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STUDY OF THE RECOMBINATION GASDYNAMIC LASERS ON ELECTRONIC TRANSITIONS (VISIBLE GDL). CURRENT STATUS

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Abstract—In report current theoretical and experimental studies on development of a new molecular electronic transition gas flow lasers on the base of S+S, Cl+Cl, Br+Br radiative recombination reactions are revued.

1. -Introduction.

High power short wavelenght lasers on electronic transitions (ET) are extremely attractive for many scientific and industrial application. But all existed now visible and UV gas lasers operate in pulsed regime (rare gas-galide excimers, halogens and others). As known the convective-flow techniques can be used to provide more efficient methods for achieving a continuous wave (CW) laser action. However, there is only one CW chemical oxygen-iodine laser operating on atomic ET of iodine in near IR spectrum [1]. CW lasers on molecular ET in visible spectrum have not been demonstrated yet. In the case of success, we expect a discovery of new generation of powerful and efficient gas flow lasers (GFL) [2-3].

2. -General Conception. Background.

In principle, from an operational viewpoint a visible GFL is similar to a well-developed CW HF/DF, CO₂ - or CO-GDLs operating on vibration-rotation transitions (VRT) in mid IR spectrum. But to extend the idea of GFL from IR to visible region (from VRT to ET), some physical and technical nontrivial problems should be overcome.

The *general conception* about the specific of ET GFL may be given without going into particulars of laser action [2]. One of the *main difficulties* is to produce a good intensive electronically excited states (EES) excitation, the *other* is to provide the conditions for population inversion during EES relaxation in gas flow. The integration of these hard-combined requirements imposes the strong limitations on the properties and parameters of active medium and dictates the choice of methods for laser pumping and operational regimes.

In Table 1 comparisational characteristics for different types of laser transitions are presented: pumping rate R^* required for a light gain $\alpha \sim 10^{-3}/\text{cm}$, specific power of spontaneous emission W_{sp} under the same conditions and stimulated emission cross-section σ_{sp} are presented. As can be seen, for a change-over from IR to visible lasing the rate of pumping should be increased by a factor of 10^3 or more. On the other hand, a molecular ET has a broad bandwidth, that leads to small cross-section of stimulated emission (in comparison with the atomic ET), thus large EES density necessitates for reasonable gain.

Table 1

Value	Molecular VRT	Atomic ET (bound)	Molecular ET (bound)	Molecular ET (continuum)
$\lambda, \mu\text{m}$	2.7...2.9	0.5	0.5	0.5±0.05
$R^*, / \text{cm}^3 \text{s}^{-1}$	10^{17}	5×10^{17}	10^{21}	10^{22}
$W, \text{W/cm}^3$	10^{-5}	10^{-3}	5	60
σ_{sp}, cm^2	10^{-15}	5×10^{-14}	10^{-17}	10^{-18}

In practice, these conclusions result in the high pressure active medium necessary to use. Therefore, the commonly used reactive flow mixing methods for visible GFL pumping could not be useful so must bring some new problems. In addition, quite a number of further important conditions should be borne in mind (reaction stability, inversion range, medium optical homogeneity, etc.) for reaching a laser effect. Still one difficulty is a lack of reliable kinetic and spectral data (EES excitation, quenching and relaxation rates, radiative lifetimes and absorption cross-sections, etc.) crucial to the selection of suitable laser molecules and chemical reactions. All this gained the impression that the visible GFLs development would require major efforts than for conventional IR GDLs.

Radiative recombination of atoms and free radicals in gases presents an important class of highly exoenergetic chemiluminescence reactions. It attracts our attention potentially as most perspective way to create a thermally (or chemically) pumped visible recombination GDL (RGDL). Unlike usual IR GDLs on VRT, RGDLs can use EES with excitation energy up to 4 eV. In contrast with relatively small fraction of vibrational energy, the energy that can be stored in the breaking chemical bonds under molecular dissociation and partitioned into EES under atomic recombination amounts to 60%. If it could produce the inversion with a quantum efficiency of 50%, one could create the laser with an efficiency up to 30%.

The simple scheme of recombination pumping mechanisms is

Here, A and B are recombining atoms, process (1) is a recombination into the ground ES of product molecules AB, (2) - recombination into EES, (3) - collisional quenching of EES, (4) - vibrational relaxation of ES and (5) - radiative ET to generate the laser output. (1) and (3) compose a "dark" channel of recombination, (2) and (5) determine a "light" channel. The reactive branching ratio into EES can be up to 0.25...0.75.

