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Electron-beam-excited high-pressure He–Ar mixture as a potential active medium for an optically pumped laser

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***Abstract.* The possibility of developing a laser with optical pumping of metastable ArI levels produced by a relativistic electron beam in high-pressure He–Ar gas mixtures is investigated. It is shown that a high population of the $1s_5$ level of the argon atom, a wide (~10 nm) absorption spectral line, and a great lifetime allow us to obtain, as a result of electron beam impact, optimal lasing conditions within the framework of the classical three-level laser scheme. The rate constants of two- and three-particle collisional relaxation processes of low-lying 4s levels of an excited argon atom in He–Ar mixtures in the pressure range from 4 to 0.75 atm are measured.**

Keywords: *electron-beam laser, laser on atomic transitions of inert gases, diode pumping.*

1. Introduction

In the last 15 years, diode-pumped alkali lasers (DPALs), which provide high-power single-aperture lasing with a high optical beam quality, have been significantly developed [1, 2]. At the same time, the high chemical aggressiveness of such a laser medium leads to a rapid damage of optical elements of the laser cell, and also to the formation of laser-scattering hydrides (laser snow) in reactions with hydrocarbons, which are used as a spin-orbital relaxation agent in dense mixtures at atmospheric pressure [3]. A transition to the scheme with the use of metastable states of atoms of inert gases formed by an electric discharge of direct current (DC) or by a radio-frequency (RF) discharge in diode-pumped rare gas lasers (DPRGLs) at a pressure below 1 atm made it possible to obtain a chemically inert active medium with preservation of the same high-level specific laser characteristics [3–6]. A He–Ar mixture with the use of pumping of the $4s[3/2]_2^o$ metastable level ($1s_5$ in Paschen notation) of an excited argon atom to the optically coupled $4p[5/2]_3$ level by means of radiation from a diode laser with a wavelength of 811.5311 nm corresponding to the $4p[5/2]_3 - 4s[3/2]_2^o$ transition proved to be the most promising for this type of lasers (Fig.1). Rapid collisional relaxation leads to excitation of the $4p[1/2]_1$ ArI level, forming a population inversion that results in lasing at the $4p[1/2]_1 - 4s[3/2]_2^o$ transition with a wavelength of 912.2967 nm. Thus, a classical three-level laser model is implemented. As is known, this model requires high levels of optical pumping and population of the $4s[3/2]_2^o$ metastable level, which is estimated by a value of no less than 10^{13} cm^{-3} (see work [7]).

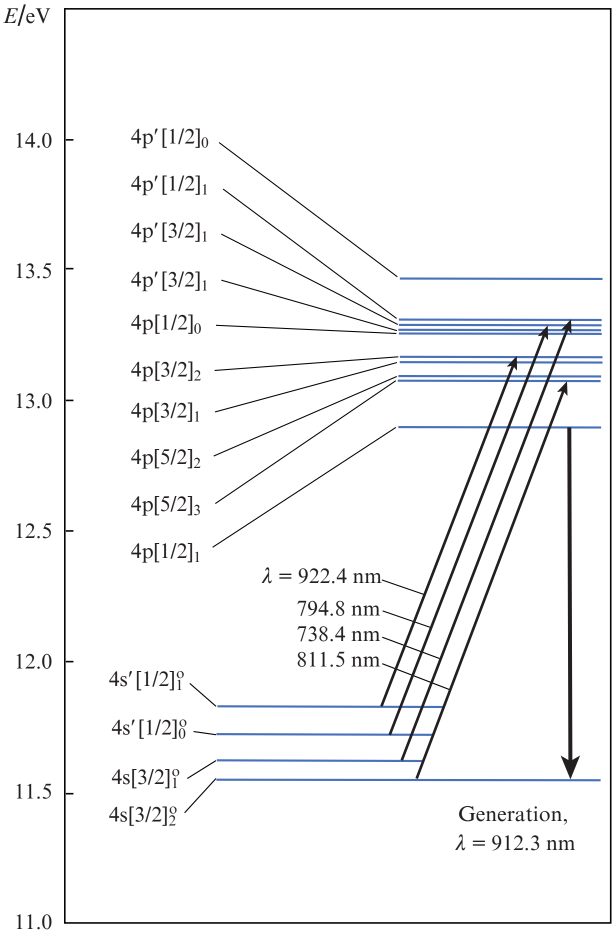

Figure 1. Diagram of low-lying excited ArI levels.

