Quantum Electronics **45** (8) 704–708 (2015)

PACS numbers: 42.55.Lt; 42.55.Xi; 42.60.Lh
DOI: 10.1070/QE2015v045n08ABEH015750

Optically pumped rare-gas lasers

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Abstract. **The modern state of the research of a new promising optically pumped laser system with an active medium formed by metastable rare-gas atoms is briefly reviewed. The kinetics of these media is similar to that of laser media based on alkali metal vapour; however, the gas medium is inert. Metastable atoms can be produced in an electric discharge. As in alkali lasers, the specific laser power output under atmospheric pressure can be several hundreds of watts per 1 cm^3. The lasing wavelengths lie in the near-IR range and fall in the transparency window of the terrestrial atmosphere. This new concept makes it possible to develop a closed-cycle cw laser with megawatt power levels and high beam quality.**

Keywords: *optically pumped laser, rare gases, metastable atoms.*

1. Introduction

Currently, principles of designing new high-power lasers, which would possess high efficiency, scalability, and diffraction beam quality, have been actively sought for. The main direction is to sum and convert radiation of diode lasers using various solid and (recently) gas media. High-power and efficient diode lasers, as well as linear and composite arrays on their basis, have been developed in the last decade; however, the beam quality of these systems is below diffraction-limited quality. Diode laser radiation can be used for efficient optical pumping of laser media in order to provide diffraction beam quality. Gas media can ensure high beam quality in the cw regime with megawatt power levels; it has been demonstrated by an example of chemical oxygen–iodine laser. These levels of laser power may deteriorate the optical homogeneity and damage solid media.

Recently, significant progress has been made in the development of a gas laser of this kind: a diode-pumped alkali laser (DPAL). Development of a caesium vapour laser with closed-cycle circulation of the active medium was reported in [1]. The laser power was ~1 kW at an active medium volume of 12 cm^3. The pump conversion efficiency (ratio of the lasing power to the optical pump power) reached 48%. Currently, the maximum value of the differential pump conversion efficiency for DPALs is ~70% [2]. Despite the encouraging results obtained in this field, some technical problems, related to the high reactivity of alkali metals, were revealed. For example, high-power pump radiation in combination with the presence of hydrocarbon molecules in the medium, which provide fast excitation transfer to the upper laser level, lead to destruction of laser cell windows [2]. A necessary condition for efficient operation in a medium free of hydrocarbons is helium pressure from several atmospheres to several hundreds of atmospheres, depending on the alkali metal chosen. A detailed description of an alkali laser can be found in reviews [2–4].

Another DPAL problem was discovered by Zhdanov et al. [5]; it is illustrated by Fig. 1, taken from [5]. This figure shows that, under conditions of pulse pumping, the output power linearly increases with increasing pump power, whereas under continuous pumping, the output power reaches a maximum and then even diminishes with a further increase in the pump power. Simulation with allowance for the heat release and various kinetic and radiative processes does not make it possible to describe adequately these experiments [6]; however, it was found that the situation can be significantly improved using active-medium circulation [1].

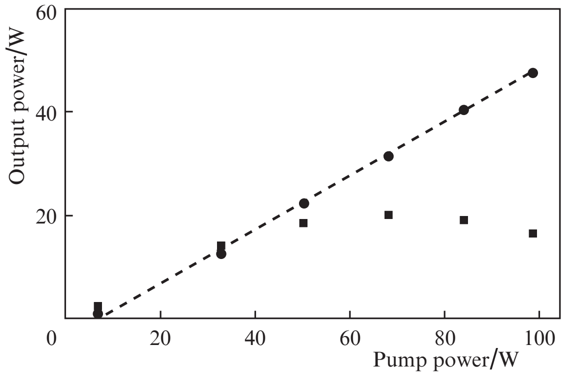

Figure 1. Dependence of the DPAL output power on the pump power for (●) pulsed and (■) continuous pumping [9].

