Chapter 7

Nuclear-Induced Plasmas of Gas Mixtures and Nuclear-Pumped Lasers

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Additional information is available at the end of the chapter

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Abstract

We briefly describe the basic processes of formation and relaxation of nuclear-induced plasmas of gas mixtures, especially the processes of inverse population creation in nuclear-pumped lasers (NPL). A review of the work to create and research nuclear-pumped lasers is in progress: on transitions of atoms and atomic ions and on molecular transitions. An increased focus is on the gas media, which we also study on WWR-K nuclear reactor and DC-60 ion accelerator. The studies on emission of heteronuclear ionic molecules of inert gases are also reviewed.

Keywords: laser, ionizing pumping, mechanism of population, recombination, direct excitation

1. Introduction

Direct conversion of nuclear energy into light energy is of great interest as it provides for application of compact and energy-intensive nuclear energy sources to create high-power generators of coherent and incoherent optical radiation. Nuclear energy pumping into active laser medium was first proposed with the appearance of first lasers [1, 2]. At present, the research on nuclear-pumped lasers (NPL) has progressed to the stage where design and engineering developments of continuous and pulse nuclear laser equipment for various purposes have become possible, that is integrated units based on nuclear engineering and physics, quantum electronics, physics of low-temperature plasma, optics, gas dynamics, and other areas of science and technology [3].

Nuclear-pumped lasers have potential in a wide range of applications, especially in cases requiring high-power and compact lasers to be placed on autonomous remote facilities. The most promising areas of nuclear-pumped laser application are as follows: laser thermonuclear fusion, long-distance transmission of radiant energy and information, rocket laser engine, laser isotope separation and photochemistry, stratospheric ozone layer recovery, and space junk removal. Considerable interest in this area research is also associated with significant difference between the mechanisms of level population during nuclear pumping and population processes in conventional gas-discharge lasers. Application of nuclear energy for active laser medium pumping can be considered not as the way to create high-power laser, but as the way to obtain energy from nuclear reactor. This necessitates consideration of fundamentally new equipment—a reactor laser designed to spatially combine nuclear laser active medium and nuclear reactor core. This approach opens up opportunity to generate qualitatively new energy.

Attempts to achieve laser action during pumping of condensed media with nuclear radiation did not yield positive results. The main obstacle on the road of creating condensed media NPLs is their radiation damage: radiation defects of crystal lattice in solid-state laser, radiolysis, and gas bulb generation on the tracks of nuclear particles in liquid lasers. Presently known gas NPLs [3] radiate in spectral range 391–5600 nm in about 50 atomic transitions of Xe, Ar, Kr, Ne, C, N, Cl, O, I, Hg; Cd^+, Zn^+, Hg^+ ions, CO molecules, and N_2^+ molecular ion.

2. Methods and sources of gas excitation by nuclear reaction products

NPLs include active media that are excited directly using nuclear radiation, or with the use of intermediate nuclear-optical converters. There are three basic sources of nuclear radiation, which can be used to pump NPLs or convert nuclear energy into light energy on transitions of atoms and molecules:

1. nuclear explosions,
2. radioactive isotopes,
3. neutron radiation of nuclear reactors.

Using the γ-ray radiation from nuclear explosion as the pump source was apparently done for the first time in VNIIEF (All-Union Scientific Research Institute of Experimental Physics) in 1971 [3]. Xenon emitted as Xe_2^* excimer molecules was used as an active media. Experiments on xenon excimer laser pumping (λ ~ 170 nm) were done in a testing area in Nevada in 1973 [4]. Experiments to develop NPLs using nuclear explosive devices were carried out up until 1987 when underground test ban was introduced.

Optical radiation of gases excited by radioactive nucleus decay products (^{210}Po, ^{238}Pu, ^{239}Pu, ^{241}Am, etc.) was studied before to create gas scintillators [5, 6]. Radioactive isotopes usage for laser pumping [7] or pre-ionization in electric discharge lasers [8, 9] is limited by lower power density deposited in gas (up to 0.6 W/cm^3). The main volume of works on NPL active media search and study of their parameters was performed on stationary and pulsed nuclear reactors. Nuclear reactors are the source of neutron and γ-radiation. Neutrons were used to pump lasers, as in this case the energy input to laser medium is several times higher than due to γ-radiation. Direct pumping of active media is usually carried out not by neutron radiation but by nuclear reaction products with thermal neutrons (**Table 1**).

Reaction (energy of reaction, MeV)	Natural composition of isotopes	Kinetic energy of reaction products, MeV	Cross-section of reaction for thermal neutrons, barns
$^{3}He(n,p)T$, (0,76)	$^{4}He(100\%)+^{3}He(1,4°10^{-4}\%)$	p—0,57; T—0,19	5400
$^{10}B(n,\alpha)^{7}Li$, (2,35)	^{11}B (80,1)% +^{10}B (19,9%)	^{4}He—1,5; ^{7}Li—0,85	3837
$^{6}Li(n,T)^{4}He$ (4,7)	^{6}Li (7,5%)+^{7}Li (92,5%)	T—2,7; ^{4}He—2,0	945
$^{235}U(n,ff)FF$ (167)	$^{238}U(99,28\%)+^{235}U(0,72\%)$	Fragments: light—99; heavy—68	582

Table 1. Nuclear reactions [10] for NPL pumping.

When nuclear reactors are used as neutron sources, two basic types of laser-medium excitation are utilized:

1. a gaseous isotope or compound thereof (^{3}He, $^{235}UF_6$, $^{10}BF_3$) is a component part of the laser medium,

2. internal surface of the gas-filled laser cell is coated with a thin layer of isotope (^{10}B, ^{6}Li, ^{235}U) or compound thereof ($^{235}UO_2$, $^{235}U_3O_8$).

In the case of volumetric source of pumping using ^{3}He, the non-uniformity of pumping comes from the absorption of slow neutrons in ^{3}He and from the reduction of energy contribution in area near the wall, owing to removal of reaction products to the walls of the cell, in case of $^{235}UF_6$—fission fragments energy loss on the walls of the cell. Results of computation of the total energy deposition and spatial distribution of the deposited energy depending on ^{3}He pressure and diameter of cylindrical cell are given in [11, 12], while [13] shows the results of computation for $^{235}UF_6$-He at different pressure of mixture and content. In Ref. [14], authors show summary of results of experimental and theoretical studies dedicated to definition of energy contribution in NPL cells. Three experimental methods were considered: pressure shock method, interferometric method, and string calorimeter method. The cell size and path length of nuclear reaction products in the gas mixture determines spatial non-uniformity at surface pumping source use. Various calculation models for spatial distribution of energy deposition, influence of non-uniformity of uranium-containing layers, and analysis of experimental data on determination of uranium fission fragments energy loss in gas medium are given in [3].

Due to high ^{3}He and ^{10}B neutron-absorption cross-section, loading laser devices using ^{3}He or ^{10}B on the walls can significantly affect reactivity charge of nuclear reactor and even lead it to subcritical state. A laser cell containing ^{235}U also serves as a fuel element of reactor. This has triggered an idea of laser reactor, which must spatially combine active laser medium and nuclear reactor core [2, 15]. Initially considered option included uranium-235 hexafluoride serving as uranium-containing medium, the only uranium compound existing in gas phase at moderate temperatures. However, the use of $^{235}UF_6$ complicated due to the chemical aggressiveness of uranium hexafluoride and products of radiolysis. Laser radiation absorption by UF_6 molecules, high speed of quenching of excited atoms and molecules in collisions with UF_6, electron attachment to molecules of UF_6 also prevents the use of uranium hexafluoride as a component of the laser mixture [16]. At present, the most realistic designs are heterogeneous reactor lasers using thin-film uranium fuel [17, 18]. The core of this reactor laser is a specific quantity of laser cells with uranium layers appropriately placed in a neutron moderator matrix. With appropriate selection of components, the conditions for the reactor-laser operation are provided without utilizing additional fuel (uranium). The number of laser cells may vary from a few 100 to 1000, the total weight of uranium from 5 to 70 kg, and characteristic linear dimensions are 2–5 m [3, 18].

Studies in recent years were set out to explore opportunity to load uranium in active laser mixture in the form of fine dust with a particle size of 100–500 nm substantially lower than the path length of fragments [19]. In this case, it is possible to minimize energy loss in the fuel and substantially improve the efficiency and uniformity of pumping. It is necessary to ensure the transparency of such a mixture at the lasing wavelength. Theoretically, it is possible by arranging the dust particles in the form of periodic structure with a mutual distance comparable to laser wavelength, that is, in the form of dust crystals.

2.1. Basic processes of formation and relaxation of nuclear-induced plasmas of gas mixtures

Currently, direct nuclear pumping is implemented in gas media in which the populating of lasing levels occurs in a low-temperature plasma formed by ionizing radiation, in nuclear-induced plasmas. This section describes the basic processes of formation and relaxation of such plasma, in relation to active media of lasers with nuclear pumping and conversion of nuclear energy into the energy of spontaneous gas emission. The most complete information about the processes in plasma of active media of gas lasers with nuclear pumping is contained in monographs [3, 20].

