NUMERICAL MODELING OF THE PLASMOCHEMICAL PROCESSES IN THE Xe-Sr-MIXTURE PUMPED WITH A HARD IONIZER

A.V. Karelin, R.V. Shirokov, and S.I. Yakovlenko

Institute of General Physics, Russian Academy of Sciences, Moscow
Received June 17, 1997

Nonstationary self-consistent kinetic models of Xe–Sr *laser on* $6s^2S \to 5p^2P_{3/2}$ (λ = 430.5 nm) *transition in strontium ion and of* Xe–Sr *lamp pumped with a hard ionizer have been developed. Optimum conditions of lasing providing the laser efficiency relative to the pump of up to 3% have been realized. The efficiency of a source of spontaneous line radiation in the range* 400–500 nm *was shown to reach* 30%.

INTRODUCTION

The vapor of metals from the second group are considered to be promising active media (hereafter AM) for developing efficient sources of spontaneous and stimulated emission in a wide spectral range.[1–5] This provided the development of a number of now widely used devices, such as Cu- and Cd-vapor lasers, Hg lamps. However, the scientists continue research and development of the AM-emitters used in a lot of applications. In particular, Xe–Sr mixture, considered in this paper, can be promising for the development of efficient sources of laser radiation (at 430.5 nm wavelength) as well as of spontaneous line radiation in the blue-green spectral region.

Experimental and theoretical investigations of lasing based on $6s^2S \to 5p^2P_{3/2}$ transition (λ = 430.5 nm) in a single strontium ion have been being carried out for many years.[6–8] Different active mixtures and methods of pumping the upper laser level have been considered:

a) He(Ne)–Sr(–H_2) mixture with the recombination pumping.[4,7–11] In this case, atoms and ions of He or Ne cool the plasma electrons and the upper $6s^2S$ level of ion Sr^+ is populated due to triple e–e recombination of Sr^{++} ions resulting from the recharge of He^+(Ne^+) ions on Sr atoms. Addition of small amounts of hydrogen to He–Sr (Ne–Sr) mixture (only if it does not decrease the Sr^{++} ions concentration) leads to a decrease in the electron temperature and to lasing during a long after-glow[7];

b) Xe–Sr mixture with the upper lasing level pumped using Xe^+ ions recharging on strontium atoms.[12,13] According to Ref. 13, the laser generation had not been obtained because of a low discharging cross section of Xe^+ on Sr, and high temperature of electrons which can not be efficiently cooled by heavy particles of the plasma;

c) theoretical investigations of He–Xe–Sr mixture with the upper level pumped due to the recharge of Xe^+ on Sr and the recombination one were considered in Ref. 10. In this model, free electrons cooled by helium, provide an efficient de-excitation of the lower laser level. Two mechanisms of the upper level pumping can result in an intense lasing (with the laser efficiency exceeding 1%).

Results of the optimization of some of the above mentioned media pumped with a hard ionizer are reviewed in Ref. 5.

An atom and ion of strontium (mixed with xenon) are promising for the development of spontaneous line radiation source in the range 400–500 nm (see Grotrian diagrams in Fig. 1). This interval comprises the wavelengths of four ion and five atom transitions in strontium. Thus, one can hope to develop a highly-efficient laser radiation source.

