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Research Article

Laser-Induced Photon-Branched Chain Reaction in a Chemically-Active Gas-Dispersed Medium

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A promising avenue in the development of high-energy pulsed chemical HF/DF lasers and amplifiers is the utilization of a photon-branched chain reaction initiated in a two-phase active medium, that is, a medium containing a laser working gas and ultradispersed passivated metal particles. These particles are evaporated under the action of IR laser radiation which results in the appearance of free atoms, their diffusion into the gas, and the development of a photon-branching chain process, which involves photons as both reactants and products. The key obstacle here is the formation of a relatively large volume (in excess of 10^3 cm^3) of the stable active medium and filling this volume homogeneously for a short time with a submicron monodisperse metal aerosol, which has specified properties. In this paper, results are presented for an extensive study of laser initiation of a photon-branched chain reaction in a gas-dispersed H_2–F_2 medium.

1. Introduction

Theoretical investigations [1–9] have been carried out for the problem of photon branching in a two-phase (consisting of a gas and disperse particles) chemically active media. It has been shown that the introduction of a certain concentration of ultradisperse particles into the active medium of a chemical laser based on a chain reaction, and further their (particles) evaporation by external infrared (IR) radiation, can result in the generation of coherent radiation of energy considerably higher than the energy of an initiating input pulse at same frequency. New laser photons generated in the cavity can also participate directly in the laser-chemical reactions, facilitating the formation of active centers of a chain reaction (free atoms). If the number of emitted photons exceeds the number of those used in the initiation process, one can speak of a new type of branched chain reaction: photon-branched chain reaction (PBCR). The PBCR has practical interest because of it possible use in constructing powerful pulsed HF/DF-lasers and amplifiers without external energy sources.

The first experimental results to achieve a photon-branched chain reaction in a two-phase medium were published in [10]. The authors showed that during the injection of a fine-disperse aluminum powder with particle diameters of 0.3 μm covered by thin oxide film into a mixture of F_2 + H_2 + O_2 + He, the system remains stable. They observed the formation of free aluminum atoms under the action of radiation of a chemical laser with intensities as high as 10^7 W/cm^2, with a demonstration of the chemical mixture ignition. Comparison of the output energy in the presence and absence of disperse particles led to the conclusions that about 15% of the output energy is due to a PBCR.

The successes of the first experiments have given a positive answer to the question of the feasibility of a photon-branched chain chemical process in a two-phase active medium of an HF laser, and it has confirmed the accuracy of the theoretical models of the chemical and vibration-relaxation kinetics. We have developed a relatively simple HF laser kinetic model [1, 2] which includes the most important physical-chemical kinetics features under the assumption of rotational-translational nonequilibrium,

making it possible to calculate the specific energy of the coherent radiation, the pulse duration, the effective chain length of the reaction between fluorine and hydrogen, and the chemical and engineering efficiencies of the laser. This model was supplemented with an electrodynamical model [2–9] of the pulsed laser based on a solution of the wave equation. The wave approach for the description of a pulsed chemical HF laser amplifier based on a PBCR in an unstable telescopic cavity enables the detection of interesting optical effects in an active two-phase medium of H_2–F_2–O_2–He and Al particles and also of the new properties of the laser. These effects, the optimal parameters and the output performances of the laser and also some characteristics of an active medium, are presented in Table 1.

It has been shown [3, 5–9] that the laser chemical reaction can be ignited in an initial small focal volume of an active medium, and then spreads out of this minimal volume spontaneously in the autowave regime, without external power sources to subsequently fill the whole volume of the laser with a highly-intense electromagnetic field with self-supporting cylindrical photon-branching zones formed by the paths of the rays inside the unstable telescopic cavity. The ignition of an autowave PBCR under the condition of external signal focusing reduces strongly the input pulse energy necessary for initiation down to $\sim 10^{-8}$ J and, thereby, enables a huge value of the energy gain of about 10^{11}. In a system of this kind, there is no danger of self-excitation, even for large values of the gain, because the medium is not inverted in advance, and the energy is reserved in the form of the free energy of reactants.