This kinetic mechanisms can provide the high rate of EES production and large enough quantum yield. The recombination reaction is a three-body process, its rate is $R = k_r [A][B][M]$. A usual order of magnitude of reaction constant is $k = 1 \times 10^{-33} \text{ cm}^6/\text{s}$ (over range of temperatures $T \leq 1000 \text{ K}$). Therefore, to reach a required value of EES pumping rate $R^* = 1 \times 10^{21} / \text{cm}^3 \text{s}$ the concentration of atoms should be as high as $[A] = [B] = 1 \times 10^{18} / \text{cm}^3$ (dilution $[M]:[AB] = 1:5$). It means the process should be proceeded at strong chemical nonequilibrium conditions to shift toward to EES production. To do it, the characteristic time of gas flow, $t_{\text{gas}} = 1/(\text{dln}T/\text{dt})$, should be much less than the recombination time, $t_{\text{rec}} = 1/k [A][M]$.

The case of achieving a population^{rec} inversion varies with the parameters of EES and presents a complex trade-off between gain and loss mechanisms. For most suitable lasants due to a shift of EES potential energy curve relative to the ground ES and because of

favourable Frank-Condon factors, a partial population inversion should be created between the low v -levels of the lasing EES and the high v -levels of the ground ES essentially unpopulated at low operational temperatures. This occurs necessarily when VT-relaxation is more rapid than EES-desactivation.

The bandwidth of radiative ET depends on both the emitting EES and the ground ES potential energy curves, as well as the distribution of excited states among the various vibrational and rotational states of laser levels. Most molecules also have metastable EES that lie close and sometimes below the upper laser level. These EES act as energy reservoirs for excitation that can funnel through the radiative EES.

To our knowledge we have a sufficient background for the GDLs: a) there are specific diatomic molecules as a potential lasants and favourable gas-phase chemiluminescence processes going by scheme (1)-(5) for which kinetics, spectrum and other properties are well known (halogens, interhalides, sulfur and other candidates);

b) high degree of dissociation of this molecules is reached at practicable temperatures of the plenum gas: for the Br² with dissociation energy $D \approx 2$ eV the characteristic temperature is $T_d = 2000$ K, for Cl² - $D \approx 2.5$ eV, $T_d = 3000$ K, for S₂ - $D \approx 4$ eV, $T_d = 4000$ K; that allows to produce a high atom densities in the plenum chamber under heating by electrical arc, shock waves, explosion or combustion (there are some fuels can contain the necessary species and can supply the necessary temperature and pressure regimes);

c) dissociated gas can be cooled to temperatures below 1000 K fast enough in the supersonic nozzle flow during the time $t_{\text{rec}} \approx 10^{-6}$ s $\leq t_{\text{rec}} \approx 10^{-4}$ s to initiate a EES recombination pumping^{gas} followed by population inversion; the supersonic flow (Mach number $\sim 3 \dots 5$) also provides to rapid remove a waste species and heat release following laser action; the presence of diluent in gas composition promotes to extend the inversion zone along the cavity flow up to a few tens cm.

The typical conditions selected in view of theory predictions [2] for the RGDLs demonstration are presented in Table 2.

Table 2

Gas composition	T ₀ K	P ₀ atm	h* mm	M	T K	P atm	α cm ⁻¹	λ μm
Br ² :Ar = 1:5	2200	30	0.2	5	300	0.10	0.002	1.2
Cl ² :Ar = 1:5	3000	30	0.2	4	500	0.15	0.003	1.0
S ₂ :Ar = 1:5	4000	30	0.2	3.5	650	0.20	0.005	0.5

3. -Results of Pre-Laser Studies. Critical Review.

Because a complexity of RGDL's development, it takes the special strategy of research: first of all, to carry out the pre-laser gasdynamic photochemical studies and only then to work out the RGDL-like machine. The elements of such strategy have been tested by our Advanced Laser Research Group in Kiev [2,4-7].

