^2. Since the gas temperature was not determined, only a rough estimate can be made. Taking the velocity of colliding particles to be 10^4 cm s^{-1}, we obtain a rate constant below 2×10^{-16} cm^3 s^{-1}. In the case of Pb–Pb collisions, the rate constants for the $^1D_2$ and $^1S_0$ levels were reported to be 4.5×10^{-11} and 9×10^{-11} cm^3 s^{-1}, respectively. The respective cross sections are 1.4×10^{-15} and 3×10^{-15} cm^2.

Table 5. Relaxation rate constants (cm^3 s^{-1}) of Pb levels for collisions with atoms and molecules [54]; $T_g \sim 300$ K.

Gas	$^3P_1$	$^3P_2$	$^1D_2$	$^1S_0$
TML	4.0×10^{-11}	3.1×10^{-10}		
TEL	$(1.4 \times 10^{-11})^*$	(5.8×10^{-11})	(5.6×10^{-11})	(3.7×10^{-11})
He	(1.1×10^{-16})	$(< 1.3 \times 10^{-17})$	$(< 2 \times 10^{-16})$	
Ar	$< 2.3 \times 10^{-16}$	$< 2.0 \times 10^{-15}$		
	(1×10^{-16})	(2.0×10^{-15})		
Xe	$< 6 \times 10^{-16}$	$< 2.3 \times 10^{-15}$	$(< 1 \times 10^{-15})$	$(< 2 \times 10^{-15})$
H_2	$< 5.7 \times 10^{-15}$	1.5×10^{-12}	$(< 1 \times 10^{-14})$	$(< 1 \times 10^{-15})$
	(2.9×10^{-15})	(1×10^{-12})		
D_2	$< 5.5 \times 10^{-15}$	8.7×10^{-12}		
	$(< 6 \times 10^{-16})$	$(< 1 \times 10^{-12})$		
O_2	4.5×10^{-11}	4.6×10^{-11}		
	(7×10^{-12})	(4×10^{-11})	(1.1×10^{-10})	(1.2×10^{-10})
N_2	1.7×10^{-15}	4.1×10^{-13}		
	(2.0×10^{-15})	(8×10^{-15})	$(< 1 \times 10^{-15})$	(1.6×10^{-15})

*The rate constants given in parentheses were taken from [52, 53]; TML – tetramethyllead, TEL – tetraethyllead.

Table 6. Collisional relaxation cross sections for the Tl (6p $^2P_{3/2}$) level [56].

Gas	$\sigma/10^{-16}$ cm^2
He	$< 2 \times 10^{-3}$
Ne	$< 2 \times 10^{-3}$
Ar	$< 2 \times 10^{-3}$
TlI	4.4
TlBr	15
TlCl	39
N_2	$< 2 \times 10^{-3}$
CO_2	$< 2 \times 10^{-3}$
H_2	0.037
CO	0.010
NO	5.2
O_2	28
CH_4	0.026

Relaxation of a metastable level of Tl. Collisional relaxation of the Tl 6p^{2}P$_{3/2}$ metastable level ($E = 7793$ cm^{-1} $= 0.97$ eV) was studied by Bellisio and Davidovits [56]. Metastable thallium atoms were produced by pulsed photodissociation of thallium halides in the range 285 – 400 °C. The results are presented in Table 6. One can see that the cross sections of relaxation by collisions with inert-gas atoms, N_2 and CO_2 are also small. Collisions with some other molecular species, in particular with O_2, NO and thallium halides, ensure much faster relaxation.

Bokhan [19, 31] measured the rate constants and cross sections of various deactivation processes for metastable levels of atoms and ions whose R – M transitions had been made (or expected) to lase. He presented the rate constants and cross sections for the deexcitation of the Ba(^{1}D$_2$), Cu(^{2}D$_{5/2}$), Eu(a^{8}D$^{\circ}$, b^{8}D$^{\circ}$), Mn(a^{6}D), Pb(^{1}S$_0$, ^{1}D$_2$), Sn(^{1}S$_0$), Ba$^+$(^{2}D$_{5/2}$), Ca$^+$(^{2}D), Eu$^+$(a^{9}D$^{\circ}$), and Sr$^+$(^{2}D) metastable levels through collisions with electrons (k_e), ground-state metal atoms ($k_{\rm Me}$) and buffer-gas atoms ($k_{\rm He}$, $k_{\rm Ne}$) and also through resonance charge transfer (k_+).

