OPTIMAL CONDITIONS FOR LASING IN A NUCLEAR-PUMPED He–Cd–CCl_4 UV–LASER

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We discuss here numerical modeling of active media of a nuclear-pumped laser in the mixture of He–Cd–CCl_4 *(*$\lambda = 325.0$ nm*) using a detailed nonstationary kinetic model. The mechanisms have been studied of the effect of* CCl_4 *admixture on the relaxation processes in* He–Cd *plasma as well as on the lasing process. The influence that may exhibit addition of nitrogen on laser emission characteristics has been studied as well. It is shown that among the main factors that may prevent lasing at the transition with the wavelength of* 325.0 nm *there are parasitic absorption of the laser emission by cadmium atoms in metastable states and capture of the pump by minor, uncontrolled, admixtures as water vapor, nitrogen, and so on. The modeling made enabled us to deduce optimal conditions for* Cd*-lasers operation when pumped with a pulsed reactor of BARS–6 type.*

INTRODUCTION

Among lasers that operate on vapor of the II-nd group metals pumped with a hard ionizing radiation the laser in He–Cd mixture is one of the most interesting as it has the lowest lasing threshold while the highest lasing efficiency. In addition, the lasers of this type deliver quite a short-wave radiation at 325.0, 441.6, 533.7, and 537.8 nm (transitions in a Cd ion) wavelengths.[1–10] Besides, the metal-vapor laser of this type attracts attention of the laser researchers by a possibility of creating quite an efficient laser with a nuclear pump of the lasing transitions in the UV region, at 325.0 nm wavelength (see Fig. 1). It is just that possibility of obtaining a quasi-stationary lasing on the cadmium ion transition $4d^9 5s^2\,{}^2D_{3/2} \rightarrow 4d^{10}5p\,{}^2P_{1/2}$ at the wavelength of 325.0 nm that makes the subject of the study presented in this paper. Earlier, in Refs. 1, 5, and 7, it was noted, based on the results of numerical simulations, that the threshold density of the neutron flux for this laser operation is $\Phi_{thr} \approx 2\cdot 10^{16}$ neutrons/($cm^2\cdot s$) at a pressure of the He–Cd mixture of 2 to 3 atm and temperature from 360 to 400°C. It was also shown in these papers that the maximum lasing efficiency of that laser can not exceed 0.2%. The high lasing threshold and relatively low lasing efficiency have been assigned to de-excitation of the upper lasing level due to collisions with atoms of the buffer gas – He, as well as by a strong parasitic absorption of the laser emission. The rates of the latter processes have been assessed from a comparison made between the results of theoretical modeling and experiments on the electron-beam pumping.[1,4,5] However, the nature of that parasitic absorption has not been so far identified. In Ref. 6 the authors assumed it to be due to the absorption by cadmium atoms on the transition yielding the emission at 325.25 nm wavelength. Later on, in Ref. 7 it was shown that the absorption cross-section for this transition, as well as the population its lower level may reach quite high values. New results that have been reported in Refs. 8 to 10 on the laser emission obtained at the transition with the wavelength of 325.0 nm when pumped by a microsecond-duration electron beam that models the leading edge of a pump pulse from a nuclear reactor made us to revise, in order to refine, the kinetic model of He–Cd–CCl_4 laser earlier discussed in Refs. 1, 4, 5, and 7. In so doing we placed special value on the parasitic absorption. The main reason for returning to the kinetic model was the circumstance experimentally observed as the break of lasing at 325.0 nm wavelength during the pump electron beam pulse.[8–10] . At the same time after adding 1 Torr of CCl_4 to the gas mixture no such a break occurred and the lasing terminated simultaneously with the pump pulse termination.[10] Thus it was experimentally demonstrated that it is possible to provide for a better lasing by adding some quantity of CCl_4 into the gas mixture of the laser active medium.

In this paper we discuss a theoretical study of the effect that is produced by the parasitic absorption and that from the CCl_4 admixture on the lasing properties of the active medium in the He–Cd mixture at the transition with the wavelength of 325.0 nm when pumped with a hard ionizer excited in different modes.