Initial stage of ionization processes in gas media. Gas medium ionization occurs by various types of radiation: uranium fission fragments and transuranium elements, fast electrons, protons and tritons, lithium nuclei, α-particles, γ-quanta. Ionization in γ-radiation of gas is induced by fast electrons produced in the process of Compton scattering, photoeffect, and effect of electron-positron pair formation. In the initial stage of ionization process, primary ionization during the immediate interaction of charged particles and secondary ionization in interaction of media atoms with electrons formed as a result of primary ionization.

The process of ionization of an atom may be viewed as a binary collision of oncoming charged particle and one of the electrons of the atom's shell [21]. Due to the large difference in masses of heavy charged particles and the electron, only a comparatively small percentage of fragment energy can be transferred to the orbital electron. The spectrum of electrons produced by ionization of heavy particles is softer compared with the spectrum produced by ionization of gas by fast electrons [22, 23]. The average energy of electrons formed in neon as a result of ionization by fission fragments is 40 eV and fast electrons 150 eV [22]. In the case of fission fragments, the secondary electron may provide additional one or two acts of ionization on average, while in the case of fast electrons, it is from 5 to 10 [23].

However, differences in the effects on gas media by different types of ionizing particles are not substantial, because the ultimate result is a combined effect of primary and secondary ionization. It follows from calculations in that the electron energy distribution and energy formation of electron-ion pair in the gas does not depend on the type of charged particles [24, 25]. The same conclusion can be drawn from the luminescent properties of plasma and gas NPLs output parameters, which do not depend on the type of charged particles, but depend on the power and duration of pumping [18].

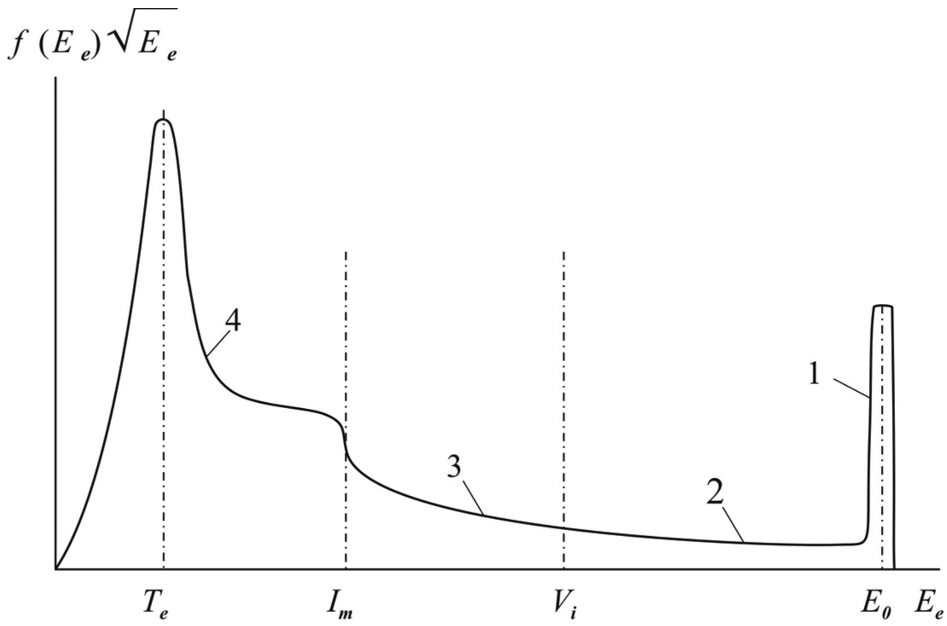

Figure 1. Electron energy distribution in the ionized gas. 1—primary electrons of source; 2—electrons of ionization cascade; 3—electrons in the inelastic excitation region; 4—thermal and subthreshold electrons.

Usually, the calculation and analysis of parameters of nuclear-induced plasmas, development of laser action and NPLs radiation, ionization of gas medium are assumed to be homogeneous. One of the features of nuclear-induced plasma is associated with the formation of tracks when passing through dense gas of heavy charged particles [26, 27]. Depending on parameters of gas media, transverse dimensions of the tracks are 1–10 μm, and the track lifetime or the time to establish uniform ionization through diffusion is 0.1–1 μs [28]. Non-uniformity of ionization associated with track structure of plasma will be most noticeable in the following cases:

- at ionization by fission fragments and other heavy ions,
- in dense gases with high atomic weight,
- at a very low degree of ionization, when there is no overlapping of tracks.

Fluctuations of plasma component concentrations induced by the track structure may have some effect on NPL characteristics excited by fission fragments, if population of upper levels is due to the fast charge process, for example, in laser on a mixture of He-Cd [28]. The influence of plasma track structure on recombination processes will be insignificant, as the track lifetime is much less than the characteristic time of recombination processes [29, 30]. Tracks' overlapping occurs at high pumping power densities; therefore, the track structure of plasma disappears. Estimates show that the overlap of tracks in atmospheric pressure helium occurs at excitation power densities ~2 W/cm^3 [29].

Ionization of gas at the initial stage is carried out directly by charged particles and secondary electrons. A picture of the electron energy distribution in the gas is shown in **Figure 1**, where f_e is the energy distribution function of the electrons, and E_e is the electron energy.

The entire electron energy range can be divided into three regions [24]:

1. The region of ionization cascade $V_i < E_e < E_0$ (E_0 is the particle initial energy, V_i is the ionization potential of the gas atoms and molecules), in which the electrons energy is sufficient for ionization of gas particles.
2. In elastic excitation region $I_m < E_e < V_i$ (I_m is the minimal threshold of electron or vibrational excitations), in which the energy of the electrons is reduced, primarily due to excitation of electron and vibrational states of the gas particles.
3. In the subthreshold region ($E_e < I_m$), electrons lose energy in small "portions" due to elastic collisions with gas particles, thus creating electron thermalization. In subthreshold region, the electrons are involved in the processes that are important in population kinetics and NPL levels' deactivation. These processes include the following: the electron-ion recombination, quenching of excited states, processes of attachment to electronegative gas, excitation, and ionization in collisions with gas particles in excited states.

At $E_e > I_m$, the function of electron energy distribution differs greatly from Maxwell distribution. Electrons in this region do not participate in recombination processes and this region supplies electrons in the subthreshold region. The electrons of inelastic region excitation and ionization cascade possibly play a major role in population of 3p levels of neon [30, 31].

2.2. Kinetics of plasma processes at nuclear pumping of gas mixtures

In quantum system, the gain (absorption) factor of the medium is described by [32]:

$$\alpha = \sigma(N_2 - N_1 \frac{g_2}{g_1}) \tag{1}$$

where indexes (1, 2) refer to upper level 2 and lower 1, N—level population, g—statistical weight of levels. The cross section of stimulated transition:

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$$\sigma = \frac{\lambda^2}{2\pi}\frac{A}{\Delta\omega} \tag{2}$$

where λ—transition wavelength, $\Delta\omega$—line width, A—transition probability. The amplifying medium ($\alpha > 0$) requires maintaining population inversion: Population of the upper level must exceed population of the lower level (adjusted to degeneracy multiplicity). Formation of population inversion requires selectivity of population of the upper or lower level. The inversion can be provided not only by the predominant population of upper laser level, but also through selective cleaning of the lower level.

The active media of gas NPLs are often the double mixtures A-B (A—a buffer gas with a high ionization and excitation potential, B—a gas with a lower ionization potential, and lasing occurs in its transitions) or triple A-B-C. In triple mixtures, the third C gas usually plays the role of deactivator of lower level and is not involved in upper level population, but can quench it to some extent. Therefore, we consider the kinetics of processes in plasma in the example a two-component mixture.

The first stage includes ionization and excitation of the buffer gas atoms A (formation of A^+ ions and excited atoms A*), in some cases, direct excitation of active gas B [31, 33]. The main channels of energy transfer from A^+ and A* to particles B are as follows:

1. Charge exchange processes

$$A^+ + B \rightarrow (B^+)^* + A \tag{3}$$

$$A_2^{\ +} + B \rightarrow (B^+)^* + 2A \tag{4}$$

2. Penning process (if A* energy is higher than B ionization potential)

$$A^* + B \rightarrow (B^+)^* + A + e \tag{5}$$

3. Excitation transfer

$$A^* + B \rightarrow B^* + A \tag{6}$$

The main type of ions in high-pressure plasma is molecular ions A_2^+, B_2^+, $(AB)^+$ which are formed in triple processes:

$$A^+ + A + M \rightarrow A_2^{\ +} + M \tag{7}$$

$$B^+ + B + M \rightarrow B_2^{\ +} + M \tag{8}$$

$$A^{+} + B + M \rightarrow (AB)^{+} + M \tag{9}$$

where M—third particle (A or B). Plasma neutralization occurs as a result of recombination processes, which, depending on specific conditions, may prevail or have dissociative recombination.

$$A_2^{+} + e \rightarrow A^* + A \tag{10}$$

$$B_2^{+} + e \rightarrow B^* + B \tag{11}$$

$$(AB)^{+} + e \rightarrow B^* + A \tag{12}$$

triple or shock-radiative recombination

$$A^{+}(B^{+}) + 2e \rightarrow A^*(B^*) + e \tag{13}$$

$$A^{+}(B^{+}) + e + M \rightarrow A^*(B^*) + M \tag{14}$$

Population of laser levels occurs during processes (3–5) for B^+ atomic ions, (6, 11–14) for B neutral particles, as well as in cascade transitions from B^{**} high levels. It was previously considered [34, 35] that the processes of dissociative recombination of molecular ions are predominantly populated by p-states of atoms, but in recent years this conclusion was questioned [3].