Recently, M. Heaven (Emory University, USA) proposed and experimentally implemented a new scheme of an optically pumped laser with an active medium based on metastable rare-gas atoms, produced in a pulsed gas discharge [7, 8]. With regard to the energy level diagram of emitting atoms and the laser kinetics, this scheme is similar to that of an alkali laser. This similarity is due to the fact that both an alkali metal atom and a metastable rare-gas atom have one electron per outer shell. This feature makes it possible to implement an

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Received 22 January 2015; revision received 3 April 2015
Kvantovaya Elektronika **45** (8) 704–708 (2015)
Translated by Yu.P. Sin'kov

optically pumped rare-gas laser (OPRGL), as in the case of a DPAL. The OPRGL wavelengths, being near the boundary between the IR and visible spectral ranges, fall in the atmospheric transparency window. The upper laser level is efficiently populated as a result of collisions of excited heavy rare-gas atoms with atoms of lighter rare gases, for example, He, Ne and Ar. This is an essential difference from the DPAL medium, where a necessary condition for fast collisional relaxation from the pump level to the upper laser level is the presence of hydrocarbon molecules. According to the data in the literature [9–11], the constants of energy transfer collisional processes between the states of rare-gas atoms involved in a lasing cycle are $\sim 10^{-11}$ cm^3 s^{-1}; thus, one can obtain, as in DPAL, laser power of several hundreds of watts per 1 cm^3 of the active medium under atmospheric pressure and at a number density of metastable atoms on the order of $10^{12}-10^{13}$ cm^{-3}.

The relative simplicity of discharge scaling in mixtures of rare gases makes it possible to develop a megawatt cw laser with an active-medium on the order of several litres or several tens of litres. Modulating the pump intensity, one can implement pulsed and repetitively pulsed regimes. In addition, the OPRGL active medium contains only rare gases; this is an important advantage over DPAL, because the chemical interaction of alkali metals with the materials of the medium and cell is absent in this case.

2. OPRGL lasing cycle

The OPRGL operation involves excited s and p states of Ne, Ar, Kr and Xe atoms. The lower laser level is the metastable state of a rare-gas atom with the lowest energy (except for He), $(n+1)$s$[3/2]_2$, which can easily be populated in an electric discharge. The OPRGL lasing cycle includes optical excitation of the $(n+1)$s$[3/2]_2 \rightarrow (n+1)p[5/2]_3$ transition, collisional relaxation to the $(n+1)$p$[1/2]_1$ level and lasing at the $(n+1)$p$[1/2]_1 \rightarrow (n+1)s[3/2]_2$ transition. This three-level scheme is illustrated in Fig. 2 by an example of the argon atom. For simplicity, the Paschen notation is often used for the s and p states of rare-gas atoms (except for He). In descending order of energy, the four s states and ten p states are denoted, respectively, as $1s_2 - 1s_5$ and $2p_1 - 2p_{10}$.

Figure 2 shows also the $1s_4$ level, which is not directly involved in the lasing cycle.

However, the experiments performed at Heaven's laboratory [8, 11] showed that the occupation of this level due to spontaneous emission from the upper laser level $2p_{10}$ with a wavelength of 965.8 nm may negatively affect the lasing at low temperatures and pressures, when the relaxation from the $1s_4$ level to the $1s_5$ level occurs insufficiently rapidly. Direct and reverse collisional transitions between the $1s_4$ and $1s_5$ levels are shown, respectively, by solid and dotted slanted arrows. Optical pumping is performed at 811.5 nm, and the lasing wavelength is 912.3 nm. The vertical dashed arrows show spontaneous transitions (here, the subscript p corresponds to the pumping level and the subscripts u and l correspond to the upper and lower laser levels, respectively).