The method of heating-fast cooling in supersonic flow conditions was realized for Cl² (B-X) EES recombination pumping and population inversion formation² in the shock tube gasdynamic pre-laser experiments [4-6]. The optical (shadow technique and interferometry) and spectroscopy (emissivity and absorptivity technique) flow visualization in complex with numerical flow modeling were used. Extremely intensive, up to 0.1 W/cm², spontaneous emission in near IR with quantum yield up to 0.001 photon/molecule was observed. Using the calibrated multichannel spectroscopic measurements ($\lambda = 325$, $\lambda = 441.6$, $\lambda = 1008 \pm 8$ nm) in various diagnostic sections h/h^* of tested² nozzles, population of ground ES [Cl² (X)] and vibrational temperature T have been defined from the gas absorptivity A_λ and population of optically active EES [Cl² (B)] - from the gas emissivity I_λ . Radiative lifetime

$\tau(B, v'=0) = 50 \mu s$ has been found from the preliminary measurements of I_{λ} in shock waves. The typical conditions and results of experiments are summarized in Table 3.

Table 3

h^* mm	T_0 kK	P_0 atm	Cl ₂ :Ar	$I\lambda$ /cm ⁴ s	A_1 %	A_2 %	T K	T_v K	Cl ₂ (X) /cm ³	ΔN /cm ³	α /cm
.11	3.3	20	1:0	1×10^{16}	-	-	365	1230	6×10^{16}	2×10^{14}	.003
.11	2.6	19	1:2	2×10^{15}	-	-	252	1120	8×10^{16}	4×10^{14}	.002
.18	3.4	41	1:0	8×10^{15}	-	-	620	980	2×10^{17}	8×10^{13}	.001
.60	2.4	18	1:4	2×10^{15}	38	7.9	430	850	5×10^{17}	2×10^{13}	.0005

Fig.1. -Evolution of recombining gas flow parameters and population inversion formation versus distance x/h^* in nozzle $h^*=0.18$ mm, $\theta^0=30^\circ$ for plenum conditions $T_0=3530$ K, $P_0=17$ atm, pure chlorine. I - observation.

initial thermodynamic parameters $T_0 = 2500-4000$ K, $P_0 = 10-50$ atm, gas composition Cl₂:Ar=1:0-1:8 and nozzle flow regimes showed that the value of light² gain $\alpha \sim 10^{-3}$ /cm is to be in good agreement with the theoretical predictions and sufficient to excite a laser oscillation. Gasdynamic quality of supersonic flow of recombining gas in the designed part of nozzles also was found to be a good enough.

The recombination scheme (1)-(5) under flash photodissociation of Br₂ was realized in photochemical pre-laser experiments [7], and light gain up to 5×10^{-4} /cm in $1.06 \mu m$ spectral region and specific laser energy up to 7 J/l was achieved without any optimization. (Potentially specific laser energy can be increased by order of magnitude, and high efficiency up to 5...10% can be anticipated).

Recently the gasdynamic recombination studies for Cl₂ and S₂(B-X) systems have been started by LASERDOT (France) with the aim to obtain a chemically driven visible or near IR GDL [8]. Unfortunately, the conditions of these experiments are insufficient for laser action, but they are promising in progress.

By a computational modeling, the set of important characteristics of Cl₂(B-X) kinetics was determined: recombination efficiency and its branching ratio, rates of EES and vibrational relaxation, etc. Example of numerical simulation of nozzle flow parameters is shown on Figure 1.

It was found in the nonequilibrium supersonic nozzle flows (Mach number ~ 4) due to the high rate of recombination pumping into radiative B-state ($R^* \geq 10^{20}$ /cm³s) and rapid VT-deactivation in absorbing X-state (T_v not for much more than T), the partial population inversion (density $\Delta N \geq 10^{13}$ cm³) is formed on ET $B, v'=0, 1 \rightarrow X, v''=14-15, \dots$ in near IR spectrum ($\lambda = 0.9 \dots 1.2 \mu m$).

Experimental varying of

It should be mentioned two unsuccessful attempts to demonstrate the RGDL when a preliminary phase of laser studies was ignored [9-10].

In experiments [9] $O(B-X)$ system was wrongly selected as laser candidate, and the direct² measurements in recombining gas flow in supersonic nozzles with a high quality mirrors has not resulted on the laser effect. According with the numerical estimating, under these conditions the light gain was not more than of $10^{-6}/cm$. The point is that EES of O_2 , populating by $O+O$ atomic recombination, have strongly forbidden ET,² thus laser action could not be possible.