In [36] based on radioluminescence intensity dependence from mercury vapor pressure (10^{-3}–10^{-7} Torr) in ^{3}He-Hg mixture was made a conclusion that in the process of Hg^+ three-body recombination mostly populated d-states of mercury atoms. Thus, D-levels may also be populated in the processes of dissociative recombination of molecular ions (at a higher density of mercury atoms).

In low-pressure gas discharge laser, the lower laser level is usually deactivated in optical transitions to lower levels, and in lasers with nuclear pumping of atmospheric pressure, the deactivation occurs in collisions with media atoms or plasma electrons, and in Penning reaction with the particles of additional gas. In excimer lasers, where at photon emission the excimer molecule passes in the lower dissociated or weakly coupled state, the lower level deactivation is feasible.

Characteristics of laser radiation at pumping by hard ionizer depend on the power and duration of energy input into active medium, but do not depend on the type of ionizer [18]. This means that kinetics of processes in active media of lasers excited by an electron or ion beam and for nuclear-pumped lasers will be identical [37, 38]. Calculation of plasma param-

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eters and laser characteristics implies for kinetic models, representing the balance of rates of formation and decay of individual components in plasma. Kinetic equations are supplemented by the equations of electron energy balance. In some kinetic models, the number of plasma chemical reactions reaches several hundred (see, for example, [29, 39]). Typically, the relevant description of plasma and laser parameters' calculation suffices it to include 10–15 basic reactions. In this regard, it is sometimes advisable to use the so-called small models for calculation, which includes only the basic plasma processes, examples of such models [40, 41]. It should be noted that in many cases the basic level population process is either not defined (e.g., lasers with Xe, Kr, Ar IR transitions), or under discussion (e.g., laser on the 3p–3s transitions of neon [30, 31]). In other cases, relevant calculation is hindered by a large uncertainty in the values of (or even the order of magnitude) processes rate constants [42], the uncertainty in coefficient of light absorption by active medium particles [43].

2.3. Design and development of experimental methods for nuclear-induced plasma research

Pulsed nuclear reactors were used as a source of neutron radiation for NPLs research [3, 27, 44–47]: in Russia—VIR-1, VIR-2, TIBR-1M, BR-1, BIGR (VNIIEF), EBR-L (VNIITF, All-Russian Scientific Research Institute of Technical Physics), BARS-6 (FEI, Institute of Physics and Power Engineering), IIN-3 (Kurchatov Institute of Atomic Energy); in USA—TRIGA Mark-II (University of Illinois), SPR-III (Sandia Labs), APRF (NASA), Godiva-IV (Los Alamos National Laboratory). Relatively recent reports were issued about experimental NPL investigations in China on the CFBR-II [48]. First, NPL works in China were carried out on a stationary INPC nuclear reactor [49].

Thermal neutron flux density at stationary nuclear reactor reaches 10^{13} to 10^{14} n/cm^{2}s, and gas mixtures' pumping power does not exceed a few W/cm^3. Therefore, research on stationary reactors (IRT-2000 in Moscow Engineering Physics Institute (MEPhI), our works on WWR-K reactor) was mainly associated with the study of the spectral characteristics of plasma [50, 51], as well as the development of lasers with non-self-maintained discharge (WWR-K reactor) [52].

WWR-K reactor (**Figure 2**) is a heterogeneous unit of water-cooled type, operating on thermal neutrons. Desalinated water serves as moderator, reflector, and coolant. Uranium is used as reactor fuel enriched by uranium-235 isotope to 36%. WWR-K reactor is a powerful source of neutrons and gamma rays. The maximum thermal neutron flux in the central channel of the reactor reaches 2°10^{14} n/cm^{2}s. The reactor core is placed in an aluminum tank filled with water and is designed as hexagonal lattice containing fuel elements, control and protection system channels and experimental channels. The water temperature is kept constant and does not exceed 40°C. The core has a shape similar to cylindrical, with diameter of 645 mm and height of 600 mm. Central vertical experimental channels with diameter of 96 mm, which was used for nuclear-pumped laser works, pass through the core center. Biological protection of staff from the reactor radiation in horizontal plane is provided by layers of water of 850 mm wide, cast iron—210 mm and limonite concrete—2250 mm. Biological protection in vertical direction is formed of 3700 mm of water layer and removable cast iron lids of 800 mm wide.

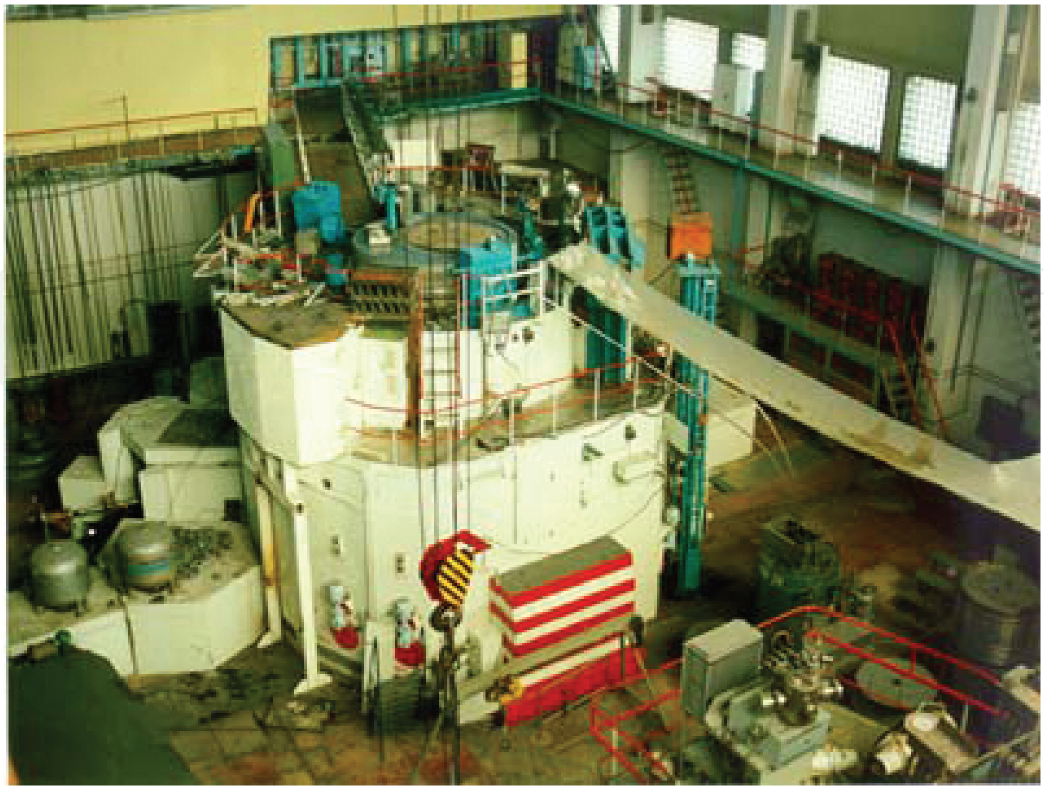

Figure 2. General view of WWR-K reactor.

The upper part of reactor tank includes rotating cast-iron lid, through which experimental channels are loaded. The left side includes the base made of plates on which vacuum pumps for pumping mixtures from laser devices under the reactor lid were placed.

Three designs of reactor core laser systems [53] were developed and tested on WWR-K reactor. One design was intended for testing mixtures of xenon laser pumped by uranium fission fragments, the second for lasers on inert gas mixtures excited by the products of $^{3}He(n,p)^{3}H$ reactions and the third to run the laser on transitions of mercury triplets [54]. **Figure 3** shows the design intended for lasers on mixtures of inert gases excited by the products of $^{3}He(n,p)T$ reactions. The laser cell is designed as electropolished pipe of 36 mm in diameter with flanges for mirrors on the edges. The distance between the mirrors is 2.1 m, and the mirrors are on a quartz substrate with a dielectric multilayer coating. Mirrors are distanced for 0.5 m (non-transmitting) and 1.0 m (half-transmitting) from the reactor core. Laser channels cutting after a 6-month settling showed that KU-1 quartz substrate has sufficiently high radiation resistance. De-gassing and gas puffing were also conducted through waveguide pipe Ø36 mm. The laser light from this tube was extracted through the window of LiF or CaF_2 and analyzed by registration system. IR radiation was simultaneously recorded by calorimeter located above the exit window and reflected on the LiF plate portion through the matrix of five PD-7G photodiodes. Radiation within the visible range was recorded by photodiodes matrix and the system for measuring the luminescence spectra based on the SPM-2 monochromator and FEU-106 photomultiplier operating in photon counting mode.

