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Mechanisms of level population in gas lasers pumped by ionizing radiation

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Abstract

The mechanisms of level population in high pressure gas lasers pumped by ionizing radiation at the 3*p*-3*s* transitions of neon, the *d*-*p* transitions of argon, krypton, xenon, and triplet lines of mercury are analyzed. It is shown that dissociative recombination of molecular ions with electrons is not the basic process responsible for populating the *p* levels of inert gas atoms. It is assumed that the most likely channel for *d*-level population is direct excitation of atoms by secondary electrons and excitation transfer from buffer gas atoms, with *p* levels being populated by transitions from upper levels. Dissociative recombination of mercury molecular ions with electrons is the basic process responsible for populating the 7^3S_1 level of mercury atoms.

Keywords: Laser; Mercury; Nuclear pumping; Population mechanism; Rare gases

1. INTRODUCTION

The study of optical (laser or spontaneous) radiation of nuclear-excited plasmas is of interest for the development of ways to extract energy from nuclear reactors as well as for the control and adjustment of reactor parameters. It is likely that, in the future, nuclear-pumped lasers will be used for a wide range of applications (Shneider & Hohl, 1984; Mel'nikov *et al.*, 2008), especially when it is necessary to place powerful compact lasers at autonomous remote sites. Direct pumping of active laser media is, as a rule, executed by products of nuclear reactions with thermal neutrons from a nuclear reactor: ^{3}He(n,p)^{3}H, ^{10}B(n,α)^{7}Li, ^{235}U(n,f)F, and others. The active medium of the laser should contain ^{235}U, ^{3}He or ^{10}B, or compounds of these isotopes should be applied to the walls of the laser chamber. There is also substantial interest in connection with the differences between the level population mechanisms in nuclear pumping and the population processes in conventional gas-discharge lasers. The characteristics of the laser radiation resulting from pumping by a hard ionizer depend on the power and duration of energy deposition into the active medium of the laser but not on the type of ionizer (Mel'nikov *et al.*, 2008). This means that the kinetics of processes in the active media of lasers excited by electron or ion beams and those of lasers with nuclear pumping will be the same (Fedenev & Tarasenko, 1998; Ulrich, 2012). Heavy ions have a very high energy loss in matter, which scales quadratically with the charge of the projectile that allows achieving high pumping power densities in the laser medium (Ulrich *et al.*, 2006).

Currently known lasers with nuclear pumping (Mel'nikov *et al.*, 2008) radiate in the spectral band 391–5600 nm at about 50 transitions of Xe, Ar, Kr, Ne, C, N, Cl, O, I, and Hg atoms, Cd^+, Zn^+, and Hg^+ ions, the CO molecule, and the N_2^+ molecular ion. Lasers with nuclear pumping at the Xe atomic transitions have been explored most thoroughly, with pulsed power values of 1.3 MW and energy in pulses of 520 J that are the highest known for lasers with nuclear pumping (Zagidulin *et al.*, 2012). At the Ar-Xe mixture pumped by uranium fission fragments laser pulse duration of 1.5 s was achieved (Mel'nikov *et al.*, 2008). A low threshold of lasing and the absence of degradation and chemical reactivity of the active gas mixture are the most important advantages of lasers with infrared (IR) *d*-*p* transitions of inert gas atoms. On the other hand, the disadvantages of these lasers include radiation in the IR region, comparatively low efficiency (about 3%) and an operating temperature (up to 350–550 K) that is low for nuclear reactor conditions (Magda, 1993; Hebner, 1995). Although lasing at the IR transitions of inert gases has been studied for about 40 years and is considered to show the most promise, the population mechanism of the upper *nd* levels (n = 5, 4, and 3 for

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Xe, Kr, and Ar, respectively) is not yet clear (Ohwa *et al.*, 1989; Shon & Kushner, 1994; Mel'nikov *et al.*, 2008; Apruzese *et al.*, 2008). The processes of deactivation of the lower $(n+1)p$ levels appear to be clearly identified, namely collision quenching on encounters with atoms of the active medium and, at high pumping powers, with electrons. The main problem with determining the mechanism of *d*-level population is connected with difficulties in measuring IR radiation under the conditions of a reactor experiment. In this work, conclusions regarding the level population mechanisms of some lasers with direct nuclear pumping are made on the basis of analysis of luminescence spectra of nuclear-induced plasmas of gas mixtures.