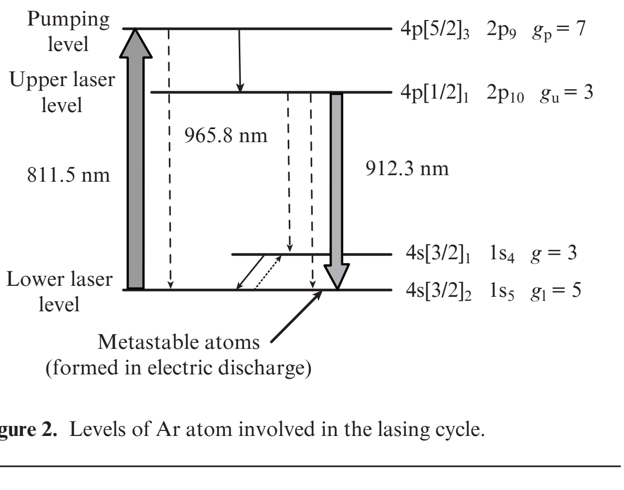

Figure 2. Levels of Ar atom involved in the lasing cycle.

3. Current state of the OPRGL study

3.1. Experiment

To date, the operating capacity of OPRGL has been demonstrated in both pulsed [7, 8] and cw [12] regimes. All experiments were performed using longitudinal optical pumping (coaxial with the output laser beam). In the first experiments [7], metastable Ne*, Ar*, Kr* and Xe* (RG*) atoms were produced using a nanosecond pulsed discharge in the discharge chamber of an excimer laser (Lambda Physik EMG 102) in an RG: He mixture or in pure argon. The RG partial pressure in helium was about 30 mbar, whereas the total pressure of the mixture was varied in the range of 200–2000 mbar. Optical pumping was performed using a tunable optical parametric oscillator with an emission line width of 30 GHz. The gain of the medium was measured using a pulsed tunable dye laser. The optical pumping and lasing (in air) wavelengths were, respectively, 640.2 and 703.2 nm for Ne, 811.5 and 912.3 nm for Ar, and 881.9 and 980.2 nm for Xe. In contrast to Ne, A, and Xe, pumping of the Kr : He mixture was performed at different wavelengths: 769.5 or 811.3 nm, and amplification was detected at 829.8 and 892.9 nm, respectively; these data indicate that different schemes of energy levels can be used to implement the OPRGL operation. In these first experiments, a pump conversion efficiency of ~13% was obtained under far-from-optimal conditions.

In the experiments performed in [8], metastable argon atoms were produced using a pulsed microsecond discharge in Ar : He $\approx$ 1 : 10 mixtures under pressures of about 360 Torr. A discharge was ignited between flat stainless steel electrodes 2.5×2.5 cm in size (the interelectrode distance was 0.5 cm). Optical pumping was performed by a cw semiconductor laser with a power up to 8 W and spectral width less than 10 GHz. It was found that the lasing pulse duration is much shorter than the discharge duration. It was also noted in [8] that spontaneous emission at a wavelength of 965.8 nm leads to occupation of the $1s_4$ level. A detailed study of the collisional relaxation kinetics [11] showed that, under the gas mixture pressures that could be attained in the discharge system described in [8], the relaxation from the $1s_4$ level to the $1s_5$ level occurs insufficiently rapidly. As a result, the lower laser level becomes depleted, with the corresponding deterioration of lasing parameters.

The parameters of a laser medium based on Ar : He mixtures depend on the energy exchange rates between the p and s states of Ar atoms. Table 1 contains the energy exchange rate constants k_{ij} (measured in [11]) for argon p levels in helium and argon (in parentheses) as collisional partners. The initial and final levels are indicated in the first column and the first row, respectively. The columns denoted by symbols q and T contain, respectively, the constants for the transition to s levels and the constants for the total depopulation rate of the corresponding level. These results show that the transitions between the argon p and s multiplets under collisions with helium atoms occur more slowly in comparison with the transition between the pumping level p_9 and the upper laser level p_{10}. The fraction of the power transmitted beyond the laser channel is small under these conditions; hence, an efficient laser can be developed based on an Ar : He mixture. In addition, since $k_{89} = 3k_{98}$ for the p_8 and p_9 levels, the energy exchange between these levels can be neglected in estimations.