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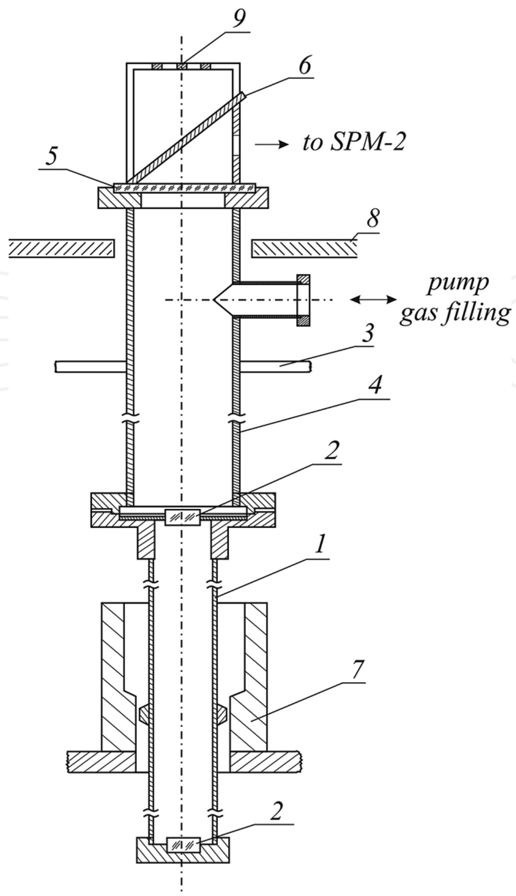

Figure 3. Scheme of laser cell of inert gases excited by $^3He(n,p)T$ reaction products. 1—laser pipe, 2—mirrors, 3—aluminum lid of reactor, 4—waveguide pipe, 5—window, 6—LiF plate, 7—propellant, 8—cast iron lid of reactor, 9—photodiodes matrix.

Although we were not able to create a continuous laser with a direct nuclear pumping, the use of continuous ionizing radiation sources: stationary nuclear reactors and radioactive sources provide for detailed study of plasma properties of gas mixtures, and stationary state of pumping simplifies the analysis of processes kinetics in plasma [53].

Currently, the research is being conducted on DC-60 heavy ion accelerator [55]. The main parameters of the accelerated ion beam: ion type—from lithium to xenon, and ion energy—from 0.5 to 1.75 MeV/nucleon. The intensity of ion beam is from 10^{12} to 10^{14} particles/s depending on type and energy of ions. Ion impulse duration is several nanoseconds and repetition rate of ions is 1.84–4.22 MHz. Mainly, the ions of argon were used as a source of ionization and excitation. The accelerated ion beam passes from evacuated transportation channel through 3-mm hole in the flange to irradiation chamber (**Figure 4**). The hole in the flange is closed by membrane of 2-μm titanium foil or 2.5-μm-thick havar foil. The gas pressure in the cell is measured by capacitance diaphragm gauge mounted at the top of the chamber. The ions passed through the foil ionize and excite the gas mixture in irradiation chamber (**Figure 5**). Argon ion energy after separation foil is about 50 MeV. The emerging light radiation passes through quartz window and condenser and focused on optical fiber. The beam falls on a compact spectrometer through the fiber, and the recorded spectrum is displayed on a computer.

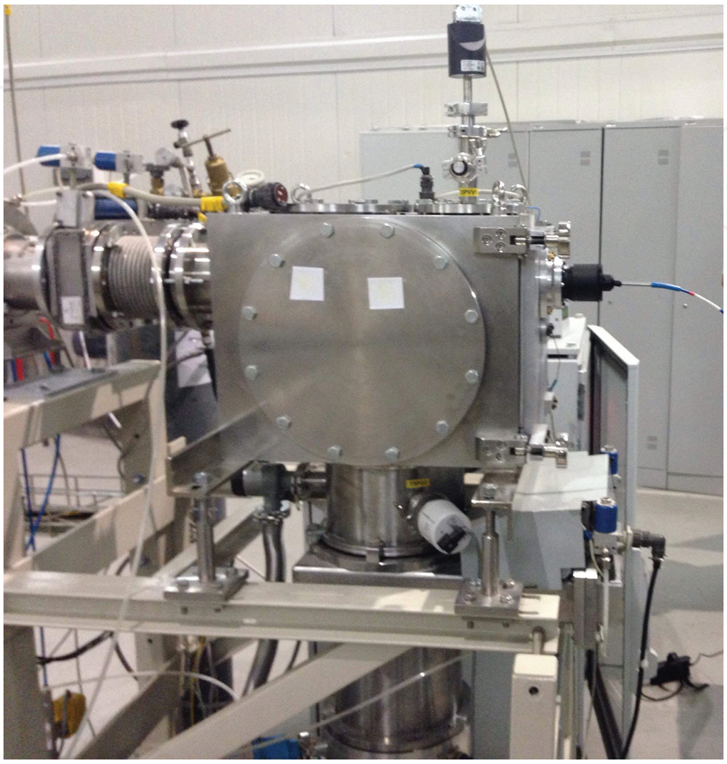

Figure 4. View of the experimental setup on the DC-60 accelerator.

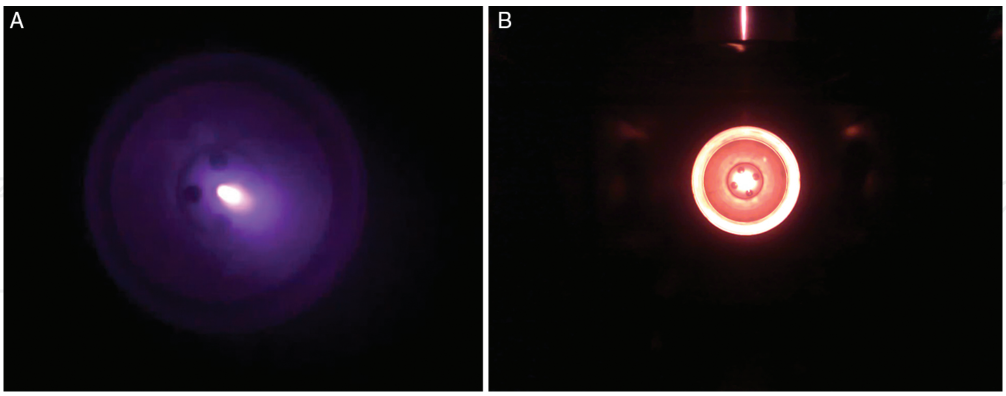

Figure 5. 400 Torr helium (a) and 600 Torr neon (b) luminescence induced by argon ions.

Beam from the narrow ion-excited region is reflected on the separating flange (a, b) and on the top of the chamber (b).

Studies on luminescence spectra of gaseous media were also conducted with the use of radioactive isotopes [36, 56–58] and pulsed nuclear reactors [59–61].

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3. NPL active media on transitions of atoms and atomic ions

3.1. IR lasers operating on transitions of Xe, Kr, and Ar

Nuclear-pumped lasers operating on IR transitions of Xe, Kr, and Ar were investigated in detail and have maximum output parameters for NPL. The research by VNIIEF in 1972 during the first experiments was performed with the VIR-2 reactor; the output power of xenon laser with an optimal pressure and composition of the He-Xe was 25 W with efficiency ~0.5%, but the results were not published that time [62]. In 1974, obtained laser action on He-Xe composition (λ = 3.51 μm) with excitation by uranium fission fragments [63]. It was one of the first publications on the achievement of lasing under direct nuclear pumping [63, 64].

Most of the lines with laser action refer to nd-(n + 1)p transitions of Xe, Kr, and Ar atoms (n = 5, 4, 3 for Xe, Kr, Ar, respectively). Xenon laser (λ = 1.7–3.5 μm) has received the most studies, as it has the highest output parameters. He, Ar, Kr, and compounds thereof served as buffer gases. The first NPL xenon laser has the maximum achieved energy parameters:

- output power 1.3 MW and energy 526 J per pulse with duration 400 ms in He-Ar-Xe compounds, with a wavelength 2.03 μm [65],
- 5.6% efficiency in Ar–Xe mixture [66], 2.5% in He-Ar-Xe [67] with λ = 1.73 μm and ~3% with 2.03 μm [65]. The differences in efficiency values may be related to differences in the evaluation of energy deposited in gas [3].

Xenon laser also has the lowest lasing threshold: $1.5°10^{12}$ n/cm^{2}s in Ar-Xe mixture (λ = 2.03 μm) [7, 68]. These advantages, as well as the absence of degradation of the laser mixture as a result of radiation and chemical reactions, join the lasers operating on IR transitions of Xe, Kr, Ar the ranks of most promising in terms of reactor-laser creation [18].