2. EXPERIMENTAL SETUP

Investigation of the luminescence spectra of nuclear-excited gas mixtures was conducted with radioisotope pumping. A cylinder of diameter 25 mm and length 70 mm, carrying on its surface 18 sources containing ^{210}Po, was installed in a stainless steel chamber. Under normal conditions, the maximum range in He of α particles of energy 5 MeV is 183 mm. Before installation of the sources, the chamber was heated and degassed in a vacuum of about 10^{-3} Pa. After installation in the chamber, the α sources were pumped without heating for two to three weeks until well-reproducible (up to 3–7% of intensity for different gases) luminescence spectra were obtained. The gas pressure was measured by a vacuum manometer and a VDG-1 vacuum meter. The purity of the spent gases was Ne 99.996%, He 99.99%, Ar 99.992%, and Kr 99.999%. Hydrogen and deuterium (enrichment by D_2 was 99%, nitrogen impurities about 0.1% and oxygen about 0.05%) were purified by passage through silica gel and active copper. The emission spectrum was analyzed by an SPM-2 monochromator with quartz prism and a FEU-106 photomultiplier operating in photon-counting mode. The activity of the α sources was 9.6 GBq, which corresponds to an average energy deposition under 2 atm of helium of $W \approx 3\times10^{-5}$ Wcm^{-3} and a mean ionization rate according to gas volume of $S \approx 4\times10^{12}$ cm^{-3}s^{-1}. The apparatus for study of luminescence spectra of gas mixtures in the active zone of a nuclear reactor is described in Khasenov (2004; 2014).

3. NEON

Lasing on three neon lines in the visible region (Fig. 1) with wavelengths of 585.3 nm ($3p'[1/2]_0$-$3s'[1/2]_1^0$ transition), 703.2 nm ($3p[1/2]_1$-$3s[3/2]_2^0$), and 724.5 nm ($3p[1/2]_1$-$3s[3/2]_1^0$) was achieved under pumping mixtures with neon by products of nuclear reactions (Voinov *et al.*, 1990; Hebner, 1993). Processes in the active media of lasers at $3p$-$3s$ transitions of NeI are considered to be well studied (Shon *et al.*, 1993*a*; Karelin & Yakovlenko, 1995): population of the upper laser level is due mainly to dissociative recombination of Ne_2^+ and $HeNe^+$ molecular ions. Under

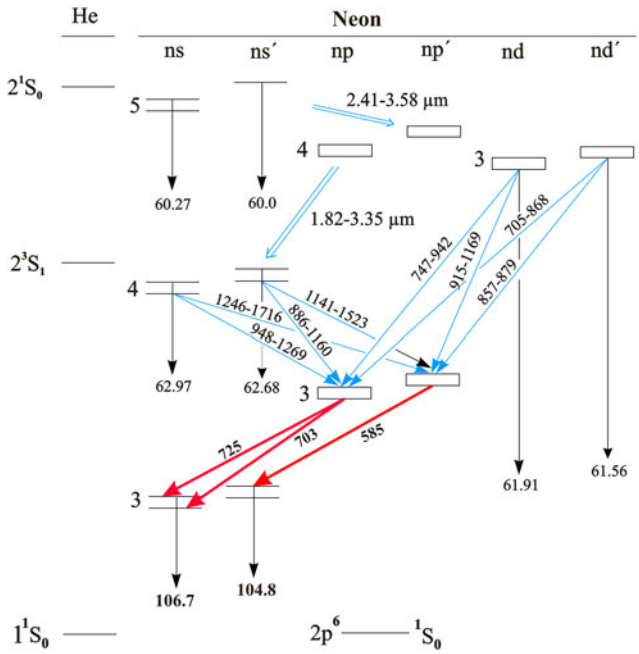

Fig. 1. (Color online) Scheme of population of Ne($3p$) levels, cascade transitions (blue), which are populated $3p$ levels of neon are indicated. Lasing lines (red) and resonance lines of Ne are also shown. Wavelengths are in nm and μm.

comparatively weak pumping, the $HeNe^+$ ions also form Ne_2^+ ions in the substitution reaction

$$HeNe^+ + Ne \rightarrow Ne_2^+ + He.$$

Assuming that the dependence of the luminescence intensity of the 585 nm line is determined by competition between Ne_2^+ charge exchange with the quenching additive and recombination of electrons with Ne_2^+, we obtain the additive concentration P at which the intensity is halved. In this case, the rate of recombination of molecular ions Ne_2^+ is equal to the rate of ions recharge on additive:

$$\beta[Ne_2^+]n = k[Ne_2^+]P,$$

where k is the coefficient of Ne_2^+ charge exchange with the additive, β is the coefficient of Ne_2^+ recombination with electrons and n is density of electrons. Taking the values of the coefficients of recombination of basic molecular ions in the plasma approximately equal to the β value for Ne_2^+ ion, we obtain:

$$\begin{aligned} n &\approx \sqrt{S/\beta}, \\ P &\approx \sqrt{\beta S}/k. \end{aligned} \qquad (1)$$