A parametrical study [7–9] of a pulsed chemical laser based on a PBCR initiated in a gaseous disperse medium, composed of H_2–F_2–O_2–He and Al particles, by focused external IR radiation has been conducted. We see (Table 1) that both the observed effects of autowave spreading of a PBCR under the condition of external signal focusing and the effect of a huge laser energy gain of 10^{11} give the pulsed chemical HF laser the important properties of autonomy and compactness. We have determined the performances of the main laser units: the minimal linear size of the cavity, the minimal energy of the master oscillator and its power source, and the dimensions of the system of gas cylinders. We have also obtained the optimal composition of the reactants and the pressure of the laser working mixture. For example, at a general pressure of the working gases of $P = 2.3$ bar, the optimal parameters of the dispersed component (Alparticles with radius $r_0 = 0.09\,\mu$m and concentration $N_0 = 1.4 \times 10^9\,\text{cm}^{-3}$) and the composition of the working mixture, $H_2 : F_2 = 1 : 2$, the HF laser ensures an output energy up to ~ 1.5 kJ in a pulse from a rather small volume of about 2 L of active medium. A specific design was proposed for a self-contained compact pulsed HF laser with small linear sizes of the unstable telescopic cavity ($\sim 13 \times 14 \times 14\,\text{cm}^3$, where the diameter of the input (large) mirror is 14 cm), which can be initiated by a small submicrojoule master oscillator powered by an accumulator. The wave studies of the spatiotemporal behavior of the output laser radiation showed that the profile of the output pulse in the near-field zone is a torus, whereas in the far-field zone, the profile is Gaussian-like.

For a full-scale demonstration of the effect of photon-branching and design of a chemical laser based on a PBCR, it is necessary to investigate the permissible range of the concentration and size of disperse particles in the laser active medium, its dynamics, and aging; to develop the technique of preparing the gas-disperse medium of required properties. In this paper, we present an overview of the optimal and permissible parameters of the disperse component of HF active medium and give recommendations for the experimental realization of PBCR in a chemically-active gas-disperse medium.

2. Initiation of a Photon-Branched Chain Reaction in Aerosol

2.1. Kinetic Scheme

The initiation of a PBCR in an active medium of H_2–F_2-laser amplifier, containing passivated fine aluminum particles covered by an oxide film with a thickness ~5–10 nm, occurs by the mechanism offered in [1, 2, 8] for a pulsed HF generator. These aluminum particles are evaporated under the action of laser radiation, resulting in the appearance of free Al atoms. These atoms diffuse into the gas, and the photon-branching process begins. The initiation can be described by the following scheme:

$$\begin{aligned} &\text{Al (solid)} + n\hbar\omega \longrightarrow \text{Al (gas)},\\ &\text{Al (gas)} + \text{F}_2\ \text{(gas)} \longrightarrow \text{AlF} + \text{F}. \end{aligned} \tag{1}$$

After the appearance of free F atoms, the atoms then diffuse into the gas, and the chain reaction starts, leading to the formation of vibrationally excited HF molecules and, further, to generation of new photons:

$$\begin{aligned} \text{F} + \text{H}_2 &\longrightarrow \text{HF}^* + \text{H},\\ \text{H} + \text{F}_2 &\longrightarrow \text{HF}^* + \text{F},\\ \text{HF}^* &\longrightarrow \text{HF} + \hbar\omega. \end{aligned} \tag{2}$$

Some of the emitted photons are absorbed, which leads to the formation of new free Al atoms, while other emitted photons contribute to the net laser energy gain. The progressive growth of photons and free atoms takes place over time, and all of the particles (photons and free F and Al atoms) participate in active chain reactions. A reaction of this type is called a PBCR. This idea has been validated by computer calculations based on the solution of a system of differential equations of chemical kinetics, vibrational relaxation transfer of laser radiation, and laser evaporation of particles [1–8].