1. KINETIC MODEL OF A He–Cd–CCl_4 LASER PUMPED WITH A HARD IONIZER

When describing the kinetics of the He–Cd plasma we took into account the following components: helium (He) and cadmium (Cd) atoms; He^+ and Cd^+

($4d^{10}5s\ ^2S_{1/2}$) ions in the ground state; molecular ions He_2^+, He_3^+, and Cd_2^+; the excited He* and Cd($5s5p^3P_{2,1,0}$) atoms; double ionized cadmium Cd^{++}; excited molecules of He_2^*, Cd_2^*, and Cd_3^*; cadmium ions in 17 excited states: $4d^95s^2\ ^2D_{5/2}$, $4d^95s^2\ ^2D_{3/2}$, $4d^{10}5p^2\ P_{1/2}$, $5p^2P_{3/2}$, $6s^2S_{1/2}$, $6p^2P_{1/2}$, $6p^2P_{3/2}$, $5d^2D_{3/2}$, $5d^2D_{5/2}$, $6d^2D_{3/2}$, $6d^2D_{5/2}$, $4f^2F_{5/2}$, $4f^2F_{7/2}$, $6f^2F_{5/2}$, $6f^2F_{7/2}$, $6g^2G_{7/2}$, $6g^2G_{9/2}$, as well as the combined ion $HeCd^+$. Figure 1 depicts the diagram of the cadmium ion terms that are allowed for in the model and the transitions the lasing on which is being studied.

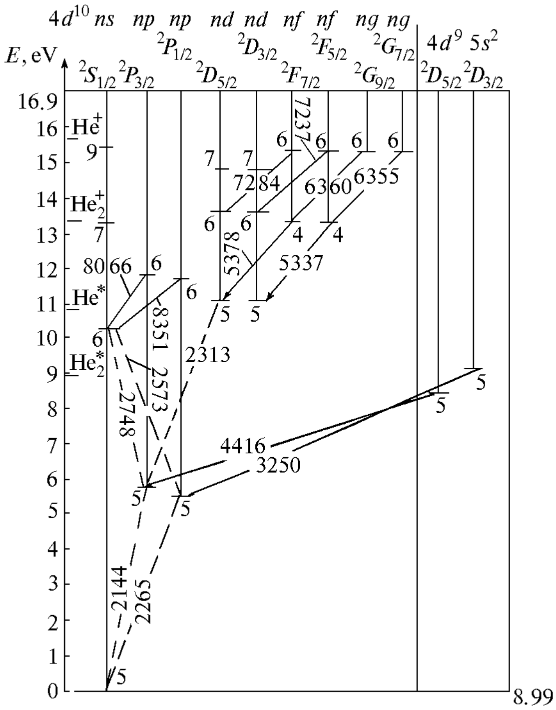

FIG. 1. Diagram of terms of the cadmium ion.

In this model we consider in a more detail the relaxation flux through the excited levels $5s5p^3P_{0,1,2}$ of the cadmium atom. The levels are being taken into account separately. Besides, we have introduced into the model a parasitic absorption of radiation with the wavelength of 325.0 nm by cadmium atoms at the transition $5p^3P_2 \to 7s^3S_1$ (325.25 nm wavelength) assuming the absorption cross-section to be $2.1\cdot10^{-17}$ cm^2. Since in the model used the dynamics of populating the metastable states of Cd atoms is an important factor we have refined, following Ref. 11, the data on the rate of the dissociative recombination of the Cd_2^+ ion.

We have also refined, when constructing the model, partial cross-sections of the charge exchange reactions for helium molecular ion on the cadmium atoms. In so doing we have lowered, by 1.5 times, the pumping of $4d^95s^2\ ^2D_{5/2,\,3/2}$ and $4d^{10}5p\ ^2P_{3/2,\,1/2}$ levels in these reactions while directing the remaining fraction off the relaxation flux towards the $4d^{10}6p\ ^2P_{3/2,\,1/2}$) levels. Total rate of the Penning reaction for metastable helium atoms He* on cadmium atoms was taken to be $8\cdot10^{-10}$ cm^3/s.[12] About 2% of the charge exchange reaction flux for helium ions on the cadmium atoms come from the upper, for green lines at 533.7 and 537.8 nm wavelengths, levels $4f^2F_{7/2,\,5/2}$ to the levels $6g^2G_{9/2,\,7/2}$. The pressure broadening coefficient for the line at 325.0 nm wavelength has been assumed to be 10^{11} (atm·s)$^{-1}$. When allowing for the reabsorption of radiation by the transitions $5p^2P_{3/2,\,1/2} \to 5s^2S_{1/2}$ we have considered the escape coefficient to be a function of time, $\Theta([Cd^+](t))$. The rate of mixing the Beitler levels by helium amounted, in this model, to 10^{-13} cm^3/s.