Figure 2 shows the measured dependence of the luminescence intensity of the 585 nm line in a He-Ne mixture on the pressure of quenching additives under radioisotope pumping. Dependence of the intensity at $\lambda = 703$ nm on

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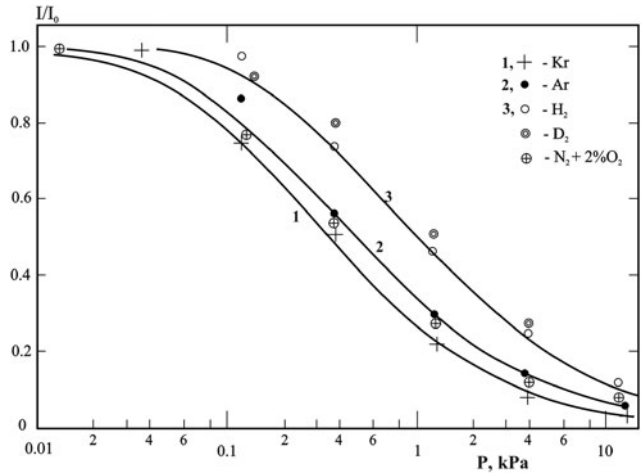

Fig. 2. Dependence of luminescence intensity at λ = 585 nm on pressure of additives to the He(200 kPa)-Ne(6.7 kPa) mixture. I_0 is the intensity in the He-Ne mixture without additives.

the H_2 or Kr pressure in the He-Ne-H_2(Kr) mixtures is similar to the dependence for 585 nm. For hydrogen, $k = 1.3 \times 10^{-10}$ cm^3s^{-1} (Mayhev, 1992); then, at the mean ionization rate, $P \approx 8 \times 10^{12} cm^{-3} \approx 3 \times 10^{-2}$ Pa. The measured values of P correspond to 370 Pa for Kr, 505 Pa for Ar, and 1070–1200 Pa for H_2 and D_2 in a mixture with 200 kPa of He and 6.7 kPa of Ne (see Fig. 2). For comparison, the P value measured for nitrogen was 465 Pa, and the k values for nitrogen in the literature are 8.2×10^{-10} cm^3s^{-1} (Mayhev, 1992) and 8.6×10^{-10} cm^3s^{-1} (Collins & Lee, 1980).

Population of the $3p'[1/2]_0$ level of NeI on excitation by heavy particles seems to be absent during the dissociative recombination of molecular ions. The addition to the He(200 kPa)-Ne(6.7 kPa) mixture of up to 13.3 kPa of nitrogen with 2% O_2 impurity led to the same drop in intensity as for pure Ar and Kr (see Fig. 2), implying that the process of electron attachment to an electronegative impurity has no effect on population of the $3p'[1/2]_0$ level of Ne. A similar result, also under radioisotope pumping, was obtained by Mavlyutov *et al.* (1993): the line intensity at 585 nm in a Ne(100 kPa)-O_2(0.3 kPa) mixture was only half that in Ne at a pressure of 100 kPa, and the intensities of the lines at 703 and 725 nm were one-quarter. In comparison, under nuclear pumping of mercury-containing mixtures, population of HgI levels occurs during dissociative recombination of Hg_2^+ ions (Batyrbekov *et al.*, 1988). Adding 13 Pa of O_2 to the ^{3}He-Hg mixture leads to an approximately 500-fold weakening of triplet lines and the resonance line of Hg (Khasenov & Smirnova, 2008), which is connected to electron attachment to O_2 molecules. Apparently, the process of He_2^+ recharge on O_2, which competes with charge exchange on Hg atoms, is not sufficient in this case, since the O_2^+ ions will also be recharged on the Hg atoms.

It was concluded by Poletaev *et al.* (1992) that population of the Ne levels occurs under direct excitation by nuclear particles and secondary delta electrons. In He-Ne mixtures, population also occurs through excitation transfer from He metastable:

$$He^m + Ne + He \rightarrow Ne(3p) + 2He.$$

These conclusions were based on a study of the spectral-temporal characteristics of pure Ne and He-Ne mixtures under pumping by heavy charged particles.