There are a number of fundamental limitations that reduce the possibility of constructing high-power DPRGLs, associated with the electric discharge physics in dense gaseous media. These problems can be solved by using a quasi-relativistic electron beam to pump the laser gas mixture. The possibility of scaling the active volume to tens or even hundreds of litres and increasing the gas mixture pressure to several atmospheres with preservation of the spatial homogeneity of elec-

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tron pumping makes the electron beam very promising for the excitation of lasers of this type. At the same time, it has been found that a number of plasma-chemical processes occurring in the dense medium of a He–Ar laser under the action of a high-energy electron beam are not sufficiently studied. First and foremost, it is necessary to refine the rate constants of the processes responsible for populating of the lower s levels of the argon atom. In work [8], we have obtained the rate constants for two- and three-particle processes relevant to the $4s[3/2]_2^o$ metastable level; nevertheless, relevant constants for the other levels are either absent or require refinement. It is also worth noting that many authors (see, for example, Refs [3–7, 9]), with the aim of construction of various models of DPRGLs on the He–Ar mixture, use data from work [10], in which, when studying VUV radiation in the afterglow of a high-voltage electric discharge in pure argon at a low pressure (~25 Torr), the rate constants for two- and three-particle collisional processes at room temperature were obtained, which constituted $(13.8 \pm 1.4)\times10^{-16}$ cm^3 s^{-1} and $(1.71 \pm 0.02)\times10^{-32}$ cm^6 s^{-1}. At the same time, work [10] presents summary data on these constants, obtained with the use of absorption monitoring methods and also of other methods of excitation of the studied gases, and these data differ significantly from the above values. The same work emphasises that the approximations used to determine the rate constants are not valid for high gas pressures (more than 100 Torr).

The processes of collisional relaxation with simultaneous participation of atoms of working (argon) and buffer (helium) gases cause special difficulties in modelling, since experimental data on the rate constants of such processes are absent in the literature, and authors commonly use a calculated estimate corresponding to half the constant for pure argon [5]. However, simplest calculations show that for such values of constants, in the case of the He : Ar = 50 : 1 mixture at a pressure of 4 atm, the lifetime of the $4s[3/2]_2^o$ metastable level does not exceed 0.5 μs, and this clearly contradicts a long (several microseconds) exponential tail of the absorption pulse at a wavelength of 912.2967 nm [8].

Another significant dissimilarity from the DPRGL of the proposed laser scheme employing the electron-beam excitation of the He–Ar mixture consists in the formation of inversion and implementation of lasing in the afterglow that follows the electron pulse impact, since a high density of secondary hot electrons formed under pumping by a high-energy electron beam results in efficient mixing of the excited p and s levels of the argon atom, which worsens the conditions for inversion formation during the pumping, while a great lifetime of the metastable level, which significantly exceeds the lifetime of secondary electrons [8], makes it possible to implement, under the condition of optical pumping at a wavelength of 811.5311 nm, population inversion in the afterglow at these laser levels, when the electron beam action stops. The aim of this work is to evaluate the possibility of using dense He–Ar mixtures excited by a relativistic electron beam as an active medium for diode-pumped lasers.

2. Experiment

The experiments were conducted on the 'Tandem' pulsed laser electro-ionisation installation. The gas mixture under study was pumped by a ribbon electron beam with a cross section of 100×5 cm with an electron energy of 150 keV at the entrance to the active volume, a current density of 1.5 A cm^{-2}, and a total pulse duration of ~3.5 μs. Earlier in work [8], we described in more detail the installation parameters and the methodology of experiments. To determine the rate constants of two- and three-particle collision processes