Upper location of laser levels suggests a weak temperature dependence of its output parameters. However, in the case of lasers operating on IR transitions of inert gases, the output power is halved at the temperature of the mixture 350–550 K [3, 69]. The reasons for this are still not fully understood and are the matter of discussions. The most probable reasons were considered:

- reduction in the rate of formation of heteronuclear $ArXe^+$ ions and mixing laser levels by electrons [69],
- destruction of $ArXe^+$ ions by buffer gas atoms [70],
- collisional quenching and mixing laser levels by buffer gas atoms [71],
- active medium contamination by impurities as a result of their desorption from laser cell walls as long as it is warming [72].

Although lasers operating on IR transitions of inert gases are studied for over 40 years and considered to be the most promising, there is still no clarity with mechanism of upper nd-levels population [3, 73]. The processes of deactivation of lower (n + 1)p-levels can be considered well established; it is a collisional quenching in collisions with atoms of active medium, and with electrons at high-power pumping. The main problem in determining nd-levels population mechanism is associated with complexities of IR radiation registration within the reactor experiment.

The main presently discussed mechanisms of levels population (B—Xe, Ar, Kr; A—buffer gas atom) [73] are as follows:

1. Shock-radiative recombination: B^{+}+e+e (M)→B(nd)+e(M), M— third particle.
2. Electron-ion recombination: B_2^{+}+e→B(nd)+e.
3. Recombination of heteronuclear ionic molecules: AB^{+}+e→B(nd)+A.
4. Transfer of excitation in inelastic collisions: Ar*+Xe→Xe(5d)+Ar.
5. Step excitation: Xe(6s, 6s′)+e→Xe(5d)+e.

Widely held hypothesis implies for levels population in processes (3) of dissociative recombination of heteronuclear ionic molecules with electrons [29, 74]. In works [3, 73, 75], the main channel of population is considered to be the process (2) of electron-ion recombination of B_2^{+} ions. In recombination mechanism of Xe levels' population, the lasing failure with addition of 5 Torr of uranium hexafluoride to Ar-Xe mixture [16] can be explained not only by quenching xenon levels by UF_6 molecules, and also by electron attachment to UF_6, by recombination of xenon ions with negative ions in the mixture.

According to [33, 76], dissociative electron-ion recombination of molecular ions of inert gases cannot be the main process of nd-levels population of inert gas atoms. It is expected to populate levels by direct excitation of secondary electrons from the ground state of atom, as well as transfer of excitation from buffer gas atom [33].

3.2. Visible-range lasers operating on Ne atom transitions

Atomic neon laser created in 1961 by Javan A. and others is the first laser with active gas medium. Therefore, the first proposals [1] and first experiments [2] on NPL creation were associated with well-known transitions of He-Ne laser with a wavelength of 632.8 nm and 1.15, 3.39 μm. In 1980, it was reported [77] on laser action of $5s'[1/2]_1^0$–$3p'[3/2]_2$ neon atom transition (λ = 632.8 nm, **Figure 6**) after excitation of ^{3}He–Ne mixture by products of nuclear reaction ^{3}He(n,p)T in stationary nuclear reactor. At the same time, laser efficiency was approximately 0.03% and the lasing threshold was reached at a very low thermal neutron flux $F = 2°10^{11}$ n/cm^{2}s. High gain in a mixture of ^{3}He-Ne (1.7°10^{-2} cm^{-1} at $F = 2°10^{12}$ n/cm^{2}s) was also measured in work of Chinese authors [49]. The results of these studies are questionable and subject to discussion [78, 79]. Simple estimates [78] show that in these conditions [77] population cannot be obtained even in the extreme case, when all power deposited in gas is fully transferred to the upper laser level, and the level is not quenched in collisions with atoms. In our experiments, the neutron flux density was gradually changed from 10^{11} to 10^{14} n/cm^{2}s; however, the lasing threshold in ^{3}He:Ne = 5:1 mixture was not reached [53, 79]. In addition, the luminescence spectrum of He-Ne mixtures at ionized radiation pumping has no 632.8 nm line (**Figure 7**).

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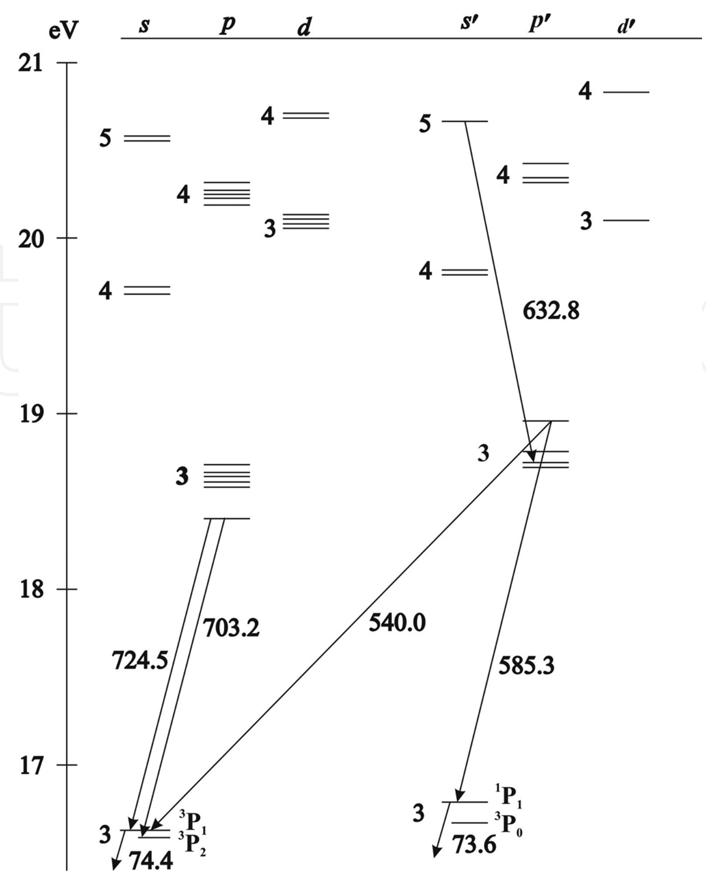

Figure 6. Scheme of laser transitions in neon. The wavelengths of laser and resonant transitions are indicated in nm.

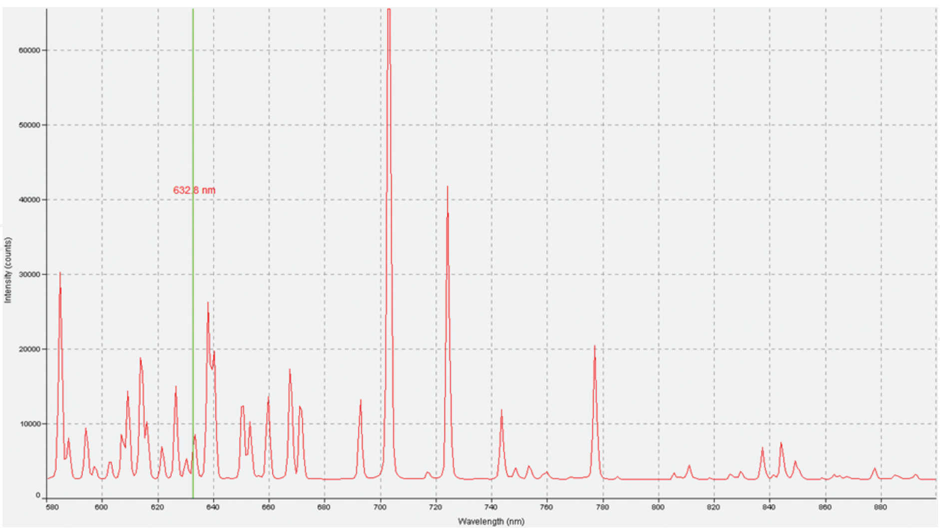

Figure 7. Emission spectrum of neon at a pressure of 605 Torr under ion beam excitation in the 570–900 nm region. The vertical green line indicates the wavelength of 632.8 nm.

3p levels of neon atoms are efficiently populating during excitation by the products of nuclear reactions of neon and its mixtures [57, 80] (see **Figure 7**). To create laser operating on 3p–3s transitions of neon, it is necessary to solve the problem of lower s-levels deactivation. These levels are metastable or resonantly coupled to the main level and have trapped radiation at neon pressures important for nuclear pumping radiation. Rapid depletion of lower laser levels at a relatively low concentration of quenching particles can be achieved in the processes occurring with Coulomb cross-sections. In Ref. [20], authors show the possibility of deactivation of excited states in the Penning process. The required selectivity of deactivation of the upper and lower levels can be achieved using the lower levels states as resonantly coupled to the main, for which the ionization cross-section of quenching by additives is particularly high [81].