From our point of view, more-probable channels for 3*p*-level population in the mixture with He are cascade transitions from the 4*s* levels:

$$He^m + Ne \rightarrow Ne(4s) + He;$$
$$Ne(4s) \rightarrow Ne(3p) + h\nu.$$

It is known that the Ne(4*s*) levels are close to the He(2^3S_1) level, and the operation of the He-Ne laser at 1.15 μm is based on excitation transfer to Ne atoms from He(2^3S_1). The absence of 4*s*-3*p* transition lines in high-pressure mixtures (Poletaev *et al.*, 1992) is connected with the location of this transition in the IR region of the spectrum, beyond the sensitivity limit of photomultipliers. In the work of Abramov *et al.* (2006), where measurements were made up to 1100 nm, in the excitation of Ne and He-Ne mixture by uranium fission fragments, there is a line at 966.5 nm corresponding to the $4s[3/2]_2$-$3p[1/2]_1$ transition. Abramov *et al.* (2006) also identified more than 10 lines of the 3*d*-3*p* transitions. In these transitions, it is mainly the lowest three 3*p* levels that are populated.

The intensity changes at 585 nm are connected not only with quenching of the $3p'[1/2]_0$ level by additives. The quenching rate constant is 4.6×10^{-11} cm^3s^{-1} for H_2 and 5.3×10^{-11} cm^3s^{-1} for Ar (Burstein *et al.*, 1991), and, considering the lifetime of the level (14.3 ns), we can obtain the value of the H_2 or Ar pressure (about 5 kPa) at which the quenching rate is comparable to the rate of spontaneous decay of the level. The decrease in luminescence intensity with increasing partial pressure of quenching additives appears to be connected mainly with the Penning process of He metastable on the additive.

Considering that the dependence of the intensity at 585 nm in He(200 kPa)-Ne(6.7 kPa)-H_2 (Ar, Kr, D_2, or N_2) is determined by competition between non-resonance excitation transfer from He(2^3S_1) to Ne and the Penning process of He(2^3S_1) on the quenching additive, we may estimate the value of the quenching additive pressure at which the intensity is halved (Table 1). A satisfactory fit of the estimated and measured P values leads to the conclusion that under ionizing pumping of He-Ne (with a substantial He content), 3*p*-level population occurs as a result of excitation transfer from He(2^3S_1) to Ne and at subsequent cascade 4*s*-3*p* transitions.

Adding 6.7 kPa of Ar to Ne at a pressure of 100 kPa as well as at 300 kPa leads to a two-fold decrease in intensity at λ = 585 nm and a three-fold decrease at 703 nm, which

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Table 1. *Processes of He(2^3S_1) quenching. Estimates of P for a He-Ne(6.7 kPa)-M mixture (M = H_2, D_2, Ar, Kr, or N_2) with the rate constants K of the processes*

Atom or molecule	Process	K (10^{-11} cm^3 s^{-1})		P (Pa)	
		Smirnov (1982)	Lindiger *et al.* (1974)	Estimate	Experimental value
Ne	$He(2^3S_1) + Ne \rightarrow Ne(4s) + He$	0.45	0.36		
H_2	Penning	5 ± 3	2.9	~900	1070
D_2	Penning		2.6	~1000	1200
Ar	Penning	9 ± 5	7.1	~400	505
Kr	Penning	11		~300	370
N_2	Penning	7.2 ± 1.4	7.1	~400	465

correspond approximately to the 3*p*-level quenching by Ar. The presence of intense radiation in the 3*p*-3*s* transitions on Ne excitation by α particles (Batyrbekov *et al.*, 1990) and by spontaneous fission fragments of ^{252}Cf ($W \approx 10^{-8}$ Wcm^{-3}) (Poletaev *et al.*, 1992) leads to the conclusion that in pure Ne, 3*p*-level population occurs through processes not connected with dissociative recombination of Ne_2^+ ions. At low degrees of ionization, Ne_2^+ ions are preferably recharged by impurities in the gas. Under pumping by uranium fission fragments (Gorbunov *et al.*, 2004), the luminescence intensity of Ne(64 kPa)-NF_3(200 Pa) is approximately equal to the emission intensity in the 3*p*-3*s* transitions in Ne with the addition of 1.06 kPa of Ar, Kr, or Xe. As with the He-Ne mixture, the presence of an electronegative impurity in Ne does not lead to a rapid drop in radiation intensity in the 3*p*-3*s* transitions.

It has been shown that a scheme in which Ne 3*p*-3*s* transitions are excited by dissociative recombination of Ne molecular ions with electrons is not consistent with experimental data on weak pumping by ionizing radiation: (1) The effective luminescence of 3*p*-3*s* transitions observed with admixtures at pressures of hundreds of Pascal's, at recombination mechanism of the levels populating the radiation intensity at $W \sim 3 \times 10^{-5}$ Wcm^{-3} would rapidly decrease at H_2 or N_2 pressures of about 0.01–0.1 Pa. (2) Processes of electron attachment to electronegative impurities (O_2 and NF_3) have no effect on population of the Ne $3p'[1/2]_0$ level.