$$\mathrm{Ar(4s) + He + Ar \rightarrow Ar_2^* + He}, \tag{1}$$

$$\mathrm{Ar(4s) + 2He \rightarrow HeAr^* + He}, \tag{2}$$

$$\mathrm{Ar(4s) + He \rightarrow Products + He}, \tag{3}$$

which are responsible for depopulating the $1s_4$, $1s_3$, and $1s_2$ levels of the argon atom in dense He–Ar mixtures at a total pressure of several atmospheres after the electron beam exposure, a method of absorption probing was used to study the time dependences of the absorption coefficients for 4p–4s optical transitions associated with the levels in question. Data on these transitions and the corresponding wavelengths are given in Table 1 and Fig. 1. The transmittance of excited gas mixtures was conducted by an ISI-1 pulsed broadband light source, while the required optical line was selected by means of an MDR-2 high-aperture monochromator. With the use of high-sensitive noise-protected semiconductor photodetectors with a time resolution of at least 100 ns and a DSO-2102 dual-channel digital oscilloscope, simultaneous recording of radiation at the input and output of the active volume pumped by an electron beam was performed.

Table 1. The ArI levels under study and corresponding optical transitions and wavelengths.

Level	Transition of ArI	Wavelength/nm
$1s_4$	$4p'[3/2]_2 - 4s[3/2]_1^o$	738.4
$1s_3$	$4p'[3/2]_1 - 4s'[1/2]_0^o$	794.8
$1s_2$	$4p[3/2]_2 - 4s'[1/2]_1^o$	922.4

As in our previous work, the absorption pulses (at the wavelengths from Table 1) for He–Ar mixtures in the pressure range from 4.0 to 0.75 atm with a ratio of components 50 : 1, 100 : 1, and 200 : 1 were characterised by long exponential tails of microsecond duration. Numerical processing by means of the least squares method of an array of experimental data at different pressures and gas component ratios for each of the 4s levels in question allows us to calculate the rate constants for processes (1)–(3) in accordance with a procedure detailed in work [8]. As a result, the values of these constants given in the generalised Table 2 were obtained for all low-lying 4s ArI levels.

The data in Table 2 show that all low-lying 4s ArI levels in the studied pressure range of He–Ar mixtures and ratios between components are long-lived. In this case, with an increase in the energy level, a slight increase in the rate constants of collisional relaxation for all processes (1)–(3) is observed. Thus, we may conclude that lasing on optical transitions terminating at any of the low-lying 4s ArI levels is possible either in the self-limited regime, or in the presence of additional channels to depopulate this level.

The significantly lower rate constant for process (2) compared to that for process (1) indicates a minor role of process (2) in depopulating the lower 4s ArI levels in mixtures with a high working gas content, which is typical of He–Ar laser mixtures. However, this process can play a very important role associated with collisional broadening of the $4p[5/2]_3$ –

Table 2. Rate constants of the processes of collisional quenching for 4s ArI levels.

Reaction	Rate constants for metastable levels		Rate constants for resonant levels	
	$4s[3/2]^{\rm o}_2$	$4s'[1/2]^{\rm o}_0$	$4s[3/2]^{\rm o}_1$	$4s'[1/2]^{\rm o}_1$
(1)	$(3.6 \pm 0.4) \times 10^{-33}$ cm^6 s^{-1} [8]	$(4.9 \pm 0.5) \times 10^{-33}$ cm^6 s^{-1}	$(4.3 \pm 0.9) \times 10^{-33}$ cm^6 s^{-1}	$(7.0 \pm 0.7) \times 10^{-33}$ cm^6 s^{-1}
(2)	$(4.4 \pm 0.9) \times 10^{-36}$ cm^6 s^{-1} [8]	$(5.6 \pm 1.0) \times 10^{-36}$ cm^6 s^{-1}	$(5.4 \pm 1.1) \times 10^{-36}$ cm^6 s^{-1}	$< 1.0 \times 10^{-35}$ cm^6 s^{-1}
(3)	$(2.4 \pm 0.3) \times 10^{-15}$ cm^3 s^{-1} [8]	$(3.9 \pm 0.4) \times 10^{-15}$ cm^3 s^{-1}	$(3.5 \pm 0.7) \times 10^{-15}$ cm^3 s^{-1}	$(6.0 \pm 0.6) \times 10^{-15}$ cm^3 s^{-1}