KINETIC MODEL

This model allows for the following constituents of the active medium (Fig. 1): atom of strontium Sr in the ground state $5s^2\ ^1S_0$, as well as its excited states: Sr*: $5s5p^3P_{0,1,2}$, $5s6p^3P$, $5s5p^1P_1$, $5s6p^1P_1$, $5s4d^3D$, $5s5d^3D_{1,2,3}$, $5s4d^1D_2$, $5s5d^1D_2$, $5s6s^3S_1$, $5s6s^1S_0$, the strontium ion in the ground state $5s^2S_{1/2}$, seven excited states of the strontium ion $4d^2D$, $5p^2P_{1/2,3/2}$, $5d^2D_{3/2,5/2}$, $6s^2S$, $6p^2P$, twofold strontium ion Sr_2^+, molecular strontium ion Sr^+, xenon atom Xe and two its excited states: the state Xe* including 6s levels, and state Xe** including 6p and 5d levels, the xenon dimers Xe_2^* and Xe_2^{**}, the xenon ion Xe^+, molecular ion Xe_2^+, the hydrogen molecules H_2, H_2^*, the hydrogen atom H, hydrogen ion H^+, and molecular ions H_2^+ and H_3^+. In addition, spontaneous radiation in the range 400–500 nm (Fig. 1) at the following wavelengths: λ = 430.5 and λ = 416.2 nm (attributed to transitions $6s^2S \to 5p^2P_{3/2,1/2}$ in the ion), resonance lines of the ion at λ = 407.8 and 421.6 nm (transitions $5p^2P_{3/2,1/2} \to 5s^2S_{1/2}$), the atomic resonance line λ = 460.7 nm (transition $5p^1P_1 \to 5s^1S_0$), λ = 496.8 nm, 487.2 nm, and 483.2 nm (transitions $5d^3D_1 \to 5p^3P_{2,1,0}$

0235-6880/97/11 827-05 $02.00

in the atom), and λ = 496.2 nm (transition $5d^3D_3 \to 5p^3P_2$ in the atom) have been considered in the model.

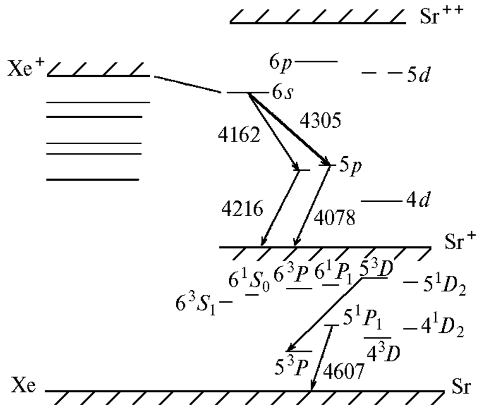

FIG. 1. System of energy levels of Xe–Sr *mixture.*

The 100 keV pulsed electron beam (duration of pulses 100–300 ns) was considered as the source for laser pumping. For a lamp, nuclear pumping with the peak neutron flux density $\approx 10^{15}$ n-n/(s·cm^2) and duration t_0 = 1 ms was considered in this model. The "bell" shaped pulses were used in both cases[9]

$$\nu(t) = \nu_0(\mu t)/[7\ (1 + e^{\mu t - 10})], \qquad (1)$$

where ν_0 is the maximum ionization frequency; $\mu = 12/t_0$, t_0 is the pulse duration measured at the level $\nu = 0.1\ \nu_0$. The ionization frequency of the xenon atoms by an electron beam is related to the density of the electron beam current j by the expression

$$\nu = 135\ j, \qquad (2)$$

where j is in A/cm^2, and the ionization frequency is in s^{-1}. Note that the change of the electron beam or a nuclear pumping for different other hard ionizers, providing the same ionization frequency, does not change the principal results.[5]

The rate coefficients for principal plasmochemical processes were taken from Ref. 10. Recharge of the excited states $5d^2D_{3/2,5/2}$ of strontium ions on xenon atoms has been considered along with the recharge of xenon ions on the strontium atoms. The direct and inverse processes were related by the principle of detailed equilibrium. We took into account time dependence of the escape coefficients Θ describing the radiation reabsorption due to transitions to the ground and metastable states of the strontium atom and ion as well as the "parasitic" photoionization of Xe** atoms and Xe_2^{**} molecules with laser radiation the cross section of this process being $1.5\cdot10^{-17}$ cm^2.

Only a limited number of reactions to account for the influence of hydrogen on the plasma kinetics, e.g.:

cooling of electrons on molecules and ions of hydrogen at a rate of $1.8\cdot10^{-7}\sqrt{T_e}$;

ionization of hydrogen atoms and molecules by pumping pulses;

triple collision-radiative recombination of the hydrogen ions H^+;

conversion of H atoms and ions H^+ into the molecular ions H_2^+, H_3^+;

dissociative recombination of H_2^+, H_3^+ ions;

recharge of H^+, H_2^+, H_3^+ ions on xenon atoms leading to formation of the excited $6p^2P$ states of the strontium ions at a rate of 10^{-9} cm^3/s.