Excitation transfer to Ne atoms from metastable He atoms and direct excitation of Ne by nuclear particles and secondary electrons are assumed to be the most likely channels of Ne(3*p*) population (see Fig. 1). In Ne, the 3*d*, 4*s*, and 5*s* levels are excited by electrons, and population of Ne(3*p*) occurs in cascade transitions from these levels. The 3*d*, 4*s*, and 5*s* levels optically connected with the ground state are effectively excited by electron impact. The absence of lines representing transitions from the 5*s* levels (Batyrbekov *et al.*, 1990; Poletaev *et al.*, 1992) is explained by the fact that the probabilities of the 5*s*-3*p* transitions are far less than those of the 5*s*-4*p* transitions in the IR region of the spectrum. Cascade transitions 5*s*-4*p*, 4p-4*s*, and 4*s*-3*p* may also contribute to population of the 3*p* levels of NeI. In He-Ne mixtures, population of the 3*p* levels occurs also in the processes of non-resonance excitation transfer from $He(2^3S_1)$ and He (2^1S_0) metastable atoms to the 4*s* and 5*s* levels of Ne and in the subsequent cascade transitions.

Lasing of the He-Ne-Ar mixtures pumped by powerful beam of electrons with duration of about 5 ns was investigated by Shon *et al.* (1993*a*). Laser radiation at 585 nm occurred with a delay of 20–40 ns, and continued for about 200 ns after the pump pulse. According to Mel'nikov *et al.* (2008), these results indicate the recombination mechanism of 3p levels population. In our opinion, the delay of laser pulse and its long duration should be associated with cascaded population of 3p levels of neon from higher levels, as well as transfer of energy from exited helium atoms to neon.

4. ARGON, KRYPTON AND XENON

The main mechanisms for population of the *nd* levels that have been discussed previously (Mel'nikov *et al.*, 2008) are as follows (B = Xe, Ar, Kr, and A = buffer gas atom): (1) impact-radiation recombination: $B^+ + e + e(M) \rightarrow B(nd) + e(M)$ (M = third particle); (2) recombination of heteronuclear ion molecules: $AB^+ + e \rightarrow B(nd) + A$ (probably in cascade transitions from the upper *p* and *s* levels); (3) electron–ion recombination: $B_2^+ + e \rightarrow B(nd) + e$; (4) excitation transfer in inelastic collisions: $A^* + B \rightarrow B(nd) + A$; (5) step-by-step excitation of Xe(5*d*): $Xe(6s,\ 6s') + e \rightarrow Xe(5d) + e$.

The most common hypothesis is the second of these processes, namely level population through dissociative recombination of heteronuclear ionic molecules with electrons (Ohwa *et al.*, 1989; Shon *et al.*, 1993*b*; Shon & Kushner, 1994). Heteronuclear ionic molecules are effectively destroyed in the collision process, so their recombination at the relatively low density of electrons in nuclear-induced plasma cannot make a significant contribution to the formation of excited atoms (Mel'nikov *et al.*, 2008). Apruzese *et al.*(2006; 2008) and Mel'nikov *et al.* (2008) considered electron-ion recombination of B_2^+ ions (mechanism 3) to be the main channel for population of the *nd* levels. In most kinetic models, dissociative recombination of B_2^+ molecular ions is considered to be the channel for losses to populate the lower laser *p* levels and $(n + 1)s$ levels.

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Recharging of molecular ions on admixtures competes with dissociative electron-ion recombination. Atoms of Kr have high rate constants for charge exchange on Ar_2^+ ions: 7.5×10^{-10} (Collins & Lee, 1979), 6×10^{-10} (Johnsen *et al.*, 1978), and 5.3×10^{-10} cm^3s^{-1} (Shul *et al.*, 1987). For Xe atoms, the rate constants for charge exchange on Ar_2^+ range from 2.3×10^{-10} cm^3s^{-1} (Shul *et al.*, 1987) to 12.5×10^{-10} cm^3s^{-1} (Collins & Lee, 1979).

The rate constant for the charge-exchange process

$$Kr_2^+ + Xe \rightarrow Xe^+ + 2Kr$$

is $k \approx 2 \times 10^{-10}$ cm^3s^{-1} (Kebarle *et al.*, 1967). From Eq. (1) at $k = 2 \times 10^{-10}$ cm^3s^{-1}, $\beta = 10^{-6}$ cm^3s^{-1} and a gas ionization rate $S = 10^{19}$ $cm^{-3}s^{-1}$ we obtain the impurity pressure at which the transition intensity in the case of the recombination mechanism for level population is halved: $P = 1.6 \times 10^{16}$ $cm^{-3} \approx 60$ Pa.