The advantages of using fine disperse particles of metals or other elements as the laser-active media are as follows.

(i) High, in principle, concentrations of free atoms.

(ii) Short time for producing high concentration of free atoms.

(iii) Uniformity of filling large active volumes with free atoms.

Table 1: Main properties and parameters of the HF-laser amplifier based on a PBCR. Here, r_0 and N_0 are the radius and concentration of the dispersed particles; I_0 and τ_{in} are the intensity and duration of the initiating radiation; E_{out} and E_{sp} are the full (i.e., for the whole volume of the laser active medium) and the specific (i.e., per unit volume) energies of the output pulse of radiation; P_{max} is the maximum generated power; k_{amp} is the energy gain factor; τ^L is the duration of the output pulse; $\tau^L_{1/2}$ is the output pulse duration at half of the maximum power; I_{thr} is the threshold intensity for optical breakdown of the active medium.

Observed effects and properties of the laser	Calculated optimal laser parameters	Output characteristics
Effect of autowave spreading of a PBCR throughout the volume of the active medium as self-supporting cylindrical zones of photon branching	Ratio of the reagents as $H_2 : F_2 = 1 : 2$. The range of general pressures of the mixture is P = 1–2.3 bar	$E_{out} = 1.5 \times 2.3$ KJ E_{sp} = 200–730 J/L $P_{max} \sim 10^{11}$ W
Effect of the giant laser energy gain of $k_{amp} = E_{out}/E_{in} \sim 10^{11}$, where E_{out} and E_{in} are the output and input energies, respectively	Ranges of Al particle radii and concentrations are, respectively: r_0 = 0.09–0.4 μm and $N_0 = 10^9$–10^7 cm^{-3}	$\tau^L \approx 800$ ns $\tau^L_{1/2} = 100$ ns.
Autonomous generation: a self-contained laser can be initiated by a small submicrojoule master oscillator powered by an accumulator	Performances of the master oscillator: input signal intensity $I_0 = 10^4$ W/cm^2, energy of the input pulse $E_{in} = 2.18 \times 10^{-8}$ J, duration of the initiating pulse, $\tau_{in} = 250$ ns	$k_{amp} = E_{out}/E_{in} \approx 10^7$–$10^{11}$
Property of compactness, which is determined by the performances of the main laser units: minimal linear sizes of the cavity, $\sim 13 \times 14 \times 14$ cm^3; minimal energy of the master oscillator, $E_{in} = 2.18 \times 10^{-8}$ J; minimal initially excited volume, $V_{in}^{min} = 10^{-7}$ cm^3. The volume of the active medium is about 2L	Geometric parameters of the unstable telescopic cavity are as follows: number of passes, $N_p = 4$; range of cavity lengths, 13–50 cm; range of magnification coefficients, β = 1.5–2; diameter of the coupling aperture, $d \le 3$ mm	The profile of the output pulse in the near-field zone is a torus, and in the far-field zone, the profile is Gaussian-like

(iv) Universality (ability to use various atoms).

(v) High specific surface area of the particles, exceeding the area of the walls (e.g., a discharge chamber), which increases the rate of depopulation of the lower active level.

The properties of an aerosol as an active medium of a pulsed chemical HF laser based on a photon-branched reaction should satisfy a number of requirements, which can be divided into two independent groups. These groups are as follows.

(i) The requirements imposed by the conditions for sustaining the photon-branched chain reaction in an H_2–F_2 mixture with fine disperse particles.

(ii) Those imposed by the aerosol optics.