The unit volume concentration of strontium atoms was estimated using the data on saturated vapor pressure (see the handbook[14]). Its temperature dependence in the range 980–1300 K can be described (accuracy 9%) by the formula

$$N = [(1.54\cdot10^{26})/T]\ e^{-16458/T}, \qquad (3)$$

where T is in K, and N is in cm^{-3}.

The ions Xe^+ and excited atoms of xenon Xe*, Xe** are formed in AM under the action of a hard ionizer. The pumping of the state $6s^2S$ of Sr^+ is performed due to recharge of xenon atoms on those of strontium (see Fig. 1). The inverse population in the transition levels $6s^2S \to 5p^2P_{3/2}$ is caused by an effective de-excitation of the lower state $5p^2P_{3/2}$ by electrons. Relaxation of the ground state of strontium ions Sr^+ is realized, mainly, through its conversion into the molecular ion Sr_2^+ and the dissociative recombination of this ion leading to the population of the higher excited states of strontium atom. The recombination flux is distributed among the levels $6p^3P$, $6p^1P_1$, $5d^3D_{1,2,3}$, $5d^1D_2$, $6s^3S_1$, $6s^1S_0$ of strontium atom depending on the statistical weight of a level. The tables and equations from Ref. 15 were used for calculating the rates of the excited states mixing due to electrons as well as the unknown values of the probabilities of the radiative transitions of strontium atom.

In calculations we used the "PLAZER" software package. The following parameters have been specified: the initial concentration of the AM constituents, the initial gas temperature T_0, effective lifetime of the photon staying in the resonator, pumping pulse shape and the peak frequency of the medium ionization depending on a pumping mode. The calculated characteristics are: non-stationary concentration of electrons, atoms, molecules and ions; temperatures of electrons and heavy plasma particles; power P_λ of every separate line of a lamp. The development of laser radiation power P was calculated using the zero-dimension approximation.

Xe–Sr LASER

The AM of a laser emitting at the wavelength 430.5 nm was suggested to have a cylindrical shape with the radius R_a = 1 cm and length L_a = 100 cm.

According to numerical modeling, the increase of the pumping pulse duration t_0 leads to improvement of the output parameters of the laser. The value $t_0 = 250$ ns was used in computations since its further increase did not influence the resulting efficiency, η, of the laser.

Addition of any amount of hydrogen to the AM was shown in capable of improving the output laser parameters. This is explained by the fact that the electron concentration decreases in spite of a decrease in the electron temperature, because their cooling favors the increase of the rates of recombination processes (especially of the dissociative recombination of molecular ions). That is why, the rate of the electron impact de-excitation of the lower laser level, playing a main role in the lower level recharge, decreases $\sim N_e/\sqrt{T_e}$. That leads to an increase in the lower level population that reduces the inverse population of the lasing transition levels.

The following results have been achieved by optimizing parameters of the AM and the resonator at a fixed duration of the electron beam: pressure of xenon is 60 Torr ([Xe] = $2.1\cdot10^{18}$ cm^{-3}), the initial temperature of AM $T_0 = 920$°C, the reflection coefficient of the resonator mirrors $r = 0.95$ at the energy contribution into a unit volume of the medium $W = 1.7$ mJ/cm^3 (current density $j = 11$ A/cm^2). The lasing process starts 0.1 ms after the pumping pulse start and its duration is 1.3 μs. The maximum radiation power is 128 W/cm^3, the energy efficiency per pulse reaches 3.8%, $N_e = 5.5\cdot10^{14}$ cm^{-3}, $T_e = 0.31$ eV. Minimum temperature of the AM allowing the lasing is 700°C at the xenon concentration [Xe] = $8\cdot10^{17}$ cm^{-3}.

