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Mathematical simulation of the amplification of 1790-nm laser radiation in a nuclear-excited He – Ar plasma containing nanoclusters of uranium compounds

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Abstract. **The possibility of applying dusty active media in nuclear-pumped lasers has been considered. The amplification of 1790-nm radiation in a nuclear-excited dusty He – Ar plasma is studied by mathematical simulation. The influence of nanoclusters on the component composition of the medium and the kinetics of the processes occurring in it is analysed using a specially developed kinetic model, including 72 components and more than 400 reactions. An analysis of the results indicates that amplification can in principle be implemented in an active laser He – Ar medium containing 10-nm nanoclusters of metallic uranium and uranium dioxide.**

Keywords: *nanoclusters, nuclear-excited plasma, kinetic processes, fission fragments, laser radiation.*

1. Introduction

Active laser media with a fissile material sputtered in them appear to be rather promising. Their main advantage is the significant (by an order of magnitude) increase in the fraction of the energy introduced by fission fragments into the active medium in comparison with the case of heterogeneous pumping, where a fissile material is deposited on the walls of laser-active element [1]. However, there are problems in mastering this undoubtedly promising technology, which are caused by the presence of dust particles in the active medium. Along with the technological problems, related to the necessity of dealing with radioactive nanoparticles, there is a fundamental problem of extinction (attenuation) of laser radiation by dust particles. In addition, dust particles may negatively affect the component composition of the medium, as a result of which lasing characteristics may significantly deteriorate. Calculations [2] showed that the problem of laser radiation extinction can be solved using 10-nm dust particles in IR-lasing media. The kinetics of media with dust particles and structures has barely been studied; this especially holds true for nuclear-excited plasmas (NEPs).

The effect of nanoclusters on the component composition of media was estimated in [2 – 4] using specially developed kinetic models of He and He – Ar NEPs. The results indicated that the presence of nanoclusters with a concentration not lower than 10^{11} cm^{-3} leads to a change in the concentrations of electrons and molecular ions in the plasma by several times. In accordance with the data of [5], neutrons can be multiplied in media with these concentrations. Amplification of a weak 1790-nm signal in a He – Ar medium was also obtained. However, to simplify the model developed in [2, 4], some processes were disregarded, in particular, possible destruction of nanoclusters colliding with fission fragments and the state-to-state kinetics of the upper laser level (which was considered as combined).

In this paper, we report the results of studying the influence of 10-nm nanoclusters of uranium compounds on the kinetics and component composition of a He – Ar NEP with allowance for their destruction as a result of collisions with fission fragments. The amplification of 1790-nm radiation in the medium under study was investigated by mathematical simulation using an extended kinetic model.

2. Kinetic model of He – Ar NEP containing nanoclusters of uranium compounds, with allowance for their destruction

The previously developed kinetic model [2] includes 51 components and more than 200 reactions. A detailed description of physicochemical processes in a dust-free He – Ar NEP, including the processes of filling and devastation of laser levels, can be found, for example, in [6]. A specific feature of this model [2] was that, along with the state-to-state kinetics of the excited states of argon and helium atoms {except for the upper laser level $\mathrm{Ar(3d[1/2]_{1,0})}$}, it took into account the discreteness of the charge transfer to nanoclusters (i.e., the reactions of all charged clusters with charged components of the medium). A comparison with the results of [6] showed that this model quite correctly describes the kinetics of a dust-free medium [4] (see Fig. 1).

Nevertheless, the model disregarded the possible destruction of clusters as a result of their collisions with fission fragments. This simplification was based on the fact that, under the conditions characteristic of a NEP and at pump powers below 1 kW cm^{-3}, these processes do not affect much the lasing characteristics of the medium. However, this mechanism must be taken into account to describe more correctly the processes occurring in the medium and make the model more variable with respect to the external conditions.

Thus, the kinetic scheme considered in this study included not only the reactions of charged nanoclusters with charged plasma components but also the processes of destruction of nanoclusters colliding with fission fragments and, correspondingly, the interaction of individual clusters with plasma components. It was assumed that a collision leads to cluster

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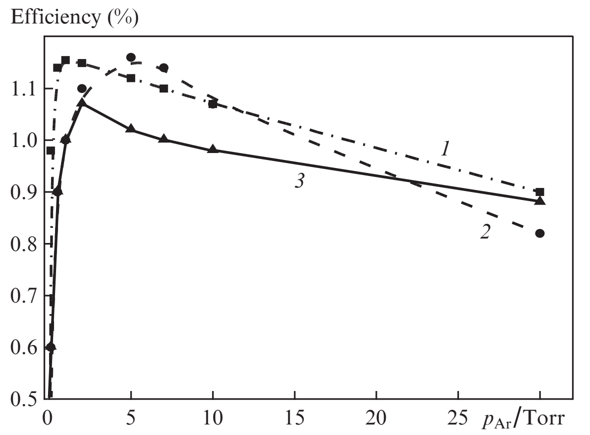