Other experimentators [10] failed to demonstrate the RGDL directly using the set-up of trivial heat pumped GDL. They run the experiments simply replacing the CO_2 -containing mixture by the SO_2 , NO and other dissociated gases but² did not have any laser effect.² Moreover, they have not been able to do any conclusions since they did not try to measure any characteristics of active medium in recombining gas flow. Such way without pre-laser investigations is not promising.

Besides the direct recombination laser pumping scheme it is known the other principal possibility to create a powerful visible GDL on ET using a donor-acceptor scheme based on E-E energy transfer. There are quite a number of fast chemical reactions, and atomic recombination reactions are among these, which have a high yield into metastable EES. Unfortunately, direct conversation of metastables's energy into laser radiation is difficult because its ET into ground ES is spin-forbidden and has a very small cross-section. Such situation may be a case with the products of recombination of oxygen - $O(a^1\Delta)$, nitrogen - $N(A)$. But using this metastables as a EES energy² donors, it can select suitable acceptors. The criteria for choice of pairs are ES excitation efficiency and radiative property of lasants. Here the halogens and halogens are good lasing also due to partial inversion arising between the low energy levels of the radiative EES and the high energy levels of the ground ES.

In conclusion, one should be stressed that all molecular candidates selected for RGDLs has been approbated in the lasers with optical pumping. If to consider this laser operation process as complex of three sequential stages: - 1) high laser levels pumping, 2) stimulated radiative ET and 3) low laser levels depopulation - experimentally two last ones has been successfully demonstrated, and V-T relaxation proved to be able "over-work" the molecular flow rate in CW lasing that comparable to analogous one for conditions of recombination pumping. This fact inspires our optimism.

4. -General Features and Perspective.

Expected RGDL performance

RGDL on ET will be substantially differs from usual GDLs on VRT by following advantageous properties:

- more short wavelength of lasing ($\lambda=0,4-1,2 \mu m$);
- wide tuning of laser spectrum (10-25%);
- high stored energy per $1 cm^2$ of cross-section of a gas flow input to the optical cavity (up to $5-20 kW/cm^2$);
- high laser efficiency (5...15%).

For example, in the gas flow at cross-section $5 \times 50 cm^2$ it can be extracted the laser output up to 50...250 kW.

RGDL intracavity active media parameters will differ from GDLs too:

- more high pressure (200...600 Torr);
- more high temperature (500...800 K);
- the length of inversion zone $\sim 10...50 cm$.

Supposed RGDL applications

Because of its unique possibilities, the RGDL systems will have potential for all known high power laser applications in science, engineering and technology.

In fact, for powerful laser technology applications, the CO₂ and YAG lasers are most widely used now: the users of CO₂ lasers take advantage of its high power output capability, and the users of YAG lasers focus on its high fiber transmission capability. The RGDLs will possess both important advantages simultaneously.

While the power levels of RGDL is superior even to that of CO₂ lasers in the same conditions, the oscillation wavelength lies near the minimum loss range of silica fibers. Therefore it is feasible to transmit and deliver CW laser power through an optical fiber network system for a multi-station production combined with a robot and X-Y material processing.

Decrease in diffraction divergence of RGDL's beam associated with the shorter wavelength results in the better focusability ($\sim \lambda^2$) which allows to increase the energy concentration on the target as high as 100...400 times more than the CO lasers of the same power level. Combining this effect with the lesser reflectivity and greater absorptivity of metals ($\sim \lambda^{-3/2}$), the RGDL may be expected to give more high quality of processing.

For laser energy production and wireless transmission applications the RGDL would operate in autonomous regime as they will not use an electric energy sources. Wide-range tunability of laser oscillation will open new possibilities both for loss-free communication and photochemical technology (laser-induced catalysis, isotope separation, etc.). Environment control will be possible as well.

At last, due to favour wavelength operation and high energy storage characteristics, an efficient repetitively-pulsed (up to 10⁴ Hz) generator-amplifier can be expected to construct as alternative laser driver system for fusion reactor research.

In spite of these fascinating properties nobody has tried to developed the RGDL for industrial application in mind probably because of some difficulties mentioned above. It motivated us to take part in this challenge because it will be a discovery of new generation of CW high power short wavelength lasers and will have a significant impact in a number of key technologies.

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