Number density of cadmium atoms, N_{Cd} (in cm^{-3}) as function of temperature T_g (in K) has been calculated by the following formula:

$$N_{Cd} = \frac{1}{T_g} \exp\left(62.51 - \frac{12550}{T_g}\right).$$

This formula describes the reference values from Ref. 13, in the temperature range from 586 to 913 K accurate to better than 0.3%.

The model we have developed enables calculations to be made for four lasing transitions in cadmium ions at 325.0, 441.6, 533.7, and 537.8 nm wavelengths (see Fig. 1).

To make assessments of the effect that CCl_4 addition may produce on the active medium kinetics we have allowed for, in our model, the following components: molecular chlorine, Cl_2, the atomic chlorine in the ground state, Cl, and in an excited state, Cl*; the molecules of CCl, CCl_2, CCl_3, CCl_4, CdCl*, HeCl*; atomic carbon, C; as well as the ions of CCl^+, CCl_2^+, CCl_3^+, CCl_4^+, Cl_2^+, C^+. Besides, in order to study the influence of impurity admixtures we involved in calculations the reactions with nitrogen whose molecules and atoms are capable of capturing the relaxation flux that pumps the Beitler levels $4d^95s^2\ ^2D_{5/2,\,3/2}$ of the cadmium ions. All in all we have taken into account, in our model, about 300 reactions in the mixture of He–Cd–CCl_4–N_2.

2. THE RESULTS CALCULATED

2.1. Pumping a He–Cd(–CCl_4) laser by a microsecond-duration electron beam

In this section of the paper we present a test of our kinetic model by comparing the calculated results with the results of experimental investigations of a He–Cd laser emitting at 325.0 nm wavelength when pumped by a pulsed electron beam of 42-microsecond duration (at the level of a pulse base). Detailed description of the experimental set up and the results obtained may be found in Refs. 8 to 10. The results we have obtained when modeling the kinetics of processes in a He–Cd mixture under the action of a low-current electron beam are as follows. The calculated and experimentally measured laser emission pulses at 325.0 nm wavelength are shown in Fig. 2.

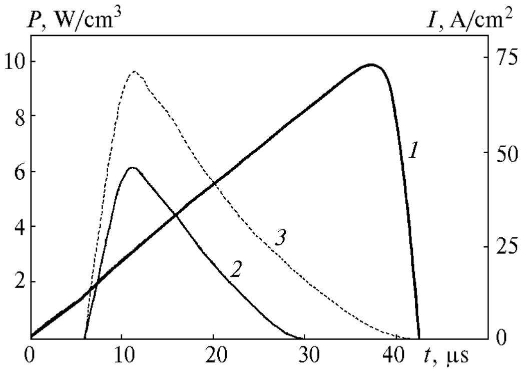

FIG. 2. The electron beam pump pulse (1) and the laser emission pulse at 325.0 nm *wavelength in a* He–Cd *mixture at* $T_g = 375°C$, $[He] = 2.7 \cdot 10^{10} cm^{-3}$, *and* $r = 99.5\%$; *experiment (2), theory (3).*

Time behavior of the lasing exhibits a fall off of the output power to a complete break of lasing during the pump pulse. This peculiarity in the lasing time behavior is explained, in Ref. 9, to be due to a decrease in the rate of the upper lasing level population. However, no concrete mechanisms of this process are being discussed in that paper. In our opinion, the limitation of the laser pulse duration is caused by the parasitic absorption of the laser emission by the cadmium atoms in the state $5s5p\,^3P_2$. Accumulation of cadmium atoms in these metastable states mostly occurs due to the dissociative recombination of the molecular cadmium ions, Cd_2^+. By the moment when the lasing reaches its maximum ($t = 10$ microseconds) the concentration of $qd^*(5p\,^3P_2)$ particles equals to $3 \cdot 10^{12}$ cm^{-3} and continues to grow in time. This amount of cadmium atoms in the metastable states is quite sufficient for effectively absorbing the laser emission regardless of the growing pump and increasing inverse population of the lasing levels.