The dependence of the energy generated per pulse E_v and the efficiency η on the initial temperature of the medium T_0 is shown in Fig. 2. The optimum conditions exist at $T_0 = 920$°C ([Sr]=$1.3\cdot10^{17}$ cm^{-3}). This can be explained as follows: the increase in strontium concentration results in an increase in the conversion rate of ions Sr^{+*} and strontium atoms Sr followed by the formation of molecular ions Sr_2^+. The latter ones participate in the dissociative recombination resulting in a decrease of the electron concentration N_e and in an increase of its temperature T_e, that, in its turn, reduces the inversion in the population of the laser levels (i.e. it decreases the de-excitation of the lower level $5p^2P_{3/2}$). Recharge of the active ions of strontium on its atoms also favors a decrease in the inversion.

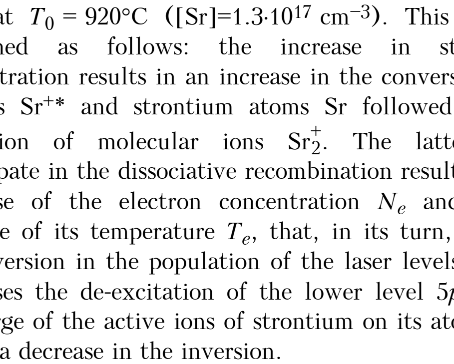

FIG. 2. The dependences of η *(1) and* E_v *(2) on the* Xe–Sr *mixture initial temperature* T_0 *at* [Xe] = $= 2.1\cdot10^8$ cm^{-3}, $j = 11$ A/cm^2, $r = 0.95$.

An increase of the energy deposition into an AM up to 2.5 mJ/cm^3 is accompanied by the efficiency growth up to η = 3.9%. Further increase of W does not influence the efficiency since the increase of the xenon ions concentration is compensated by the electron de-excitation of the upper level. The threshold of the radiation lasing is $W_{th} \approx 0.5$ mJ/cm^3.

The reflection coefficient of the output mirror r does not considerably affect the above mentioned value of E_v. Optimum value of r is 0.95. Minimum value of r allowing the lasing is ≈10%.

SOURCE OF THE LINE SPONTANEOUS RADIATION

Geometrical parameters of the AM used in calculations of Sr-lamp were kept the same. We have optimized parameters of a nuclear pumped AM with the above mentioned parameters. In particular, the optimal parameters are found to be as follows. The xenon pressure 42 Torr ([Xe]=$1.5\cdot10^{18}$ cm^{-3}) and temperature of the AM $T_0 = 850$°C. These conditions provide the following output parameters: total power in a pulse peak over the spectral lines of interest is 6.3 W/cm^3, efficiency η□= 30%, $T_e = 0.34$ eV, $N_e = 1.6\cdot10^{13}$ cm^{-3}, the peak power of the energy contribution to a unit volume is 21 W/cm^3. It is the atomic resonance line 4607 Å that makes the main contribution to the emission power.

The temperature dependence of power of a line group is shown in Fig. 3. The behavior of resonance lines of ion is attributed to the competition between the increase in pumping power of $5p^2P_{3/2,1/2}$ levels and their conversion into the Sr_2^+ ion. The population of the level $6s^2S_{1/2}$ is affected both by the conversion and by the Penning reaction with the atom itself. This explains the observations of maxima of 4305 Å, 4162 Å lines at lower temperatures, unlikely to the maximum in ion resonance lines. The existence of the parasite relaxation channel $5p^2P_{3/2,1/2} \to 4d^2D \to 5s^2S$ results in a decrease of the power of 4078, 4216 Å lines in comparison with that of 4305, 4162 Å at a relatively low temperatures. The increase in the strontium molecular ion Sr_2^+ concentration along with the gas temperature increase T_0 leads to a decrease in the electron concentration in plasma.