Abramov *et al.* (2006) and (Gorbunov *et al.*, 2004) measured luminescence spectra of inert gases and their mixtures under pumping by ^{235}U fission fragments; the pumping power was 40 W cm^{-3}, which corresponds to a gas ionization rate of 10^{19} $cm^{-3}s^{-1}$. Table 2 shows the intensity values for the 4*p*-4*s* transitions of Ar atoms in Ar and in Ar mixtures with Xe and Kr based on data from Abramov *et al.* (2006). As can be seen from the table, on the addition of 1 kPa of Kr, the line intensity decreases 1.5-fold, and on the addition of Xe, it is halved. With a lifetime of the 4*p* levels of Ar atoms equal to $\tau \approx 30$ ns, this corresponds to quenching of 4*p* levels of argon by Kr or Xe atoms with a quenching rate constant of about 10^{-10} cm^3s^{-1}. In the case of a recombination mechanism for 4*p*-level population, Ar_2^+ ion charge exchange on additives should lead to a 50- to 100-fold decrease in line intensity. Similar conclusions may be drawn in relation to the 5*p*-5*s* transitions of Kr. The intensities of the Kr lines are given in Table 3, which is also based on data from Abramov *et al.* (2006).

The total intensity of lines of the 4*p*-4*s* transitions of the Ar atom in the spectral range 696.5–842.5 nm decreases only 1.5-fold on addition of 200 Pa of NF_3 to 0.45 kPa of Ar (Gorbunov *et al.*, 2004), confirming that recombination of molecular ions with electrons is not the basic mechanism for population of the 4*p* levels of Ar.

Thus, dissociative recombination of molecular ions of Ar and Kr are not the basic processes responsible for population of the lower *p* levels of lasers on *d*-*p* transitions of atoms. As the *p* levels are populated intensively as a result of *d*-*p* transitions, it appears that similar conclusions may be drawn in relation to the *nd* levels of Ar and Kr. The absence of any temporary delay of luminescence on excitation of Xe by a nanosecond electron beam proves the absence of a channel for population of the $5d[3/2]_1$ level of Xe via formation of the Xe_2^+ molecular ion (Denezhkin & D'yachenko, 2009).

The results obtained are consistent with the data of Barrios *et al.* (1992) and Ramos *et al.* (1995) concerning the predominant formation of Ar, Kr, and Ne atoms in the $(n + 1)s$ or ground (np) states upon dissociative recombination of Ar_2^+, Kr_2^+, and Ne_2^+ ions. Populating the *nd* levels of Ar, Kr, and Xe under pumping by hard ionizers apparently occurs mainly as a result of direct excitation by secondary electrons and excitation transfer from atoms of the buffer gases as well.

Different mechanisms of level population can be engaged depending on experimental conditions. When pumped by nanosecond electron beam Ar-Xe mixture lased pulse with delay of tens of nanoseconds, while the duration of lasing pulse at atmospheric pressure reached several microseconds (Peters *et al.*, 1989; Scrobol *et al.*, 2009). Investigation of luminescence at $\lambda = 1.73$ microns of He-Ar-Xe mixtures excited by an electron beam of 2 ns duration and 500 A current was reported in (Denezhkin & D'yachenko, 2013). It was shown that there were two channels of $5d[3/2]_1$ level population. Characteristic time constants of these channels depended on composition of the mixture and amounted to the order of a hundred nanoseconds for one channel and few microseconds for the second channel. These channels are well separated in He-Ar-Xe mixtures,

Table 2. *Emission power in the 4p-4s transitions of Ar atoms in pure Ar and in Ar-Kr and Ar-Xe mixtures under pumping by uranium fission fragments*

λ (nm)	Transition	Specific luminescence power ($mWcm^{-3}$)		
		Ar(45 kPa)	Ar(44.01 kPa)–Kr(0.99 kPa)	Ar(44 kPa)–Xe(1 kPa)
750.4	$4p'[1/2]_0-4s'[1/2]_1^o$	5.1	3.2	2.3
763.5	$4p[3/2]_2-4s[3/2]_2^o$	3.7	3.1	4.6
772.4	$4p'[1/2]_1-4s'[1/2]_0^o$	5.1	2.6	1.9
794.8	$4p'[3/2]_1-4s'[1/2]_0^o$	2.2	1.6	1.5
800.6	$4p[3/2]_2-4s[3/2]_2^o$	3.4	2.7	1.8
852.1	$4p'[3/2]_1-4s'[1/2]_1^o$	3.6	2.6	1.1
866.8	$4p[3/2]_1-4s'[1/2]_0^o$	1.1	0.5	—
912.3	$4p[1/2]_1-4s[3/2]_2^o$	5.1	4.7	4.7
922.4	$4p[3/2]_2-4s'[1/2]_1^o$	3.7	2.0	—
965.8	$4p[1/2]_1-4s[3/2]_1^o$	3.9	2.8	—
Total		36.9	25.8	17.9