Those measurements were performed in the afterglow plasma of a dc discharge which was periodically switched off by a thyratron, or in that of a low-current pulsed discharge (pulse duration of 0.05 – 0.3 μs), using for the most part the resonance fluorescence resulting from transitions from the metastable level to a resonance level during exposure to appropriate laser radiation. The emission intensity was measured as a function of the delay relative to the excitation pulse. To separate the numerous deexcitation channels, conditions were chosen so that one channel was expected to prevail over the others. One parameter of the system, e.g., the metal atom or buffer gas concentration, was varied, whereas the other parameters were assumed to remain unchanged. That assumption appears insufficiently substantiated because plasma characteristics are usually interrelated, and, varying one of them, one cannot be absolutely sure that the others remain unchanged. Moreover, neither the spectral composition of the exciting laser beam nor the absorption spectrum on the excitation line was monitored during the measurements. In addition, Bokhan [19, 31] did not specify how he had verified the lack of self-absorption on the transition from the resonance level. It is well known that, without checking the emission and absorption spectra and self-absorption on the transition of interest, large errors may arise in such measurements. For this reason, the results under discussion do not appear reliable.

Detailed results of those measurements can be found in the table in Ref. [19]. Here we note only that k_e is in the order of 10^{-8} to 10^{-7} cm^3 s^{-1}, $k_{\rm Me} \sim 10^{-11}$ to 10^{10} cm^3 s^{-1} ($\sim 10^{-9}$ cm^3 s^{-1} for europium), $k_{\rm He} \sim 10^{-12}$ cm^3 s^{-1}, $k_{\rm Ne}$ is slightly smaller than $k_{\rm He}$, and $k_+ \sim 10^{-10}$ cm^3 s^{-1}.

In most cases, analogous results from other, independent sources are missing. The only exception is the Ba laser transition (^{1}P$^{\rm o}_1 - {}^1$D$_2$) probability A. According to Bokhan [31], $A = (0.9 \pm 0.1) \times 10^4$ s^{-1}, which is about a factor of 28 lower than the recently reported $A = 2.5 \times 10^5$ s^{-1} [57, 58]. This casts doubt on the reliability of the measurement procedure and results in [19, 31].

The data on the deexcitation of atomic levels by collisions with heavy particles in the above-mentioned reports indicate that the deexcitation of atomic levels by collisions with atoms of the light inert gases He and Ne, which are used in the vast majority of cases as buffer gases, is ineffective, especially at high ΔE. A considerably more effective process is the depopulation of atomic levels through collisions with molecules and atoms of heavy inert gases, particularly with those of Xe. However, when used as buffers or quenchers with significant densities, these gases may substantially reduce T_e attainable in a steady-state discharge because of the large excitation and ionisation losses, in particular through vibrational excitation of molecules, in comparison with those when He and Ne are used. Previous results for pulsed R – M lasers, such as copper, lead and manganese vapour lasers, show that even small amounts of Xe substantially degrade the laser performance or fully suppress laser action.

An appreciable deactivation rate of metastable levels can be achieved by 'chemical cleaning', as exemplified by the laser on the Ca ^{1}P$^{\rm o}_1 - {}^1$D$_2$ transition in the mid-IR ($\lambda = 5.547$ μm, see Section 3.1). Note, however, that chemical cleaning removes laser working atoms from the active medium, thereby terminating the lasing process for a relatively short period of time. An effective means is then needed to recover the working atoms in the active medium after a chemical reaction that depopulates the lower laser level.

Of special note is the feasibility of rapid deexcitation of metastable levels by collisions with helium atoms at $\Delta E \gg kT_g$. The claim that this is possible relies on the fast relaxation of the lower laser level population in the helium – europium laser on Eu$^+$ transitions (see Section 3.2), which was accounted for by deexcitation through collisions with helium. Another explanation for the fast relaxation process assumes recombination through autoionising states of the atom in quasi-resonance processes at relatively low ΔE. In our opinion, this mechanism is much better substantiated than direct deactivation through collisions with He at $\Delta E > 9kT_g$. At least, we do not know of any reports that present evidence against the recombination mechanism. Nevertheless, Bokhan [59] has recently stated again, without discussing the recombination mechanism, that the fast relaxation of the lower laser level population in the above-mentioned laser on Eu$^+$ transitions is due to direct collisions with helium atoms at $\Delta E > 9kT_g$. He also holds that this mechanism is responsible for the relaxation of the metastable levels of the Eu atom and Ca^+ ion. Thus, there is no consensus as to the nature of the fast relaxation at $\Delta E \gg kT_g$. Further research is needed to shed light on this problem.