The mixing rates of the strontium atom levels excited with electrons can influence the electron temperature T_e. The increase of concentration of the ground state strontium atoms provides for the excitation of $5s^2\,{}^1S_0 \to 5s5p^3P_{2,1,0}$ levels (accompanied by the electron cooling) to dominate over the inverse process of de-excitation. This leads to a decrease in T_e while T_0 is increasing. In spite of the strong radiation reabsorption, the transition $5s5p^1P_1 \to 5s^2\,{}^1S_0$ is most powerful radiation channel of relaxation in strontium atom. This fact explains high intensity of the atomic resonance line P_{4607}. Mixing of the excited states by electrons strongly affects the value of P_{4607} (it can both increase and decrease it). In particular, the absence of the resonance level mixing by electrons with other excited levels of the atom under optimal conditions can provide an 1.5 times increase of the resulting value of P_{4607}. This phenomenon is caused by domination of the inverse de-excitation over the electron excitation of highly populated states $5s5p^3P_{2,1,0}$ and $5s4d^3D_{3,2,1}$. Such processes result in some cooling of the plasma electrons (the so-called radiation cooling). The decrease in P_{4607} at $T_0 > 850°C$ (see Fig. 3) is attributed to an increase of the radiation reabsorption (the coefficient $\Theta \sim 1/\sqrt{[\mathrm{Sr}]}$) at quite an effective de-excitation.

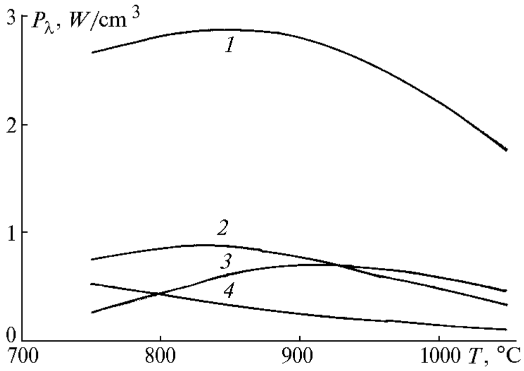

FIG. 3. Dependence of the intensity P_λ of lines on the initial temperature of AM at [Xe] = 1.5·10^{18} cm^{-3}*: P_{4607} (1), $P_{4305} + P_{4162}$ (2), $P_{4078} + P_{4216}$ (3) and $P_{4968} + P_{4872} + P_{4832} + P_{4962}$ (4).*

The intensity of the majority of lines increases with increasing concentration (see Fig. 4) of xenon. However, the optimal efficiency of the source is achieved at [Xe] = 1.5·10^{18} cm^{-3}. It is explained by the increase of the energy deposition to AM. Weakening of the ion lines with respect to the atomic ones at high pressure of xenon is caused by the conversion of strontium ions into the molecular ones.

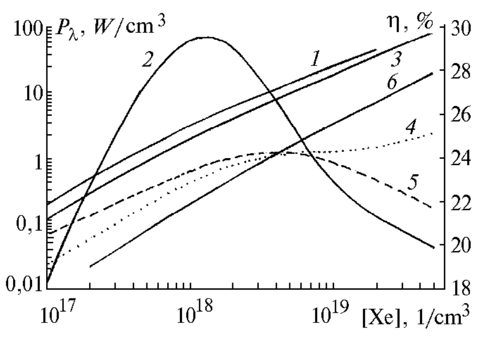

FIG. 4. Dependences of the source total power P and efficiency η as well as of power of some lines P_λ on the xenon concentration in AM at T_0 = 850°C: P (1), μ (2), P_{4607} (3), P_{4305} + P_{4162} (4), P_{4078} + P_{4216} (5), and $P_{4968} + P_{4872} + P_{4832} + P_{4962}$ (6).

CONCLUSION

The nonstationary kinetic model of a Xe–Sr laser pumped with a high energy electronic beam lasing on the transition $6s^2S \rightarrow 5p^2P_{3/2}$ of the single strontium ion at the wavelength 430.5 nm has been developed and studied by the authors. The results obtained prove the possibility of an effective lasing on this transition.