A comparison between the experimental[10] and results calculated for a He–Cd laser in a mixture with an addition of CCl_4 is presented in Fig. 3.

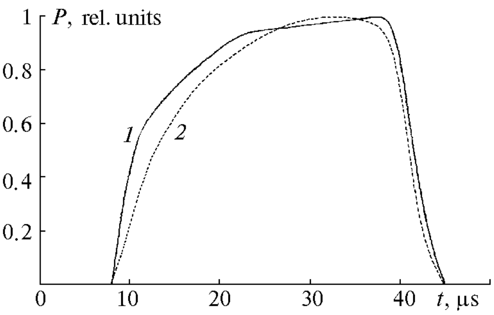

FIG. 3. The laser emission pulse in a He–Cd–CCl_4 *mixture at* $T_g = 375°C$, $[He] = 2.7 \cdot 10^{19} cm^{-3}$, $[CCl_4] = 3.6 \cdot 10^{16} cm^{-3}$, *and* $r = 99.5\%$; *experiment (1), theory (2).*

It is seen that a longer time delay of the lasing pulse occurs in the mixture with CCl_4 added as compared to that in the mixture without it. This is explained as that the relaxation flux is intercepted by the CCl_4 admixture molecules instead of the ions He^+, He_2^+, and atoms He^*. Certain delay is observed of the calculated pulse with respect to the experimental one and this may be assigned to a little bit higher rates of Penning reactions

$$He^* + Cql_4 \rightarrow He + Cql_2^+ +$$
$$+ Cl_2 + e \; (k_{\Pi} = 1 \cdot 10^{-10} \; cm^3/s)$$

and of the charge exchange reaction

$$He_2^+ + Cql_4 \rightarrow 2He +$$
$$+ Cql_2^+ + Cl_2 \; (k_{recharge} = 5 \cdot 10^{-11} \; cm^3/s).$$

taken in the model.

The occurrence of a quasi-stationary lasing when adding CCl_4 into the mixture is caused by a decrease in the $Cd^*(5p\,^3P_2)$ atoms concentration. The latter, in its turn, is explained by the fact that by the end a pump pulse the concentration of negative chlorine atoms is so high, $[Cl^-] = 2 \cdot 10^{13} cm^{-3}$, while that of electrons being so low, $N_e = 3 \cdot 10^{11} cm^{-3}$, that the molecular cadmium ions, Cd_2^+, enter the reactions of ternary and binary recombination with the chlorine ions more actively than with the electrons. This leads to a decrease in the concentration of molecular cadmium ions and, consequently, to a decrease in the rate of parasitic atoms formation, during the dissociative recombination of Cd_2^+, ions.

Thus, the dissociative capture of electrons, resulting in formation of negative chlorine ions and their subsequent ion-to-ion recombination on the molecular and atomic cadmium ions play the main role in lowering the parasitic absorption of the laser emission. In this case the interception of the relaxation flux due to the reactions of $He^+ + Cl^-(+He) \rightarrow$ $\rightarrow HeCl^*(+He)$ type becomes essential only in the final microseconds of the pump pulse when the concentration of Cl^- ions reaches its maximum value.

Optimal content of the CCl_4 in the mixture is $2 \cdot 10^{15}$ cm^{-3} (0.1 Torr). The decrease in lasing power at $[qql_4] > 2 \cdot 10^{15}$ cm^{-3} is due to an enhanced capture of the pump by this admixture at already inessential role of the parasitic absorption in formation of the laser emission.