$4s[3/2]^{\rm o}_2$ optical transition at a wavelength of 811.5311 nm. Indeed, it can be assumed that, in our absorption experiments, we observe, as process (2), the sum of two processes: direct process of three-particle collisional relaxation of the lower metastable 4s level with the formation of a heteronuclear HeAr* excimer with a very low ($\sim$0.01 eV) binding energy that can be estimated on the basis of Table 1 of work [9] [this process is comparable in speed to the process (1)], and an equally rapid reverse process. As a result, the spectral shock broadening of this level depends quadratically on pressure and significantly exceeds the standard estimates of the broadening by pressure. To verify this, we measured the absorption coefficient over the entire length of the active volume near the wavelengths of the transitions studied, and also near the wavelength of the $4p[5/2]_3 - 4s[3/2]^{\rm o}_2$ transition, and obtained the dependences shown in Fig. 2. These dependences were constructed for a He:Ar = 100:1 mixture at a pressure of 3.0 atm; in this case, the absorption maximum was observed immediately after the termination of electron beam pulse.

In addition, we measured the instrumental function of a monochromator using a He–Ne laser. In the same optical scheme and at the same dimensions of the input and output slits, and at identical filling by radiation of diffraction gratings (600 lines mm^{-1}) of the MDR-2 monochromator, the measured half-width of the He–Ne laser spectrum, that is, the instrumental function, was 1.3 nm. The half-widths of the spectral lines ending at $4s[3/2]^{\rm o}_1$, $4s'[1/2]^{\rm o}_0$ and $4s'[1/2]^{\rm o}_1$ levels were quite close to this value, although they slightly exceeded it, whereas for the 811.5311 nm line and, accordingly, the $4s[3/2]^{\rm o}_2$ level, the corresponding half-width turned out to be almost an order of magnitude larger. A similar pattern was observed in our work [8] for the 912.2967 nm line ending at the same lower metastable level. The only possible reason for this difference in linewidth may be that, under our experimental conditions, after the impact of electron beam, the $4s[3/2]^{\rm o}_2$ level is the main populated excited ArI level with a population density of $10^{13} - 10^{14}$ cm^{-3}, and its population significantly exceeds the population of other levels. Accordingly, the direct process (2) and the corresponding inverse process develop