2.2. Laser Evaporation of Particles

The formation of active centers of the chain reaction is achieved through the fast reaction of a fluorine molecule with an evaporated aluminum atom from the surface of a disperse particle under the action of external IR-laser radiation:

$$\mathrm{Al\ (gas)} + \mathrm{F_2} \longrightarrow \mathrm{AlF} + \mathrm{F}. \tag{3}$$

The characteristic time of this reaction is $\le 10^{-9}$ s at the concentration $[F_2] \ge 10^{18}$ cm^{-3}. Laser heating and evaporation of the disperse component can be described within the approximation of free molecular flow, which is valid for particles whose size does not exceed the mean free path of molecules in a laser mixture (see, e.g., [8]):

$$\begin{aligned} 4\pi r_0^2 \frac{dr_0}{dt}\rho_0 &= -\eta 4\pi r_0^2 \overline{V}_s(T_s)\rho_s(T_s), \\ \frac{4}{3}\pi r_0^3 \rho_0 C_s \frac{dT_s}{dt} &= \sigma_{abs} I - (T_s - T_g)\sum_M C_M 4\pi r_0^2 n_m \overline{V}_M(T_g) \\ &\quad + L_{evap} 4\pi r_0^2 \frac{dr_0}{dt}\rho_0 . \end{aligned} \tag{4}$$

Here, C_s and L_{evap} are, respectively, the specific heat and heat of evaporation of aluminum; I is the radiation intensity inside the cavity; σ_{abs} is the cross-section of absorption of laser radiation by an Al particle; T_s and T_g are the temperatures of the aluminum particles and the laser active medium, respectively; C_M, $\overline{V}_M$, and n_M are, respectively, the molar specific heat, average velocity, and concentration of the molecules of the Mth component of the laser mixture; η is the accommodation coefficient; $\overline{V}_s$ is the average velocity of the vapor molecules; $\rho_s(T_s)$ is the saturated vapor density of aluminum at T_s.

2.3. Optimal Concentration of Particles

Formation of active centers of the reaction chain and their spreading into the medium is thus possible around each fine particle in the IR laser radiation field. The distribution of free atoms in the active volume can become sufficiently homogeneous in a time τ_h, which is limited by the duration of the laser chemical process ($\sim$1 microsecond), provided that the condition $R_{ev} \le 2R_{diff}$ is satisfied, where R_{ev} is the average distance between the particles, $R_{diff} \sim \sqrt{D\tau_h}$,

and D is the diffusion coefficient of the active centers in the laser medium. This sets the lower limit to the required concentration of disperse particles as $N_0 \approx (1/R_{ev}^3)$. Optimization of the aerosol parameters from the point of view of the laser chemical process under the conditions of initiation of a pulsed HF amplifier was reported in [8]. Calculations showed that the optimal concentration of aluminum particles is $N_{Al} = 1.4 \times 10^9\,cm^{-3}$ for the radius $r_0 = 0.09\,\mu m$.

2.4. Optimal Particle Size

The optimal particle size is governed, for the selected concentration, by the aerosol optics, so that the attenuation coefficient of laser radiation representing scattering and absorption in a disperse medium does not exceed the local active medium gain α. Calculations of the optical characteristics of spherical disperse particles at a given radiation wavelength λ were based on the Mie diffraction theory and the single-scattering approximation [11].

The Mie formalism requires the use of two dimensionless input parameters, $\rho = 2\pi r_0/\lambda$ and $\delta = \rho m$, where m is the relative value of the complex refractive index of the aerosol at the wavelength λ. Computer calculations of the dependences of the attenuation K_{att}, scattering K_{sc}, and absorption K_{abs} coefficients at the HF laser wavelength ($\lambda = 3.3\,\mu m$) in the size range $r_0 = 0.5$–$1\,\mu m$ of aluminum particles ($m = 3.2 - 29.5i$ at 300 K) were performed in [2, 8] and plotted in Figure 1. It is evident from Figure 1(a) that for particle radii $r_0 > 0.5\,\mu m$, the scattering coefficient K_{sc} is considerably greater than the absorption coefficient K_{abs}, that is, lasing is impossible in such a strong scattering medium. Figure 1(b) demonstrates, on a much larger scale, the region of intersection of the $K_{sc}(r_0)$ and $K_{abs}(r_0)$ curves. The absorption of IR radiation predominates over the scattering by aluminum particles, $K_{abs}(r_0) \geq K_{sc}(r_0)$, if $r_0 \leq 0.15\,\mu m$. It follows that the aerosol optics imposes stringent conditions on the maximum permissible particle size in a two-phase active medium of the laser, which should satisfy the condition $\rho = 2\pi r_0/\lambda < 1$. An aluminum aerosol with the parameters $r_0 = 0.09$–$0.4\,\mu m$ and $N_{Al} = 10^7$–$10^9\,cm^{-3}$ can be recommended as a model two-phase active medium of an HF laser based on a PBCR.