Quasi-continuous lasing in allowed neon atom transition $3p'[1/2]_0$–$3s'[1/2]_1^0$ ($\lambda = 585.3$ nm) was first observed in the afterglow of discharge [82], and then at pumping by powerful electron beam of Ne-H_2, Ne-Ar, Ne-Ar-He mixtures [83]. In [84] by reducing the concentration of neon and quenching the lower level of additives by an order compared to [83], and pumping power by electron beam ~100 kW/cm^3 with He:Ne:Ar = 96:3:1 mixtures at a pressure of 3 atm at the same transition obtained laser efficiency of 1–2%. The laser efficiency with ionizing pumping at λ = 585 nm in the opinion of other authors cannot exceed 0.5% [85]. The electron beam pumping provided for laser action in a row of 3p–3s neon transitions in the spectrum red region [86] (see **Figure 6**).

Progress in creating efficient lasers of visible range operating on neon at the electron beam pumping power 10–100 W/cm^3 stimulated work on nuclear-pumped lasers operating on 3p–3s transitions of neon atom. Laser action operating on 3p–3s neon transitions at pumping by uranium fission fragments was obtained in 1985 at VNIIEF on VIR-2 reactor. These results were published in 1990 [3, 87]. In 1985, we have also conducted experiments on WWR-K stationary nuclear reactor for excitation of triple mixtures of argon or krypton, neon, and ^{3}He, which has led to a negative result [53, 79].

Obtained within the experiment efficiency values for NPL operating on neon (to 0.1%) are much lower than at pumping by electron beam [84, 85]. Threshold values of thermal neutron flux density are significantly higher than 10^{14} n/cm^{2}s [87–93]. Report [90] on NPL lasing in ^{3}He-Ne-H_2 mixture with a lasing threshold of 10^{14} n/cm^{2}s is doubtful, as hydrogen at a pressure of 0.57 bars must be considerably quenching the upper laser level. Unlike lasers operating on IR transitions of Xe, Kr, Ar, the lasing mechanism in lasers on 3p–3s transitions of neon was considered reliably established [39, 87, 94]. Already the first works on neon lasers pumped by an electron beam or nuclear radiation contain roughly the same ideas about the lasing mechanism. The lower 3s-states are deactivated in the processes of:

- Penning for metastable states $3s[3/2]_2^0$, $3s'[1/2]_0^0$ [20],
- release of an electron for resonance states $3s[3/2]_1^0$, $3s'[1/2]_1^0$ [81].

The main process of 3p levels population is considered to be dissociative recombination of Ne_2^+ ions with electrons; most studies assumed that all of these levels are populated in recombination of molecular ions in the main vibrational state. In [80, 84] to explain changes in the efficiency of various levels pumping with the neon pressure, was made a conclusion on population of $3p'[1/2]_0$-level in processes of recombination of vibrationally excited levels of

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Ne_2^+. Population efficiency of level $3p'[1/2]_0$ increases in triple mixtures with high helium content, which is explained in [80] by the formation of vibrationally excited levels of Ne_2^+ in the following processes:

$$Ne^+ + 2He \rightarrow HeNe^+ + He \tag{15}$$

$$HeNe^+ + Ne \rightarrow Ne_2^+(v>0) + He \tag{16}$$

Another mechanism for neon levels' population when excited by a hard ionizer is proposed in [79]. Based on the study of spectral-temporal characteristics of pure neon and He-Ne mixtures pumped by heavy charged particles, the conclusion was made that the population of neon levels occurs in direct excitation by nuclear particles and secondary delta electrons, and in He-Ne mixtures also in the process of excitation transfer from metastable helium atoms (He^m):

$$He^m + Ne + He \rightarrow Ne(3p) + 2He \tag{17}$$

However, this work did not receive recognition: In the review article [18], it was not mentioned, and in monograph [3], it was questioned with reference to [95]. During the study of luminescence of He-Ne mixtures with quenching additives [30], we have obtained results confirming the main conclusion in [79]—the principal and clearly dominant channels of neon 3p levels population in pumping by a hard ionizer are the processes unrelated to dissociative recombination of molecular ions of neon.

3.3. Metal vapor lasers

3.3.1. Mercury vapor lasers

HgII laser. The first report on the lasing of ion NPL operating on mercury vapor appeared in 1970 [96]. This work presents results obtained at ^3He(350 Torr)+Hg(3 Torr) mixture pumping on IIN pulsed reactor with neutrons flux up to 5°10^{16} n/cm^{2}s. According to authors, the registered lights power ~10 mW was associated with laser action of mercury ion transition $7^2P_{3/2}$–$7^2S_{1/2}$ (λ = 615.0 nm). Further on, these results were questioned, and in ^4He(600 Torr) +Hg (2.5–10 mTorr) mixture pumping by $^{10}B(n,\alpha)^7Li$ reaction products has proved lasing with a wavelength of 615 nm [97]. The conditions [97] had no lasing at partial pressure of mercury vapor ~3 Torr.

Pumping mechanism at the transition of $7^2P_{3/2}$–$7^2S_{1/2}$ mercury ion at ionizing pumping was considered in [53, 98, 99]. Discrepancy between the results of [96, 97] is possibly due to the difference in mechanisms of upper laser level population at low density and high density of mercury vapor [53, 98]. Calculations [99] indicate a low energy laser characteristics at λ = 615 nm (0.04% maximum efficiency) even with optimal parameters.

HgI laser. The emission spectra of mixtures of ^3He + Hg and ^3He + Hg + Kr in excitation by products of nuclear reaction $^3He(n,p)T$ studied in [100]. It was concluded that the excitation of low-lying HgI levels by electron impact is predominant in high-pressure plasma:

$$Hg(6^1S_0)+e \rightarrow Hg(6^3P_1)+e \tag{18}$$

$$Hg(6^3P_{0,1,2})+e \rightarrow Hg(nx)+e \tag{19}$$

Continuous laser λ = 546.1 nm with optical pumping [101] uses similar scheme of excitation (electron replaced with photon): a beam with a wavelength of 253.7 nm, corresponding the mercury resonance line, excites 6^3P_1 mercury level and beam λ = 435.8 nm transmits excitation to 7^3S_1-level. However, an attempt to ensure deactivation of 6^3P_2 state by N_2 molecules according to the scheme used in optically pumped laser at ionizing radiation pumping was not successful [102].

We have proposed another scheme of creating inverse population in laser on mercury triplet lines [54]. Our works [36, 98] have shown that population of 7^3S_1-level of mercury atom occurs in the process of dissociative recombination of molecular ions and not in direct or stepwise excitation by electrons. It is proposed to use H_2 to destruct the lower level at the transition of 7^3S_1–6^3P_2; H_2, D_2—on the transition of 7^3S_1–6^3P_1. As the pumping is carried out through ion channel, xenon should be used as buffer gas and charge exchange from xenon to hydrogen is slow. The use of krypton is less justified due to low value of rate constant of Kr_2^+ ion recharge on mercury atoms. High selectivity of dissociative recombination of Hg_2^+ is largely driven by relatively low temperatures involved in recombination of electrons. Therefore, by increasing the pumping power to the level required for laser operation, it is useful to use helium to cool the secondary electrons. Thus, the optimal gas mixture of laser operating on mercury triplet must be four-component He-Xe-Hg-H_2 [54].

Quasi-continuous lasing at 7^3S_1–6^3P_2 transition of mercury atoms using this scheme was obtained on pulsed nuclear reactor EBR-L in VNIITF [103]. Kinetic model of He-Xe-Hg-H_2 nuclear-pumped laser based on VNIITF experiments was developed in [104]. Externally similar scheme is implemented in excitation of mercury mixtures with inert gases by electron beam [105]. This work uses a mixture of He+Ne+Ar as a buffer gas at a total pressure of 2300 Torr. With reference to the paper later than [36, 98], recombination of Hg_2^+ as the main population channel of 7^3S_1 has been specified. An attempt to use H_2 for 6^3P_2 level population was unsuccessful, and at hydrogen pressure of 20 Torr laser action failed. What was also interesting in this study was the absence of molecular additives quenching the lower level. Apparently, the de-excitation of lower laser level took place in formation of excimer molecules (HgR)*:

$$Hg(6^3P_2)+R+M \rightarrow (HgR)^*+M \tag{20}$$

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where R—Ne or Ar.

3.3.2. Cadmium and zinc vapor lasers

The first nuclear pumping of visible range laser operating on cadmium vapor was carried out by MEPhI researches on BARS pulse reactor [106]. In ^{3}He-^{116}Cd mixture was obtained laser action on $4f^2F^0_{5/2,7/2}$–$5d^2D_{3/2,5/2}$ transitions of ion Cd^+ (λ = 533.7 and 537.8 nm), and later on transition from $\lambda = 441.6$ nm [107]. The first successful experiments on laser pumping on metal vapor (mixture of He-^{116}Cd, $\lambda = 441.6$; 533.7 and 537.8 nm) by uranium fission fragments were carried out in 1982 by researches of VNIIEF and VNIITF on EBR-L reactor [3]. Maximum parameters of Cd^+-laser with nuclear pumping is also obtained in this reactor: 1000 W at an efficiency of 0.4% on the blue line and 470 W on the green lines [44].