Figure 1. Dependences of the lasing efficiency for the line with λ = 1790 nm on the Ar pressure at a He–Ar mixture pressure of 1 atm, calculated (*1*) in this study and (*2*) in [6] and (*3*) experimentally found in [7].

splitting into two identical parts. In addition, the model was refined by considering the state-to-state kinetics of the Ar(3d) level, which includes the upper laser sublevel $Ar(3d[1/2]_{1,0})$.

The extended kinetic model includes more than 400 reactions and 72 components: helium atoms in the ground state and in ten lower excited states; atomic and molecular helium ions He^+ and He_2^+; argon atoms in the ground state and in the excited 4s, 4s′, 4p, 4p′, 3d, and 3d′ states; atomic and molecular argon ions, Ar^+ and Ar_2^+; heteronuclear $HeAr^+$ ions; argon excimers; electrons; and charged nanoclusters with charges up to 10.The reactions making the largest contribution to the kinetics within the developed kinetic model are listed in Table 1.

Along with the plasma-chemical reactions presented in Table 1 and other reactions between the plasma gas components, the scheme includes also the interactions of charged plasma components with nanoclusters, which can schematically be presented (as in [2]) in the following way:

$$e + D^{n-} \rightarrow D^{(n+1)-}, \qquad (1)$$

$$I^+ + D^{n-} \rightarrow D^{(n-1)-}, \qquad (2)$$

where e, D^{n-}, and I^+ are, respectively an electron, a charged nanocluster (n is the nanocluster charge in elementary charge units), and any positively charged atomic or molecular ion of the gas mixture. We took into account specifically the interaction of dust particles with charged plasma components as the strongest one.

The mathematical model includes the standard system of integro-differential kinetic equations:

$$\frac{d[X_i]}{dt} = \sum_k (\nu_{ik} - \mu_{ik}) R_k, \quad i = 1, \ldots, 72. \qquad (3)$$

The stiff nonlinear system of differential equations (3) was solved by the Gear method. The electron kinetics was calculated by solving (using an interpolation method) the Boltzmann equation for the spherically symmetric part of the electron energy distribution function. The calculations were performed with the aid of the upgraded software package described in [13].

Table 1. Main reactions in a helium–argon mixture.

Reaction	Rate constant/ $cm^{3(m-1)}\ s^{-1}$	References
$He_2^+ + e \rightarrow He^* + He$		[8]
$He^+ + He + He \rightarrow He_2^+ + He$	4.85×10^{-32}	[8]
$He^{**} + He^* \rightarrow He^+ + e + He$	4.5×10^{-9}	[9]
$He + He^* \rightarrow He_2^+ + e$	8.7×10^{-11}	[9]
$He + e \rightarrow H^+ + e + e$		[10]
$He + e \rightarrow He^* + e$		[10]
$He_2^+ + Ar \rightarrow He + Ar^+ + He$	2.0×10^{-10}	[6]
$He_2^+ + Ar + He \rightarrow He + Ar^+ + He + He$	5.0×10^{-31}	[6]
$He + Ar^+ + He \rightarrow HeAr^+ + He$	$(0.026/T_g)^{0.75}\times10^{-32}$	[6]
$He + Ar^+ + Ar \rightarrow Ar_2^+ + He$	1.9×10^{-31}	[11]
$HeAr^+ + Ar \rightarrow He + Ar_2^+$	3.6×10^{-9}	[6]
$He^+ + He + Ar \rightarrow He_2^+ + Ar$	$0.8\times10^{-33}/T_g$	[6]
$Ar + e \rightarrow Ar^* + e$		[12]
$Ar(4p[3/2]_{1,2}) + Ar \rightarrow Ar + Ar$	9.5×10^{-11}	[6]
$Ar(3d[1/2]_{0,1}) + Ar \rightarrow Ar + Ar$	3.1×10^{-11}	Estimate according to [6]
$Ar(4p[3/2]_{1,2}) + He \rightarrow He + Ar$	6.0×10^{-12}	Estimate according to [6]
$Ar(3d[1/2]_{0,1}) + He \rightarrow He + Ar$	2.0×10^{-12}	Estimate according to [6]
$Ar(3d[1/2]_{0,1}) \rightarrow Ar(4p[3/2]_{1,2}) + h\nu$	1.1×10^{6}	[6]
$Ar_2^+ + e \rightarrow Ar(3d[1/2]_{0,1}) + Ar$		Estimate according to [9]

Note: T_g is the gas mixture temperature in kelvins and m is the number of the components involved in reaction; the rate constants are omitted for the reactions with participation of electrons, because the calculations were performed using the energy dependences of the interaction cross sections.