Table 1. Energy exchange rate constants for Ar p levels (in Ar–He mixtures) in 10^{-11} cm^3 s^{-1}.

Levels	p_8	p_9	p_{10}	q	T
p_8		4.5(1.1)	0.4(1.1)	<0.1(1.5)	4.9(3.7)
p_9	1.5(0.4)		1.6(2.6)	(2.5)	(5.5)
p_{10}					<0.005(0.6)

The relaxation rate constant from the $1s_4$ level to the $1s_5$ level for an Ar : He mixture was also measured for the first time in [11]; this constant was found to be $ks_{45} = (1 \pm 0.3)\times10^{-13}$ cm^3 s^{-1} at room temperature. The rate constant for the reverse process, measured for the first time in [13], turned out to be $(2.1 \pm 0.2)\times10^{-15}$ cm^3 s^{-1}. These values are in good correspondence within the principle of detailed balance. In addition, ks_{45} was found to depend on temperature: it increased to 2.8×10^{-13} cm^3 s^{-1} with increasing temperature to 397 K.

The first laser experiments under atmospheric pressure in a continuous discharge were performed in [12]. A nonequilibrium plasma was formed using a linear array of high-frequency microdischarges, developed at Taft University [14]. The array of discharges operated at a frequency of 900 MHz; the sizes of the discharge plasma region were $0.3\times0.9\times19$ mm, with discharge power of 9 W. Dividing formally power by volume, we obtain the discharge power density to be 560 W cm^{-3}. Rawlins et al. [12] used a tunable diode laser to measure the number densities of metastable Ar($1s_5$) atoms in a 2 % Ar : He mixture, which turned out to be $(3-5)\times10^{12}$ cm^{-3}; this number density level is quite sufficient for lasing experiments.

Optical pumping was performed using a cw tunable Ti : sapphire laser with a power of 1 W and a lasing linewidth less than 2 GHz. This spectral width of pump radiation is obviously smaller than the width of the Ar absorption line under atmospheric pressure and at a temperature of 600 K, which was measured by the authors in a discharge plasma using a tunable diode laser over the 912.3-nm absorption line. The focusing optics provided an averaged pump intensity up to 10 kW cm^{-2}.

At pump intensities of 1.3–4 kW cm^{-2}, the small-signal gain in the centre of the gain line was measured to be 1 cm^{-1}. Gain saturation was observed at pump intensities of 1–2 kW cm^{-2}. CW lasing was obtained at a wavelength of 912.3 nm, with a power of 22 mW; the power absorbed at 811.5 nm was 40 mW. Thus, the OPRGL optical efficiency in these experiments turned out to be 55 %. The values of optical efficiency and gain are in good agreement with theoretical estimates [15]. However, gain saturation occurred at pump intensities an order of magnitude higher than those obtained in [15] and the estimates of the authors [12]. These estimates showed also that satisfactory agreement with the experimental gain and [Ar*] number density can be obtained on the assumption of exponential increase of constants ks_{45} and $k_{\rm pl}$ with heating.