At present, the basic processes of population of upper laser levels of Cd^+ are considered to be established:

$4f^2F^0_{5/2,7/2}$ upper levels of laser transitions from $\lambda = 533.7$ to 537.8 nm are populated due to charge exchange processes

$$He^+ + Cd \to (Cd^+)^* + He \tag{21}$$

forming higher-lying levels of 6f, 6g, 8d, 9s [108] and subsequent cascade transitions in 4f state.

$5s^{22}D_{3/2,5/2}$ upper levels for transitions with $\lambda = 441.6$ and 325 nm are populated in Penning and charge exchange processes

$$He_2^+ + Cd \to (Cd^+)^* + He \tag{22}$$

$$He^*(2S) + Cd \to (Cd^+)^* + He \tag{23}$$

Laser action at 325 nm was achieved only when pumped by an electron beam, although the threshold pumping power was only 10 W/cm^3 [109].

Deactivation of lower levels can occur as a result of radiative transitions, quenching by electrons, conversion processes of atomic ions into molecular ions

$$(Cd^+)^* + Cd + He \to Cd_2^+ + He \tag{24}$$

and for the levels lying above $6^2S_{1/2}$, in Penning process on its own atom

$$(Cd^+)^* + Cd \to Cd^+ + Cd^+ + e \tag{25}$$

Lasing mechanisms of NPLs on ion transitions of cadmium and zinc are very similar. Laser action by pumping ^{3}He-Zn mixture observed on transition of $4s^{22}D_{5/2}$–$4p^2P_{3/2}$ (λ = 747.9 nm) [110], 60 W output power obtained during pumping by uranium fission fragments at this transition [44].

Information about cadmium-vapor atomic lasers with ionizing pumping is scarce. During experiments was registered laser action (1–2 W) with a threshold density of 10^{16} n/cm^{2}s neutron flux on the lines of 1.648 and 1.433 μm at pumping He-Cd mixture by uranium fission fragments [44]. When pumping He-Cd mixture by an electron beam was obtained laser action on the line 361 nm of cadmium atom [111]. Kinetic model [112] included processes involving excited cadmium atoms and attempted to calculate some laser characteristics for 1.648 μm line.

Luminescence of ^{3}He-Cd and ^{3}He-Xe-Cd mixtures in the radiation region of stationary nuclear reactor investigated in [113]. Measured value of rate constant of Xe_2^+ charge exchange on cadmium atoms is small (~10^{-13} cm^{3}s^{-1}), in contrast to the constant of charge exchange on mercury atoms. A sufficiently high density of cadmium vapor (~3×10^{18} cm^{-3}) was established at a temperature of about 700°C, and such density of cadmium requires consideration of quenching 6^3S_1 state by its own atoms. Perhaps krypton ions charge exchange on Cd will be faster. In addition, cadmium atoms in krypton can be ionized in the Penning processes.

4. NPL active media on molecular transitions

4.1. Lasers operating on first negative system of nitrogen and carbon monoxide

N_2^+. When pumping active medium of a high pressure by electron beam with moderate (~3 A/cm^2) current density was obtained quasi-continuous lasing mode on the first negative system of nitrogen (λ = 391.4 and 427.8 nm) with an efficiency ~1% [114, 115]. In this collision lasers on $B^2\Sigma^+_{u,v=0} \rightarrow X^2\Sigma^+_{g,v''=0,1}$ transitions of nitrogen ion, deactivation of lower level was carried out at low hydrogen concentrations (~0.1%) in He-N_2-H_2 mixture in the process with proton transfer:

$$N_2^+(X) + H_2 \rightarrow N_2H^+ + H \tag{26}$$

In 1996 was obtained laser action at λ = 391.4 nm with excitation of He-N_2-H_2 mixture by uranium fission fragments on EBR-L [116]. This was the first NPL emitting in the UV spectral region: Laser power was ~10 W, efficiency ~0.01%. To date, it remains to be the most short-wavelength nuclear-pumped laser. On BARS-6 reactor were conducted studies of facility with active core length of 250 cm and volume of 4 liters with the average over the length of the laser element-specific energy deposition to 300 W/cm^3 [117]. The lasing threshold on B-X nitrogen ion transitions with λ = 391.4 and 427.8 nm was achieved with low for molecular laser power density of energy deposition 50–60 W/cm^3. It was determined that active medium had non-resonant losses ~5×10^{-5} cm^{-1}. These authors explain the laser efficiency (~0.1–0.2%) was significantly lower than expected. In [118] developed a multi-

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component spatially homogeneous model of kinetic processes of NPL active medium on He-N_2-H_2 mixture. According to the model, the maximum laser efficiency (0.5–0.8%) was achieved at pumping power of 0.5–3 kW/cm^3, for specific pumping power 90–130 W/cm^3 the maximum instantaneous efficiency is 0.1%.

The [119] searched mixture compounds that improve NPL efficiency on 1^-—nitrogen system, including study of deuterium and neon impact on the laser parameters. Replacement of hydrogen additive deactivating lower level by deuterium provided no improvements to laser efficiency. The use of neon as the buffer gas at partial pressure of 30 Torr resulted in efficiency decrease for 30–40% for both wavelengths. It has previously been shown [120] that helium replacement with neon impairs laser parameters for B-X transitions of N_2^+. At pumping by an electron beam, lasing energy of mixture with addition of 60 Torr of neon was 1.5 times less than for three-component mixture of He-N_2-H_2 with a total pressure of 6 atm.

In [121], the constants of quenching processes rates of N_2^+ (B) with nitrogen and helium, as well as two- and three-particle charge exchange processes of He_2^+ to H_2, D_2, Kr, CO, were determined on the luminescence in 0–0 transition of the first negative system of nitrogen in excitation by ∝–particles of polonium-210.

CO^+. Gain on the first negative system of carbon monoxide by powerful electron beam pumping was obtained in 1975, the same way as in N_2^+-laser, by Waller et al. [122]. Quasi-continuous lasing mode can be implemented with addition of H_2, D_2 or Kr to He-CO [123]. In [123, 124] by spectra of radioluminescence of the gas mixtures with carbon monoxide was determined constants of quenching rate of CO^+(B) with helium, neon, CO molecules, evaluated the upper limit of constant of quenching rate with hydrogen, deuterium and krypton.

Kinetic model of nuclear-pumped laser on $B^2\Sigma^+_{u,v'} \rightarrow X^2\Sigma^+_{g,v''}$ transition of carbon monoxide ion was developed in [125]. Unfortunately, the only information given on the results of calculations of medium gain, efficiency, but there is no information on considered processes, values of constants for processes rate. This provides for the opportunity to discuss the work. In addition, specified calculated wavelength transition for which the calculations were made, 210 nm; the closest wavelength—211.2 nm—corresponds to 1–0 transition [126]. That is, laser action, according to [125], should not take place from the ground vibrational level of CO^+(B), as the wavelength of 0–0 transition—219.0 nm. It should be noted that the gain in [122] was observed at 0-2 transition (242 nm) and Baldet-Johnson's system (391 and 425 nm, B→A transitions) from the ground vibrational level of CO^+(B).

4.2. Excimer lasers operating on halides of inert gases

Excimer lasers operating on halides of inert gases have been studied for a long time [127]. At present, they are the most powerful lasers that emit in UV region of spectrum. Optimal operation of excimer lasers corresponds to pumping powers of several megawatts per cm^3 and a pressure of several atmospheres. Such pumping powers are achieved by electron beams or space discharge. Radiation of nuclear explosions [128] and ion beams [129] also has been used to pump these lasers.

Excimer lasers can operate in a quasi-continuous mode, as in the photon emission, an excimer molecules pass into the lower dissociating or weakly bound state. Much of the research on creation of NPL operating on the inert-gas halides is associated with XeF-laser (λ = 351 and 353 nm). Experiments on SPR-III reactor have shown that in ^{3}He-Xe-NF_3 mixture the gain on the 351 nm band is about 7°10^{-3} cm^{-1} at pumping power q≈5 kW/cm^3 [130], and at pumping by uranium fission fragments of Ne(Ar)-Xe-NF_3 mixture, registered gain of ~2°10^{-3} cм$^{-1}$ at q≈2 kW/cm^3 [131]. Experiments in pulsed nuclear reactors with neutron flux up to 10^{17} n/cm^{2}s aimed at obtaining lasing at XeF [131] and XeF, KrF [59] did not yield positive result. The work [132] contains theoretically investigated characteristics of laser operating on Ne-Xe-NF_3 mixture at 1 atm with nuclear pumping duration 0.1–1 ms at half-height in the near-threshold lasing region. The lasing threshold is 400–500 W/cm^3, and the maximum calculated efficiency of ~1% is achieved at pumping power of 1.5–5 kW/cm^3. Predicting the possibility to create XeF-nuclear pumped laser is mainly limited by the uncertainty of absorption coefficient in active medium, which greatly affects calculation results.