Optimum conditions for lasing (L_a = 100 cm, R_a =1 cm) are the following: duration of the pumping pulse t_0 = 250 ns; the initial temperature of the AM T_0 = 920°C ([Sr]=1.3·10^{17} cm^{-3}); xenon concentration [Xe] = 2.1·10^{18} cm^{-3} (partial pressure of the xenon p_{Xe} = 60 Torr); the energy contribution to the unit volume of the AM W = 1.7 mJ/cm^3 (electron current density j = 11 A/cm^2, frequency of xenon ionization ν_0 = 1500 s^{-1}); reflection coefficient of the output mirror of the resonator r = 0.95. Such conditions provide for a specific output power per unit volume E_v = 6.6·10^{-5} J/cm^3 (total energy of radiation E = 0.02 J), the efficiency of the AM η = 3.9% and the lasing duration τ = 1.3 μs.

Modeling of the source of spontaneous line radiation in Xe–Sr mixture in the range 400–500 nm with a pulsed nuclear pumping allows one to predict the possibility of developing an effective lamp with the efficiency reaching 30%. For this case, the optimum parameters of Xe–Sr mixture were found to be the following:

the initial temperature of strontium T_0 = 850°C ([Sr]=6·10^{16} cm^{-3});

concentration of xenon [Xe] = 1.5·10^{18} cm^{-3} (corresponding pressure p_{Xe} = 42 Torr).

Mixtures of other metal vapor with inert gases are expected to be promising for development of various radiation sources emitting in other spectral ranges.

REFERENCES

1. A.M. Prokhorov, ed., *Laser Handbook* (Sov. radio, Moscow, 1978), Vol. 1, 504 pp.
2. Ch.K. Rhodes, ed., *Excimer Lasers* (Springer-Verlag, Berlin, 1979), 248 pp.
3. A.N. Soldatov and V.I. Solomonov, *The Metal Vapor Gas Discharge Lasers* (Nauka, Novosibirsk, 1985), 152 pp.
4. I.G. Ivanov, E.L. Latush, and M.F. Sem, *The Metal Vapor Ion Lasers* (Energoatomizdat, Moscow, 1990), 256 pp.
5. A.V. Karelin and S.I. Yakovlenko, Kvant. Elektron. **20**, No. 7, 631–651 (1986).
6. E.L. Latush and M.F. Sem, Kvant. Elektron. **3(15)**, No. 1, 66–71 (1973).
7. V.V. Zhukov, V.S. Kucherov, E.L. Latush, and M.F. Sem, Kvant. Elektron. **4**, No. 6, 1257–1267 (1977).
8. S.M. Babenko and S.I. Yakovlenko, *"Analysis of the kinetics of* He–Sr *laser processes*,B Preprint No. 3192, Kurchatov Institute of Atomic Energy, Moscow (1979), 40 pp.

9. V.I. Derzhiev, V.F. Tarasenko, S.I. Yakovlenko, and A.M. Yancharina, *Proceedings of the General Physics Institute* (Nauka, Moscow, 1989), Vol. 21, pp. 5–43.
10. V.I. Derzhiev A.G. Zhidkov, A.V. Karelin, and S.I. Yakovlenko, "*Kinetic models of high pressure* Sr-*inert gases (helium, xenon, neon) mixture lasers pumped by a hard ionizer,*" Preprint No. 90, Institute of General Physics of the Academy of Sciences of the USSR, Moscow (1988), 60 pp.
11. S.N. Atamas', E.L. Latush, and M.F. Sem, Sov. J. of Laser Research. **15**. No. 1, 66–68 (1994).
12. K.B. Butterfield, D.S. Gerstenberger, T. Shay, W.L. Little, and G.J. Collins, J. Appl. Phys. **49**, No. 6, 3088–3096 (1978).
13. V.V. Zhukov, E.L. Latush, and M.F. Sem. Zh. Prikl. Spektrosk. **32**, No. 4, 738–740 (1980).
14. A.P. Babichev, N.A. Babushkina, A.M. Bratkovskii, et al., eds., *Physical Values (Handbook)* (Energoatomizdat, Moscow, 1991), 1232 pp.
15. L.A. Vainshtein, I.I. Sobel'man, and E.A. Yukov, *Cross Sections of Exciting of Atoms and Ions by Electron* (Nauka, Moscow, 1973), 144 pp.