3.2. Theoretical estimates

The efficiency and optimal parameters of the laser medium for the cw regime of OPRGL operation was estimated theoretically for the first time in [15] by an example of the Ar : He mixture, for which the known set of constants of collisional processes in a discharge plasma turned out to be the most complete. The analytical model took into account the radiative and collisional losses of p states; the collisional transfer from the optically pumped $2p_9$ state to the emitting $2p_{10}$ state; and the loss of the lower metastable $1s_5$ state on the formation of excimers and transfer to the higher lying $1s_4$ state, which is radiatively coupled to the ground state (with allowance for the radiation trapping). The excitation of the lower metastable state by an electric discharge, optical pumping and stimulated emission were also taken into account. The energetic efficiency of the formation of metastable atoms in a discharge was calculated by solving the Boltzmann equation. Variable parameters were introduced to take into account the influence of the processes with unknown constants. Estimations were performed with reasonable suggestions about unknown parameters, by analogy with the known processes in other mixtures of rare gases. As a result, it was found that the kinetics of collisional processes at pressures close to atmospheric and an Ar content of ~1 % in the mixture may provide specific laser energy output of several hundreds of watts per 1 cm^3 of the medium at number densities of metastable atoms on the order of $10^{12}-10^{13}$ cm^{-3}.

The structure of the lower energy levels of the Ar s multiplet includes two metastable levels (with radiative lifetimes of the $1s_3$ and $1s_5$ levels of ~1 and ~50 s, respectively) and two emitting levels (the radiative lifetimes of $1s_4$ and $1s_2$ are, respectively, ~10 and ~2 ns). The energies of s levels differ by ~0.1 eV. Fast energy exchange between these levels occurs in a plasma discharge (mainly because of the collisions with neutral particles), and the effective lifetime of metastable levels is much shorter than the radiative lifetime. However, at a high number density of RG atoms in the ground state, which are necessary for the OPRGL medium, and a characteristic size of the medium on the order of 1 cm, the radiation is trapped. The effective radiative lifetime of metastable levels is fairly long (about 1–2 μs) [16, 17]. Another source of loss for RG* is the quenching by polyatomic molecules with gas-kinetic rates, which imposes certain requirements to the purity of gases used and the discharge system design [18].

Under atmospheric pressure, the collisional rate of pump energy transfer to the upper laser level exceeds the effective loss rate for the lower laser level by three orders of magnitude; correspondingly, each metastable atom is involved in lasing cycle several thousands times during its lifetime. Therefore, the discharge power necessary to provide a sufficiently high [Ar*] number density is low in comparison with the optical pumping power; it was calculated to be 10–20 W cm^{-3} [15]. The pump conversion efficiency, with allowance for the collisional and radiative loss kinetics, is approximately up to 70 %, while the total efficiency is few percent lower. The small-signal gain was calculated to be several units of cm^{-1} at an optical pump power density of several hundreds of W cm^{-2}. Based on the consideration of possible loss mechanisms for the p and s states and estimation of the Ar* production efficiency in a discharge, it was concluded in [15] that the OPRGL gas mixture should contain few percent of Ar in He or, more generally, heavy RG in a light RG.

Despite the fact that the active medium is in the state of plasma, the influence of charged particles on the kinetic processes important for laser operation can be disregarded. The rate constants of the processes with participation of charged particles are 10^{-7} cm^3 s^{-1}, while the rate constants of energy exchange with participation of neutral particles are 10^{-11} cm^3 s^{-1}. The electron number density in a glow discharge, which is expected to provide a necessary number of metastable atoms, is ~10^{-12} cm^{-3}, and the number density of RG atoms that is necessary for efficient OPRGL operation is on the order of 10^{19} cm^{-3}. Therefore, the frequwncies of the processes with participation of neutral particles and charged particles are, respectively, on the order of 10^8 and 10^5 s^{-1}. Only under the conditions of a high-frequency atmospheric-pressure microdischarge, where, as was stated in [12], the electron number density is $10^{13}-10^{14}$ cm^{-3}, charged particles may significantly affect the laser kinetics.