3. Results and discussion

We considered the kinetics of a helium–argon NEP containing nanoclusters of uranium compounds. A He : Ar (400 : 1) mixture was investigated at pressures of 3 and 5 atm and specific power inputs to the medium of 125 and 250 W cm^{-3}. The pulse width was chosen to be 50 ms. Nanoclusters were considered to be spherical particles 10 nm in size; the attachment coefficient of electrons and ions to them was assumed to be unity.

Figure 2 shows the dependences of the quasi-steady-state concentrations of charged plasma components on the initial nanocluster concentration at different pressures and a power input of 125 W cm^{-3}. It can be seen that the concentrations of electrons and molecular helium and argon ions decrease with an increase in the nanocluster concentration above 10^{11} cm^{-3}. A detailed kinetic analysis of the medium showed that, under the conditions considered here, the contribution of dust particles to the destruction of molecular argon ions does not exceed 20%. At the same time, the main channel for filling the upper laser level is the dissociative recombination of molecular argon ion. The contribution of other reactions is small in comparison with this channel. This means that nanoclusters affect significantly the lasing characteristics of the helium–

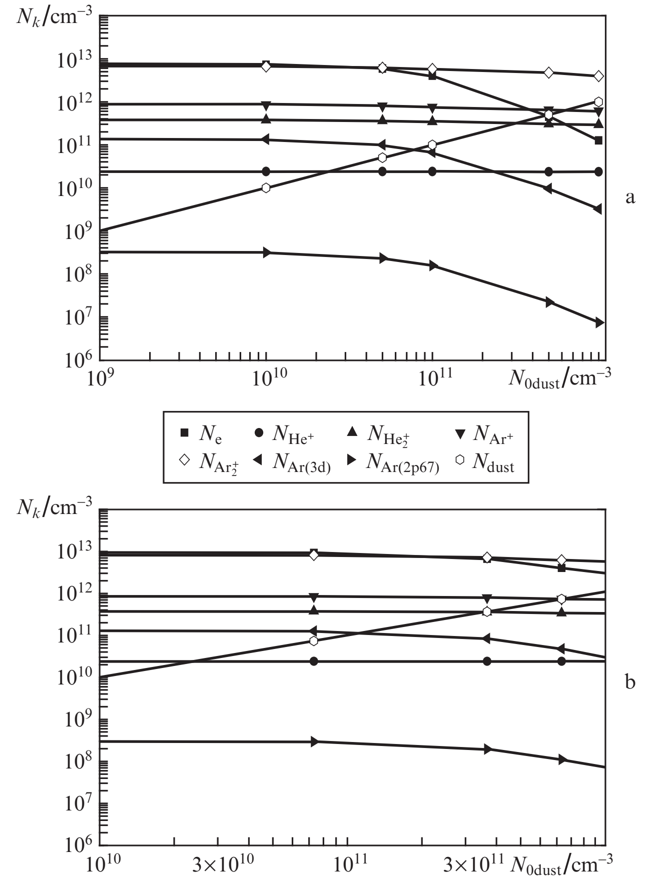

Figure 2. Dependences of the quasi-steady-state concentrations of the main plasma components (N_k) on the initial nanocluster concentration N_{0dust} at mixture pressures of (a) 3 and (b) 5 atm.

argon medium. In this context, we calculated the linear small-signal gain of a helium–argon medium containing nanoclusters of different uranium compounds, with allowance for the attenuation of laser radiation by dust particles (Fig. 3). The attenuation coefficients were calculated, as in [5], for 10-nm particles of metallic uranium and uranium dioxide and a laser radiation wavelength of 1790 nm.

Thus, the extension of the kinetic model by adding the processes of nanocluster destruction, as well as the state-to-state kinetics of the Ar(3d) level, allowed us to refine the lasing characteristics. This is evidenced by a comparison of the linear gains calculated within the previous and extended models (Fig. 4).

Note also that, according to the mathematical simulation results, the fraction of nanoclusters destroyed as a result of their collision with fission fragments does not exceed 13% (depending on the external parameters: mixture pressure and maximum specific power input). Under these conditions, the contribution of directly decomposed destroyed clusters to the destruction of molecular argon ions does not exceed 1%.

Having analysed the results obtained, one can state that a helium–argon medium excited by fission fragments and containing dust particles of metallic uranium or uranium dioxide allows low-intensity laser radiation at a wavelength $\lambda = 1790$ nm to be amplified for specific power inputs characteristic of nuclear pumping; this is one of necessary conditions for lasing.