7. Conclusions

It follows from the above that the problem of creating an efficient cw collision laser that operates on atomic transitions and conforms to the concept proposed by Bennett [1] and Gould [2] has proved more challenging than anticipated at the beginning of research in this area. Many reports presented rather optimistic predictions regarding collision laser development and no less optimistic estimates of the performance of such lasers. However, the actual progress made in the four decades of efforts in this direction appears rather modest. Indeed, moderate-power cw lasing has only been achieved on the $^1P_1^o - {}^1D_2$ R–M transitions of atomic calcium and strontium in the near-IR at 5.547 and 6.457 μm. In the case of the laser on a Ca transition, cw lasing has been obtained through chemical cleaning of the lower laser level.

Quasi-cw lasing at ~1 μm on Eu^+ transitions has been well documented. Several papers reported quasi-cw lasing on other transitions of atoms and atomic ions, but neither the experimental conditions nor lasing characteristics were described in sufficient detail, and those results have not been supported by any solid evidence in subsequent work. Thus, the strategic goal of the collision laser concept-to create efficient, high-power cw lasers on atomic transitions-has not been achieved.

In most cases, collision lasing was attempted on transitions from resonance levels to metastable levels (R–M transitions) of metal atoms and ions, with lower laser level relaxation through collisions, primarily with helium and ground-state metal atoms, at high released energies ($\Delta E \gg kT_g$). The above data on relaxation by collisions with heavy particles indicates that the relaxation rate in collisions with light inert gases at high ΔE is very low. The relaxation rate in collisions with ground-state atoms is usually substantially higher, but, at the atomic metal concentrations that are commonly expected to ensure lasing in a discharge, it is unlikely to be sufficient for the required lower laser level relaxation.

Relatively high relaxation rates were reported for collisions with many molecular species. The energy released in collisions may go into the excitation of vibrational and rotational levels. It is not yet clear what molecules and at what concentrations can be used in a particular system for lower laser level deexcitation without impairing the pumping of the upper laser level.

High relaxation rates are ensured under favourable conditions by chemical cleaning. As mentioned above, chemical cleaning of the lower laser level in the laser on a Ca transition enabled cw lasing with a duration limited by the removal of calcium atoms from the discharge as a result of chemical reaction.

Special attention should be given to the relaxation of metastable levels of ions. The example of the $He-Eu^+$ laser demonstrates that the relaxation rate of the metastable level (lower laser level) of Eu^+ increases with helium pressure and is rather fast at considerable pressures (relaxation time of ~5 ns at a pressure of ~1 atm). This led to the conclusion that fast relaxation by collisions with He was possible at $\Delta E \approx 9kT_g$. That conclusion, however, appears insufficiently substantiated because another relaxation mechanism has been proposed, which assumes the relaxation by collisions with He to be due to recombination of metastable ions via autoionising states of the atom. This mechanism applies only to the relaxation of ion levels and seems to be similar to the resonance mechanism, i.e., to take place at low ΔE values.

Given that only modest advances have been made in collision laser development, the crucial question is what obstacles have to be overcome for the collision laser concept to become reality?

In our opinion, the major obstacles are as follows:

1. The relaxation of the lower laser levels of atoms in collisions with buffer gas atoms and ground-state metal atoms is too slow.

2. In contrast to what was initially assumed, the electron excitation cross sections of metastable levels of atoms are not small. As a result, the population of the resonance levels prevails over that of the metastable levels only above $T_e \sim 1$ eV. The optimal T_e is even higher. This poses the question of in what discharges and under what conditions the desirable T_e can be achieved.

3. The above analysis shows that the positive column of a glow discharge, where the ion loss is determined by diffusion, is poorly suited for efficient collision laser development because, in the presence of metal vapour, the required T_e can only be attained at low gas pressures and in narrow discharge tubes. In our opinion, one reason why only modest advances have been made in collision laser development is that little attention has been paid to the choice of the type of discharge.

4. Reliable data on many of the processes that determine the operation of collision lasers on atomic transitions are still missing.

The question that now arises is what else can be done to create an efficient collision laser on transitions of atoms or atomic ions? Note first of all that the attempts made to date to develop a collision laser implemented not all of Gould's recommendations. In almost all cases, the lower laser level was relaxed through a single, high-ΔE transition, rather than through a group of closely spaced levels. The upper laser level was populated by direct electron impacts and not from a higher lying donor level. Attempts to achieve cw lasing utilised a glow-type discharge, and the electron temperature was not measured.

Efforts should probably be focused on the search for systems where collisional relaxation would occur not through a single, high-ΔE transition but through a group of closely spaced levels, preferably in both the pumping of the upper laser level and deexcitation of the lower level. Use should also be made of discharges controlled by volume recombination during electron attachment followed by ion ion recombination. It is very desirable to substantially extend the range of data on the characteristics of the processes that determine the operation of collision lasers. It should be kept in mind that creating an efficient, high-power laser on atomic transitions will hardly be an easy-to-solve problem.

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