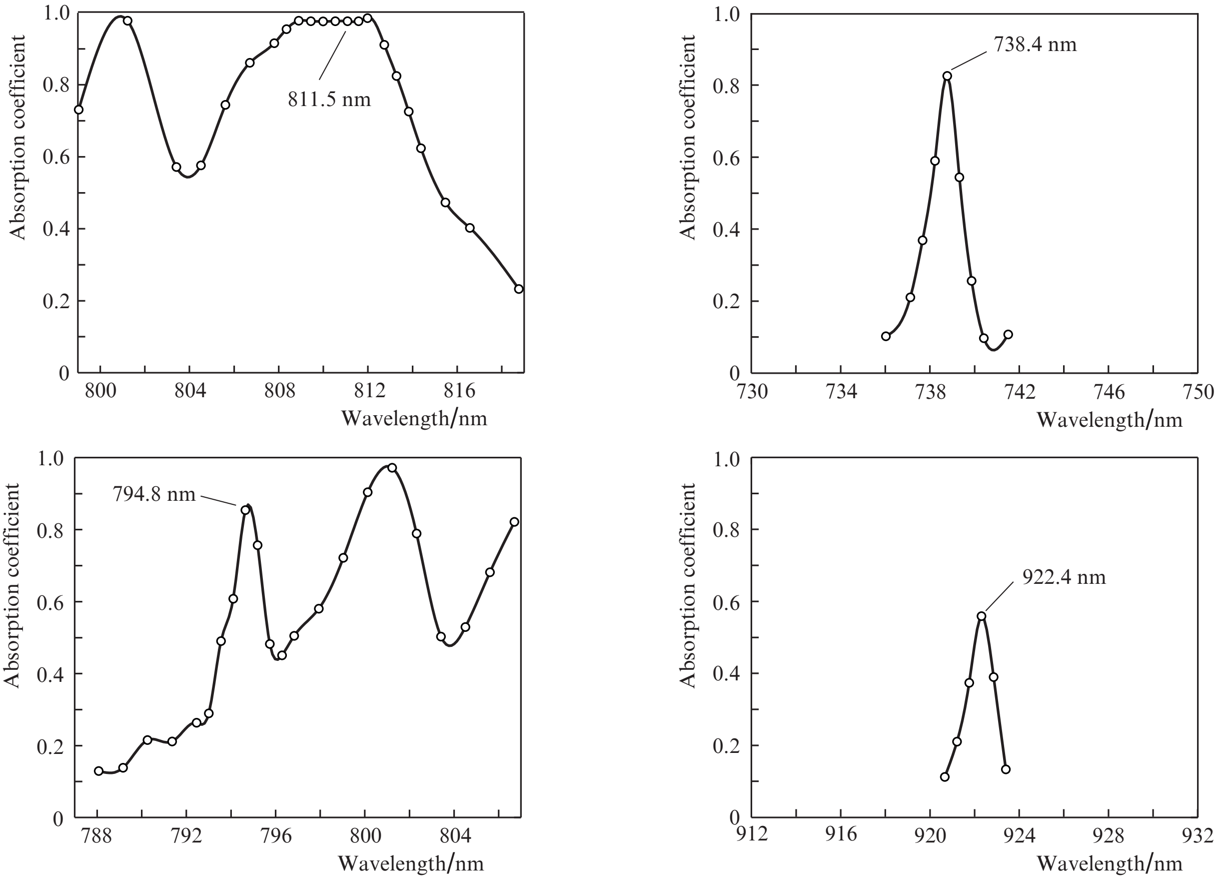

Figure 2. Spectral dependences of the absorption coefficient α near spectral lines under study with wavelengths of 811.5, 738.4, 794.8, and 922.4 nm.

most intensively at this level, which leads to its additional spectral broadening.

The wide ArI absorption line at a wavelength of 811.5311 nm overlaps the lasing spectrum of serial high-power semiconductor lasers with a radiation wavelength in the range of 808 – 810 nm, which allows them to be used for efficient optical pumping of a He – Ar laser. To verify this assumption, we used a five-millimetre array of ten cw laser diodes with a total power of 20 W and a lasing spectrum width of ~2 nm at half width (FWHM). By selecting the coolant temperature in the cooling system of this array, it was possible to adjust the lasing wavelength for the $4p[5/2]_3$ – $4s[3/2]^{\circ}_2$ transition. The radiation from this array was focused into a spot of 3×1 mm at the centre of active volume, which, according to the estimates based on the data from work [9], provided the radiation power density at focus at the level of 670 W cm^{-2} (FWHM), which is below the lasing threshold for DPRGLs. In addition, in order to simplify spectral measurements, an optical scheme was chosen with transverse pumping of the active He–Ar medium by radiation from a diode laser. Since the region of high power density of optical pumping in this case was small compared to the volume of our measuring camera (its active length was 100 cm, and the aperture was 5 cm), a new measuring camera with an active length reduced to 1 cm and a light aperture of 1.5 cm was manufactured to reliably record the impact of optical pumping on spectral characteristics of an electron-beam-excited high-pressure He–Ar mixture (Fig. 3). The camera was located opposite the electron beam output window in the 'Tandem' accelerator, and, before entering the camera active volume, the beam passed through two layers of 25 μm thick titanium foil and a 1 cm thick layer of air. Radiation from an array of diode lasers was injected through a window located at the camera bottom transverse to the camera optical axis. At one end of the chamber, there was installed a highly reflecting flat mirror, and at the other end there was either a glass substrate with an antireflecting coating at 912 nm, or a semi-transparent mirror, forming in this case a 50% plane-parallel cavity.