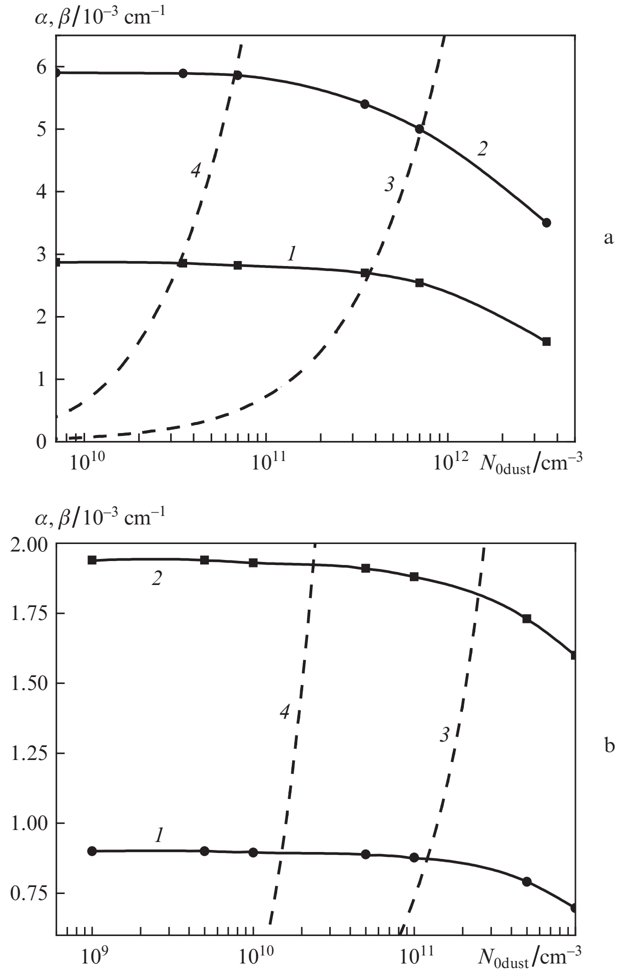

Figure 3. Dependences of the linear gain α of laser radiation in a He–Ar gaseous medium at a wavelength of 1790 nm and specific power inputs of (*1*) 125 and (*2*) 250 W cm^{-3} and the attenuation coefficients β by (*3*) U and (*4*) UO$_2$ nanoclusters on their concentration at mixture pressures of (a) 3 and (b) 5 atm.

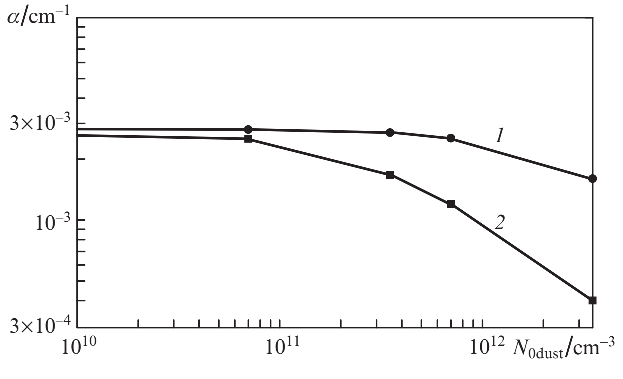

Figure 4. Linear gains calculated using the (*1*) extended and (*2*) previously developed kinetic models at a mixture pressure of 3 atm and a power input of 125 W cm^{-3}.

4. Conclusions

(i) We have developed a kinetic model of a He–Ar NEP containing nanoclusters of uranium compounds. The model includes 72 components and more than 400 reactions. The consideration of the destruction of nanoclusters colliding with fission fragments and the state-to-state kinetics of the Ar(3d) level has made it possible to refine the values of linear small-signal gain at a wavelength of 1790 nm in a He–Ar NEP containing nanoclusters of uranium compounds with different concentrations.

(ii) The results of mathematical simulation indicate that, at concentrations not lower than 10^{11} cm^{-3}, nanoclusters affect mainly electrons and molecular helium and argon ions. Specifically, they change their concentrations by several times. At the same time, at nanocluster concentrations below 10^{12} cm^{-3}, the linear small-signal gain decreases only slightly, a fact suggesting that amplification can in principle be implemented in the medium under study.

(iii) A kinetic analysis of the medium showed that the fraction of clusters decomposed as a result their collision with fission fragments in the medium under consideration does not exceed 13 %.

(iv) Metallic uranium and uranium dioxide nanoclusters 10 nm in size with concentrations below 10^{12} cm^{-3} barely attenuate 1790-nm laser radiation in a He–Ar NEP; thus, nanoclusters of specifically these materials appear to be most promising.

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