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Table 3. *Emission power in the 5p-5s transitions of Kr atoms in Kr and Kr-Xe under pumping by uranium fission fragments*

λ (nm)	Transition	Specific luminescence power (mW cm^{-3})	
		Kr(32 kPa)	Kr(31 kPa)–Xe(1 kPa)
758.7	$5p[1/2]_0-5s[3/2]_1^o$	18	8.6
760.2	$5p[3/2]_2-5s[3/2]_2^o$		
768.5	$5p'[1/2]_0-5s'[1/2]_1^o$	5.5	2.3
785.5	$5p'[1/2]_1-5s'[1/2]_0^o$	2.8	1.2
805.9	$5p'[3/2]_1-5s'[1/2]_0^o$	1.8	1.5
811.3	$5p[5/2]_3-5s[3/2]_2^o$	9.9	7.8
819.0	$5p[3/2]_1-5s[3/2]_1^o$	11	4.5
850.9	$5p'[3/2]_1-5s'[1/2]_1^o$	3.5	1.5
877.7	$5p[5/2]_2-5s[3/2]_1^o$	9.3	4.7
Total		61.8	32.1

which results in significant fraction (70–80%) of photons emitted during the first 300 ns. Ar-Xe mixtures exhibited a single population time constant of about one microsecond with delay of radiation pulse of tens of nanoseconds.

Since the fraction of xenon in optimal mixtures (He)-Ar-Xe is small, the main pumping occurs with the transfer of excitation from the argon to xenon atoms (Fig. 3). In our opinion, the long lasted lasing and luminescence emission continued after the pump pulse stems from the stepwise excitation processes:

$$\mathrm{Xe}(6s, 6s') + e \rightarrow \mathrm{Xe}(5d) + e;$$
$$\mathrm{Xe}(6s, 6s') + e \rightarrow \mathrm{Xe}(6p) + e;$$
$$\mathrm{Xe}(6p) + e \rightarrow \mathrm{Xe}(5d) + e.$$

The formation of xenon atoms in the (6*s*, 6*s*') states can be the result of several processes: direct excitation from the ground level during the pump pulse, excitation transfer from argon atoms, cascaded transitions from the upper levels, the recombination of Xe_2^+ ions with electrons, and possibly other mechanisms. The microsecond fraction of the radiation is less pronounced in He-Ar-Xe then in Ar-Xe mixtures because electrons are faster thermalized in He then in Ar. In pure xenon this channel is negligible due to the high rate of formation of Xe_2^* excimers generated from Xe (6*s*, 6*s*').

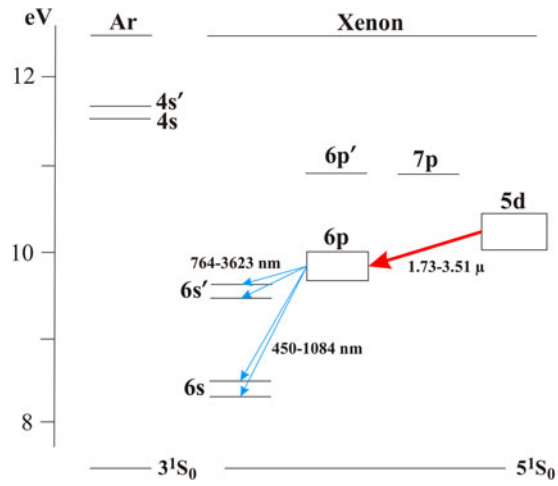

Fig. 3. (Color online) Scheme of some levels of xenon and argon. The arrows indicate the direction of the laser transitions (red) and 6*p*-6*s* transitions (blue), analogous to 4*p*-4*s* transitions in argon and 5*p*-5*s* transitions in krypton.

It is worth mentioning that in the reported computational models of the Ar-Xe laser (Klopovskii *et al.*, 1989) the major channels population of $5d[3/2]_1$ level of xenon are stepwise excitation via 6s metastable level, and the transfer of excitation from the buffer gas.