2.2. Numerical simulations of a nuclear pumped laser emitting at 325.0 nm wavelength when pumped from a BARS reactor

In this section we present model calculations of a He–Cd laser pumped by the products of nuclear reactions from a self-quenching pulsed reactor of BARS–6 type.[14,15] In the simulations we calculated the parameters of output laser radiation at 325.0 nm wavelength that may be obtained per a single pump pulse. When optimizing the laser operation regarding the characteristics of active medium and of the resonator we have arrived at the conclusions that the optimal temperature of the active medium should be T_g = 290°C, helium concentration [He] = 10^{19}cm^{-3}, the transmission of the output mirror of the resonator T = 2% (the spacing between the mirrors was taken to be 40 cm), and the specific power deposition being 34 W/cm^3. Figure 4 presents the development in time of the lasing under these conditions. It is seen from the figure that the laser emission power reaches its maximum value of P_m = 27 mW/cm^3 by the moment in time, t = 200 microseconds, when the pump power is only at 63%-level of its maximum. Then the output emission power falls off. Although the inversion of the working levels population is still increasing the concentration of Cd*($5p^3P_2$) atoms reaches, by that moment, the value of $6\cdot10^{12}$ cm^{-3} and continues to grow either. It is just this fact that explains that the emission pulse terminates before the pump pulse terminates. The lasing efficiency regarding the energy deposited into the active medium per a pump pulse is 0.08%.

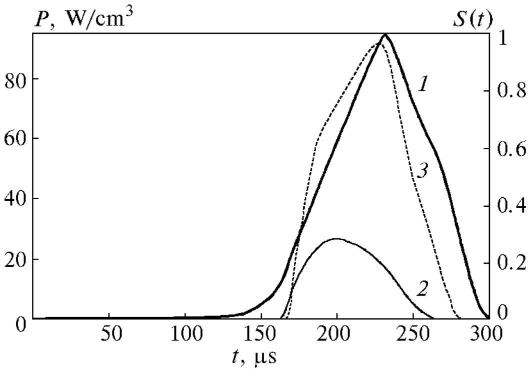

FIG. 4. The neutron pulse S(t) (1) from a BARS-6 reactor, the laser pulse emitted by the He–Cd *mixture under optimal conditions of* T_g=290°C, [He]=10^{19} cm^{-3}, T = 2% *(2) and the generation pulse (3) obtained in the mixture of* He–Cd–CCl$_4$.

Also shown in Fig. 4 is the laser pulse in the He–Cd–CCl$_4$ mixture under optimal conditions of T_g=350°C, [He] = $2.4\cdot10^{19}$ cm^{-3}, [CCl$_4$] = 10^{15}cm^{-3}, T = 2%, and the energy deposition into the active medium of 82 W/cm^3. In this case the output laser power is 90 mW/cm^3 and the lasing efficiency η = 0.11%. The cause for the optimal conditions to occur at higher temperature and pressure is the same as in the case with the laser pump by a low-current electron beam, namely, the addition of CCl$_4$ which reduces the parasitic absorption of laser radiation by the cadmium atoms in the excited state Cd*($5p^3P_2$).

Note that a foreign admixture present in the active medium, even a small amount of such, is capable of essentially reducing the population of the upper lasing level, especially at low temperatures. Thus the calculations made for a model mixture of He–Cd–N$_2$ showed that adding only 0.01% of nitrogen to He–Cd mixture under conditions optimal for lasing makes the emission power maximum at 360°C while being only 0.11 mW/cm^3.

CONCLUSIONS

We have carried out a numerical simulation of the active medium of a cadmium-vapor laser based on a detailed nonstationary kinetic model. The comparison made between the modeling results obtained and the experimental data available shows that the model proposed provides for quite a good description of the basic kinetic processes in the active medium studied. We have also studied the mechanisms through which the addition of CCl$_4$ into the active medium may affect the relaxation processes in the He–Cd plasma as well as the lasing on the transition $4d^95s^2\,^2D_{3/2} \to 4d^{10}5p^2P_{1/2}$ (with the wavelength of 325.0 nm) in the cadmium ion.

It has been shown in this study that the main factor limiting the lasing at 325.0 nm wavelength is the parasitic absorption of laser emission by the cadmium atoms in metastable states. For this reason it is advisable, in order to achieve the lasing at this wavelength to add into the working mixture a tiny quantity (below 1 Torr) of an electrically negative gas like, for instance, CCl$_4$ that is characterized by low rates of the charge exchange and Penning reactions with helium atoms in metastable states which produce the pumping of the upper lasing level.

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