The luminescence efficiency in chlorine-containing gas mixtures with xenon (to 15% [133], ~11% [134]) is about three times higher than the efficiency of emission at the transition of XeF. The maximum luminescence efficiency can be achieved at low chloride content (<0.7 Torr [134], 0.05 Torr [135]). In [133] reported about the lasing on XeCl molecule (λ = 308 nm) by pumping Ar-Xe-HCl(CCl_4) mixture by uranium fission fragments in experiments on EBR-L reactor. According to the authors, the narrowing in emission spectrum in 308 nm band, the height of film blackening, 1.3–1.5 times less than at other wavelengths, indicates the presence of laser radiation. However, these data are not sufficient for this conclusion [3].

High radiation resistance of ^{3}He-Xe-CCl_4 mixture was noted in [134, 136]. The densities of electrons and negative ions in the plasma of gas mixture of ^{3}He-Xe-CCl_4 and ^{3}He-Xe-NF_3 irradiated with thermal neutron flux 10^{11}–10^{14} n/cm^{2}s into the active core of stationary nuclear reactor, measured in [137].

4.3. Lasers operating on vibrational transitions of carbon monoxide and carbon dioxide

CO. Molecular nuclear-pumped laser operating on carbon monoxide was one of the first NPLs, the creation of which was reported in the press [64]; the laser action was observed in vibrational-rotational transitions of the CO molecule with λ = 5.1–5.6 μm. Potential of medium on carbon monoxide was determined by the fact that, unlike lasers on electronic transitions, active medium of CO-laser does not require high pumping selectivity. It is important to get energy into the broad band of vibrational levels of the ground electronic state of CO molecule. Furthermore due to the autonomy of vibrational subsystem and anharmonicity of molecular vibrations, this energy at relatively low translational temperature redistributing in the process of exchange of vibrational molecules to provide full or partial inversion on vibrational levels of CO [138]. This property associated with the anharmonicity of the vibrational levels, as well as the cascade mechanism of lasing in CO, allows directing significant energy share of nuclear reaction products to the laser level.

Experiments have not yet confirmed this finding. In [64], carbon monoxide pumping at a pressure of 0.13 atm and temperature of 77 K was carried out by uranium fission fragments,

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and the radiation power was 2–6 W with an efficiency of 0.1–0.3%. In further work, the authors [64] achieved lasing power of about 100 W by using a multiple-pass resonator with an active length of 120 cm. Lasing on vibrational transitions of CO was also obtained in [139] at excitation of ^{3}He-CO by nuclear reaction products of ^{3}He(n,p)T. Laser radiation power at the mixture pressure of 3 atm exceeded 200 W from an active volume of 300 cm^3, and the lasing threshold was reached at $F = 3\times10^{16}$ n/cm^2s.

Excitation of vibrational levels of CO can occur due to molecular collisions with plasma electrons. In [140] based on calculation of electrons spectrum formed in a molecular gas under the action of ionizing radiation, it was shown that the efficiency of CO nuclear-pumped laser cannot exceed 0.5%. Further work [141] showed the main mechanism of molecular formation in the form of dissociative recombination of cluster ions with formation of electronically excited molecules and subsequent collision of these molecules with molecules in ground state. According to the authors [141] plasma chemical processes in active medium can make a significant contribution to the energy pumping into vibrational modes of molecules and allow achieving the efficiency pumping up to 18%. The use of argon as a buffer gas instead of helium should increase 1.5-fold the efficiency of pumping in vibrational levels and reduce by an order the threshold energy for active medium pumping. It should be noted that a record efficiency of electron beam-controlled laser operating on carbon monoxide—63% [142]—was achieved due to Ar:CO = 10:1 mixture.

Presently achieved low parameters of NPL operating on CO, the need to cool down an active medium reaching cryogenic temperatures, apparently, makes carbon monoxide medium insufficient for creation of nuclear-pumped lasers.

CO_2. The possibility of CO_2-laser pumping (λ = 10.6 μm) by nuclear radiation was considered in the earliest stages of NPL study [2], and gas-discharge laser operating on carbon dioxide had the highest output parameters for that time. Numerous attempts to create NPL on CO_2 have yield negative results. Experiments on pumping ^{3}He-CO_2 and ^{3}He-CO_2-N_2 mixtures with products of ^{3}He(n,p)T reaction showed no gain in band of 10.6 μm at a wide variation of pressure (0.28–0.8 atm) and composition of mixture [143]. Moreover, these experiments showed probe laser radiation absorption, which indicates preferential population of the lower laser level of CO_2 when excited by ionizing radiation. Calculations of kinetic processes in CO_2-N_2-He mixture also support the conclusion on ineffectiveness of direct pumping of CO_2-laser with nuclear radiation [144].

Apparently, the most promising method of nuclear energy conversion into radiation on vibrational transitions of CO or CO_2 is the creation of nuclear power plant with electroionization laser based on thermionic converter reactor [145, 146].

4.4. Radiation of heteronuclear ionic molecules of inert gases

Molecular bands in radiation spectra of pair inert gas mixtures were first discovered more than half a century ago. The [147] recorded the band of 507–550 and 496–508 nm in Ar-Xe mixture, and the authors attributed the presence of these bands with emission of heteronuclear molecules or ions. When pumping Ar-Xe mixture by electron beam, the molecular band at 510 nm was detected and the band of 495–460 nm in Kr-Xe was registered for the first time [148]. Two emission systems were observed, in 600–670 and 670–685 nm regions, when Xe was added to Kr flowing afterglow at a pressure of 30 Pa [149]. Kugler [150] obtained similar results for Ar-Xe mixture and discovered new band in Ar-Kr in the region of 605–642 nm. He explained these bands as transitions of neutral heteronuclear molecule formed in the processes of metastable atoms of argon.

In 1975, Tanaka et al. [151] have published data on radiation spectra of 10 pair mixtures of inert gases in the region of 100–700 nm. Molecular bands observed in radiation spectra in the discharge were identified as transitions between states of heteronuclear ionic molecules:

$$M^{+}N \rightarrow MN^{+} + h\nu \tag{27}$$

where molecular states of M^+ N asymptotically correspond to states of M^++N, and MN^+ to the state of M+N^+; here M, N—atoms of inert gases, and N—a heavier atom. If the plasma of low pressure in an electric discharge in paired mixtures of inert gases has up to 5 similar bands [151], there are no transitions from levels corresponding to the states of atomic ions $^2P_{3/2}$ [152, 153] when excited by ionizing radiation of medium- and high-pressure mixtures.

Kinetics of Ar-Kr, Ar-Xe, and Kr-Xe mixtures' excitation by low activity ^{241}Am alpha particles was studied in [152, 154]. Constants of processes rates in these mixtures were identified; however, constants' values of a number of processes are underestimated: ~10^{-15} cm^3/s for two-particle and ~10^{-34} cm^6/s for three-particle processes. Emission of Ar-Xe and Kr-Xe mixtures when excited by ^{210}Po alpha particles with activity of ~0.5 Cu investigated in [153, 155–157] determined the rate constants of processes of formation and destruction of levels of heteronuclear ionic molecules. In [153] includes first noted high luminescence efficiency of $(ArXe)^+$, $(KrXe)^+$ at pumping by ionizing radiation. Luminescence of Ar-Xe mixture pumped by powerful electron beam was studied in [158], attempts to obtain lasing on transitions of $(ArXe)^+$ at pumping by an electron beam had yield negative results [155, 158].

In [42] was built kinetic model of Ne-Ar mixture relaxation pumped by a hard ionizer with regard to the possibility of lasing on transition $Ne^+Ar \rightarrow NeAr^+$. When using typical rates of plasma chemical reactions, calculations show that lasing is only possible at high pressure (above 16 atm) and powerful pumping (1 MW/cm^3), and lasing efficiency should not exceed 0.05–0.25%. In this work were considered the triple (with Kr) instead of the binary mixtures of inert gases, as the authors suggested that the constant of deactivation rate of the lower level in exchange processes:

$$NeAr^{+} + Ar \rightarrow Ar_2^{+} + Ne \tag{28}$$

may occur negligible. It was assumed that the lower laser level will be deactivating in the processes with Kr atoms:

$$NeAr^{+} + Kr \rightarrow Kr^{+} + Ne + Ar \tag{29}$$

$$NeAr^{+} + Kr + M \rightarrow Kr^{+} + Ne + Ar + M \tag{30}$$

5. Conclusions

Nuclear-pumped lasers are of great interest as the way to extract high-quality energy from a nuclear reactor core. Presently achieved pulse power of NPL in quasi-continuous mode exceeds 1 MW. However, the most promising active media on transitions of inert gas atoms have a number of disadvantages: relatively low efficiency, radiation in IR region, low operating temperature. There is no clarity as to the basic mechanism of upper laser level population: direct excitation by secondary electrons, excitation transfer from buffer gas atoms, electron-ion recombination of molecular ions (dimers or heteronuclear ions). This area requires further research.

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