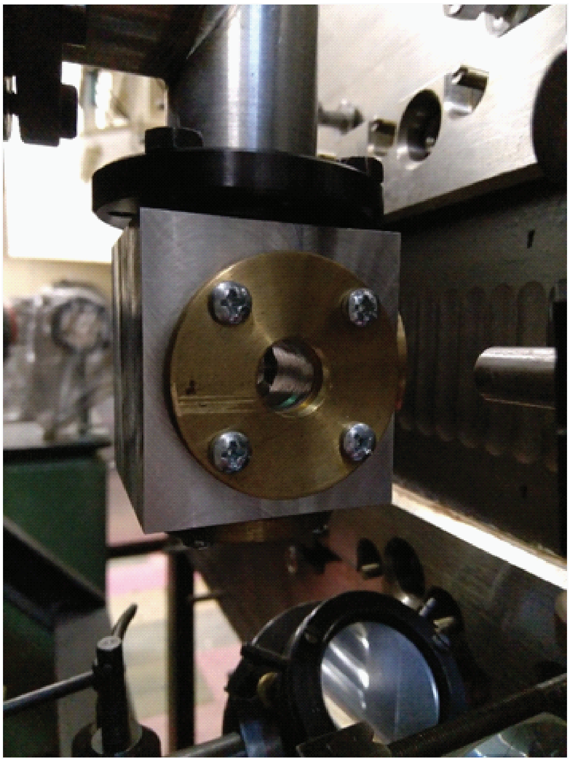

Figure 3. Camera for studying photoluminescence of high-pressure He–Ar mixtures in electron beam afterglow pumped by radiation from an array of diode lasers.

Using a six-meter bundle of silica fibre with a diameter of 5 mm, photoluminescence radiation from an electron-beam-excited active He–Ar mixture was transmitted to the entrance slit of the MDR-2 monochromator tuned to a wavelength of 912.3 nm and then recorded by a silicon photodiode and a digital oscilloscope on the monochromator output slit, as we did it previously in our absorption measurements. Photoluminescence was observed under pumping by an electron beam, both with and without cw optical pumping.

The most pronounced were the changes in the photoluminescence pulse for a mixture with a fairly high working gas content (He : Ar = 50 : 1) at a gas pressure of 4 atm, which was maximal under our experimental conditions (Fig. 4). The launch of optical pumping resulted in a significant increase in the amplitude and duration of photoluminescence, which can be explained by an increase in the $4p[1/2]_1$ level population as a result of rapid collisional relaxation of the $4p[5/2]_3$ level pumped by radiation from a diode laser with simultaneous termination of parasitic mixing of the 4p and 4s ArI levels by secondary electrons in the gaseous He–Ar mixture excited by

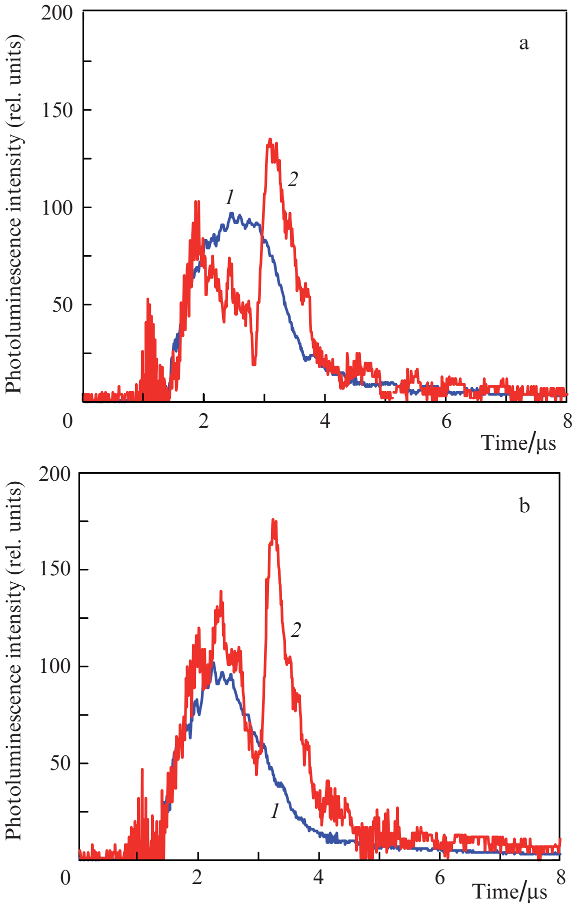

Figure 4. Photoluminescence pulse at a wavelength of 912.3 nm: (a) without a resonator (with a single highly reflecting mirror); (b) with a semi-transparent plane resonator [(*1*) electron beam pumping only, (*2*) combined electron-optical pumping]. The pump electron pulse duration in the camera active volume is ~2 μs.