The following relation for estimating the OPRGL optical efficiency was obtained in [15]:

$$\eta_{\mathrm{opt}} = \frac{W_{\mathrm{out}}}{W_{\mathrm{p}}} = \eta_{\mathrm{q}} \frac{1 - Y(1 + I_n^{-1})}{1 + \Pi}, \qquad (1)$$

where W_{out} is the output laser power, which is determined by the kinetics of the medium; W_{p} is the absorbed optical pump power; $\eta_{\mathrm{q}} \approx 0.9$ is the quantum efficiency (ratio of the pump and lasing wavelengths); and I_n is the ratio of the pump intensity to the saturation intensity with respect to pumping. Parameters Π and Y characterise the fraction of the power transmitted beyond the laser channel; they are determined as

$$\Pi = \frac{\nu_{\mathrm{pl}}}{\nu_{\mathrm{pu}}} = \frac{A_{\mathrm{pl}}}{4.4\times10^8\,\Xi} + \frac{k_{\mathrm{pl}}^{\mathrm{Ar}}}{k_{\mathrm{pu}}} y_{\mathrm{Ar}} + \frac{k_{\mathrm{pl}}^{\mathrm{He}}}{k_{\mathrm{pu}}}(1 - y_{\mathrm{Ar}})$$

$$\approx 7.5\times10^{-2}\,\Xi^{-1} + 1.4 y_{\mathrm{Ar}}, \qquad (2)$$

$$Y = \frac{g_{\mathrm{u}}}{g_{\mathrm{p}}}\frac{\nu_{\mathrm{pl}}}{\nu_{\mathrm{pu}}} = \frac{g_{\mathrm{u}}}{g_{\mathrm{p}}}\left[\frac{A_{\mathrm{ul}}}{4.4\times10^8\,\Xi} + \frac{k_{\mathrm{pl}}^{\mathrm{Ar}}}{k_{\mathrm{pu}}} y_{\mathrm{Ar}} + \frac{k_{\mathrm{pl}}^{\mathrm{He}}}{k_{\mathrm{pu}}}(1 - y_{\mathrm{Ar}})\right]$$

$$\approx \frac{3}{7}(5.4\times10^{-2}\,\Xi^{-1} + 1.1 y_{\mathrm{Ar}}). \qquad (3)$$

These parameters are the ratios of frequencies (in the kinetic sense) of transitions from the pumping level ν_{pl} and the upper laser ν_{ul} to the lower laser level to the transition frequency from the pumping level to the upper laser level ν_{pu}. Here, k_{ij} are the rate constants for the corresponding transitions; A_{ij} are the Einstein coefficients; $\Xi = N/(2.46\times10^{19})$ is the number of particles per 1 cm^3, normalised to the number of particles at a pressure of 760 Torr and a temperature of 300 K; and y_{Ar} is the argon fraction in the mixture. The numerical coefficients in the expressions for Π and Y were calculated with allowance for the measurements performed in [11], which revealed the absence of intermultiplet p–s transitions at collisions of Ar atoms with He atoms ($k_{\mathrm{pl}} = k_{\mathrm{ul}} = 0$); correspondingly, the third terms in the expressions for Π and Y are absent. Under the experimental conditions implemented in [1] (pressure of 769 Torr and temperature of 600 K), the Ξ value is 0.5; therefore, the Π and Y values are determined mainly by the spontaneous emission from the pumping and upper laser levels. The analysis performed in [1] showed that the 1s$_4$ level relaxes fairly rapidly to the lower 1s$_5$ level, presumably, because of the exponential increase in the corresponding rate constant with temperature; therefore, the occupation of the 1s$_4$ level can be neglected. Under experimental conditions ($I_n \approx 1$), we find that the calculated optical efficiency is ~70%. This value is in good correspondence with the result of [12] (55%), with allowance for the fact that the cavity efficiency is below unity and that the spatial distribution of pump intensity is nonuniform.

Thus, the experimental results of [12] (values of optical efficiency and gain) are in good agreement with the theoretical estimates [15]. The difference in the pump saturation intensities by an order of magnitude may be related to the spatial distribution of pump radiation in the experiment; however, this problem calls for further study.