5. MERCURY

It was shown by Batyrbekov *et al.* (1987; 1989) that dissociative recombination of molecular ions is the basic process populating the 7^3S_1 level of the Hg atom under ionizing pumping, possibly in cascade transitions from the 7^3P levels. The following results point to the recombination mechanism of population: (1) The intensity of the Hg triplet lines in He-Xe-Hg mixtures was practically independent of Hg pressure in the range 0.1–40 Pa in a radioisotope installation and 0.1–13 kPa in an in-core installation, which excludes the possibility of direct excitation of the 7^3S_1 level from the ground state by electron impact. (2) In a He-Xe-Hg-D_2 mixture, on increasing the partial pressure of D_2 to 13 kPa, the intensity of the resonance line of Hg (253.7 nm) decreased more than 300-fold, whereas the intensity of triplet lines decreased only 2.5-fold. This indicates that population of the HgI levels cannot be explained by step-by-step excitation through the 6^3P_1 level. (3) On addition of an electronegative impurity (13 Pa of O_2) to a ^{3}He-Hg mixture, the intensity of triplet lines decreased 500-fold (Khasenov & Smirnova, 2008).

In the work of Bochkov *et al.* (1992) and Rhoades and Verdeyen (1992), in which laser action was obtained on the 546.1 nm line of the Hg atom, recombination of Hg molecular ions with electrons was also considered as a basic mechanism for population of the 7^3S_1 level. The 7^1S_0 level is also populated; notably, although the 407.7 nm line is comparatively weak under our conditions, the probability of the IR transition from this level (λ = 1014 nm) is approximately seven times higher than that for the 7^1S_0-6^3P_1 transition (Benck *et al.*, 1989). From the lifetime of the 7^1S_0 level, the probability of the 7^1S_0-6^3P_1 transition and the intensity of the 407.7 nm line, it is possible to determine the ratio between the number of photons radiated from the 7^1S_0 level and the number of photons radiated from the 7^3S_1 level (Table 4). Line intensities of mercury were measured at 22 GBq total activities of nuclides sources in the radioisotope installation and at thermal neutron flux of 10^{13} n/cm^{2}s on the reactor facility. Due to the proximity of lines 407.7 nm and 404.7 nm and the known values of the transition probabilities of 7^3S_1

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Table 4. *Intensity of transitions (relative units) from 7^3S_1 and 7^1S_0 levels of Hg atoms under excitation by products of $^3He(n,p)^3H$ reaction in the core of a nuclear reactor and by α particles from ^{210}Po*

Source	Nuclear reactor		^{210}Po		
Mixture	^{3}He(80 kPa)–Hg(130 Pa)	^{3}He(40 kPa)–Xe(40 kPa)–Hg(130 Pa)	Ar(100 kPa)–Hg(0.2 Pa)	Kr(100 kPa)–Hg(0.2 Pa)	Xe(100 kPa)–Hg (0.2 Pa)
$I(7^1S_0)$	160	250	28	75	8.0
$I(7^3S_1)$	900	3540	230	440	300
$I(7^1S_0)/I(7^3S_1)$	0.18	0.07	0.12	0.17	0.027

level it was not necessary to calibrate the spectral sensitivity of the apparatus. It appears that Xe (and, to a lesser extent Ar) quenches the 7^1S_0 level, with level deactivation occurring through the process

$$\mathrm{Hg}(7^1S_0) + 2\mathrm{Xe} \rightarrow \mathrm{HgXe}* + \mathrm{Xe}.$$

Estimation of the corresponding rate constant from the results shown in Table 4 gives a value of about 3×10^{-31} $\mathrm{cm}^6\mathrm{s}^{-1}$. Khasenov and Smirnova (2008) showed the possibility of population of the 7^3S_1 level of the Hg atom by recombination of Hg atomic ions and negative oxygen ions:

$$\mathrm{Hg}^+ + \mathrm{O}^- + \mathrm{M} \rightarrow \mathrm{Hg}* + \mathrm{O} + \mathrm{M}.$$

6. CONCLUSIONS

It has been shown that dissociative recombination of molecular ions with electrons is not the dominant process for population of the $(n + 1)p$ levels of Ne, Kr, and Ar under ionizing pumping. As these levels are intensively populated in transitions from the corresponding *nd* levels, similar conclusions are likely to hold in relation to population of the *d* levels. It is assumed that direct excitation of atoms by secondary electrons and excitation transfers from atoms of buffer gas are the most probable channels for *d*-level population. This conclusion is important with regard to the development of optical detectors for neutron radiation. In the case of a recombination mechanism for level population, the intensity of optical radiation under weak ionization is more sensitive to the presence of impurities in the gas.

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