a relativistic electron beam. The use of a semi-transparent plane resonator significantly increased the radiation intensity of the active medium in simultaneous electron-beam and optical pumping, which indicates the observation of ASE. Too small length of active medium and insufficient density of optical pumping did not allow us to obtain lasing in our experiments.

At the same time, our experiments demonstrated the possibility of effective optical excitation of the $4p[1/2]_1$ ArI level by radiation from a standard array of laser diodes without the use of special methods of narrowing and fine-tuning into the optical transition of the generation line of these diodes, which holds out a hope of the use of high-power industrial assemblies of kilowatt-power and more high-power diode lasers, widely used at present for pumping of solid-state lasers, in the studied electron-beam pumped high-pressure lasers.

3. Conclusions

Our research on the possibility of using relativistic electron-beam-excited high-pressure He–Ar mixtures as an active medium for optically pumped lasers yielded the following results:

(i) It is shown that all low-lying 4s ArI levels are long-lived (the lifetime varies from several to tens of microseconds) in the studied pressure range with a ratio of the buffer and working gas components from 50:1 to 200:1. The previously unknown rate constants for two - and three-particle processes (1)–(3) of collisional relaxation have been determined.

(ii) For gas mixtures under study, an abnormally wide radiation absorption line (~10 nm at half width) near the 811.5311 nm line corresponding to the $4p[5/2]_3 - 4s[3/2]_2^o$ ArI transition was found. This opens up the possibility of efficient optical pumping of the studied active media by radiation from broadband high-power commercial laser diode arrays.

(iii) Experimentally, by varying the photoluminescence signal at a wavelength of 912.3 nm, optical pumping of an electron-beam-excited high-pressure He–Ar mixture has been demonstrated using a laser diode array with a lasing wavelength of 811.5 nm. It is shown that the most optimal photoluminescence and amplification conditions for such a laser mixture at a wavelength of 912.3 nm are realised after the electron beam impact. An increase in the region length and optical pumping density will make it possible to attain lasing at a wavelength of 912.3 nm in a laser utilising a high-pressure He–Ar mixture with electron-beam optical pumping.

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References

1. Krupke W.F. *Proc. SPIE*, **7005**, 700575 (2008).
2. Bogachev A.V., Garanin S.G., Dudov A.M., Eroshenko V.A., Kulikov S.M., Mikaelyan G.T., Panarin V.A., Pautov V.O., Rus A.V., Sukharev S.A. *Quantum Electron.*, **42** (2), 95 (2012) [*Kvantovaya Elektron.*, **42** (2), 95 (2012)].
3. Han J., Glebov L., Venus G., Heaven M. *Opt. Lett.*, **38** (24), 5458 (2013).
4. Han J., Heaven M. *Opt. Lett.*, **37** (11), 2157 (2012).
5. Demyanov A.V., Kochetov I.V., Mikheyev P.A. *J. Phys. D: Appl. Phys.*, **46** (37), 375202 (2013).
6. Demyanov A.V., Kochetov I.V., Mikheyev P.A., Azyazov V.N., Heaven M.C. *J. Phys. D: Appl. Phys.,* **51** (4), 045201 (2018).
7. Yang Z., Yu G., Wang H., Lu Q., Xu X. *Opt. Express*, **23** (11), 13823 (2015).
8. Ionin A.A., Kholin I.V., L'dov A.Yu., Seleznev L.V., Ustinovskii N.N., Zayarnyi D.A. *Laser Phys.*, **27** (12), 125803 (2017).
9. Emmons D.J., Weeks D.E. *J. Appl. Phys.*, **121** (20), 203301 (2017).
10. Wieme W., Lenaerts J. *J. Chem. Phys.*, **74** (1), 483 (1981).