4. Problems to solve

An analysis of the data in the literature showed that the temperature dependences of the collisional relaxation constants of RG levels, which are especially important for estimating the OPRGL parameters at high plasma-discharge temperatures, are completely unknown. There are hardly any reliable measured values of the number density of heavy RG* in a discharge plasma under conditions of strong dilution with a lighter RG, when the loss on excimer formation is small (a condition preferred for forming an active medium). These data are of key importance for justified estimation of the prospects of developing a high-efficiency cw OPRGL.

To estimate the pump radiation absorption length and the plasma temperature from the shape of the RG* absorption line, it is necessary to know collisional-broadening coefficients for the corresponding spectral lines. Some reliable experimental data on the collisional broadening of the 811.5-nm Ar line in an Ar–He mixture have been obtained only recently [19, 20]. The broadening coefficients (HWHM) at 300 K are as follows: $\xi_{\mathrm{Ar-Ar}} = (2.85 \pm 0.1)\times10^{-10}$ s^{-1} cm^3 and $\xi_{\mathrm{Ar-He}} = (3.3 \pm 0.1)\times10^{-10}$ s^{-1} cm^3. These data are absent for mixtures of Ar, Kr, and Xe with Ne and for mixtures of these RGs.

An important problem in implementing the cw regime of OPRGL operation is to provide sufficiently high RG* number densities in the continuous-discharge plasma at elevated pressures. Stable operation of the diffuse discharge in He, Ar and mixtures of these gases under pressures on the order of atmospheric was reported in [21–23]. The number density of metastable Ar* atoms under conditions of a dc microdischarge was measured in [21] to be 10^{14} cm^{-3}; this value is an order of magnitude larger than the minimum value necessary for OPRGL operation (according to theoretical estimates). However, it is not yet clear how to implement a high-pressure dc discharge in a volume of about 1 L. Sufficiently high [Ar*] number densities were produced in [12]; however, the formally obtained discharge power density exceeds the theoretical estimates by an order of magnitude. One might suggest that this discrepancy is due to the following factors ignored in experiment: power dissipation in the design elements of microdischarge array, quenching of Ar* on sputtered electrode materials, or the existence of discharge structures similar to near-electrode layers. It is also obvious that necessary [Ar*] number densities cannot be produced in large volumes under microdischarge conditions. Thus, the problem of an appropriate discharge system still remains to be solved.

The width of atomic lines of both rare gases and alkali metals due to the collisional broadening under pressures on the order of atmospheric is known to be ~10 GHz (0.03 nm); thus, to implement efficient radiation conversion, one should take necessary measures to narrow the pump spectrum. Currently, narrow-band pump sources, in particular, for pumping DPALs, have been actively developed. For example, Gourevitch et al. [24] used an external cavity with a volume Bragg mirror (OptiGrate) to narrow the spectrum of a 30-W laser-diode linear array to 10 GHz with minimum (~10%) power loss in comparison with the case of free lasing. A 250-W laser module with a spectral width less than 10 GHz for pumping rubidium vapour, developed based on this technology, was described in [25].

Thus, the laser medium with metastable RG atoms is of great practical interest and must be comprehensively investigated. The first results in this field give grounds for certain conclusions. In particular, the results of measuring the collisional-relaxation constants in an Ne:He mixture [26] suggest that an OPRGL with a high optical efficiency cannot be based on Ne because of the presence of intermultiplet p–s transitions. On the contrary, the absence of intermultiplet transitions in Ar^* upon collisions with He makes it possible to design an efficient laser system using widespread and inexpensive rare gases. Krypton- and xenon-containing mixtures have not been investigated sufficiently thoroughly to date.

Acknowledgements. The part of the work carried out at the Lebedev Physics Institute (Samara Branch) was supported by the Russian Foundation for Basic Research (Grant No. 14-05-97013), and the work performed at the Samara State Aerospace University (SGAU) was supported by the Ministry of Education and Science of the Russian Federation within the Program for Improving the Competitiveness of SGAU for 2013–2020 and State Contract for Research at Institutions of Higher Education (No. 3.161.2014/K).

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