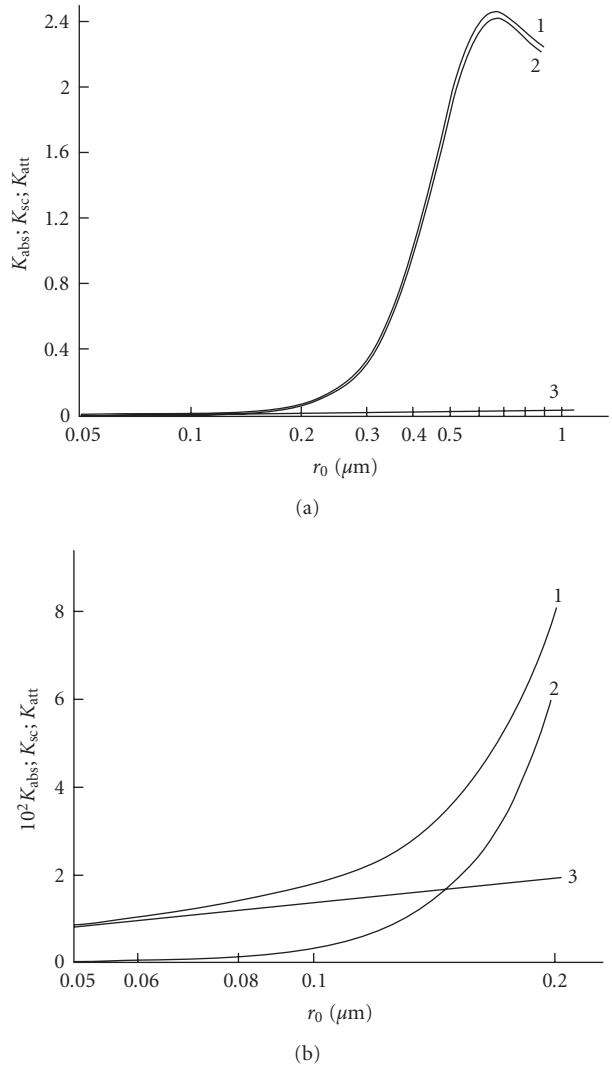

Figure 1: Dependencies of the attenuation K_{att} (1), scattering K_{sc} (2), and absorption K_{abs} (3) coefficients at the HF-laser wavelength ($\lambda = 3.3\,\mu m$) for (a) the size $r_0 = 0.5$–$1\,\mu m$ of aluminum particles ($m = 3.2$–$29.5i$ at 300 K) and (b) the region of intersection of curves 2 and 3 shown on an enlarged scale.

2.5. Lifetime of a Disperse Component

The main disadvantage of lasers with two-phase active media is the fast degradation of the disperse component and the consequent short lifetime of the active medium with specified properties. Continuous variation of the properties of the disperse phase with time results in a deterioration of the output characteristics of a laser or in complete quenching of the laser action.

The first attempt to model degradation of the disperse phase in a laser active medium was made for the operating conditions in a pulsed chemical HF laser [12]. The degradation processes are practically the same for all types of lasers with disperse media. We adopted an approximate coagulation model [13] and assumed the Stokes law. In the first approximation, we studied the time dependence of the overall characteristics of an aerosol in a laser active medium, including the average size and the total concentration of disperse particles, and we also calculated the formation time and lifetime of a two-phase active medium of an HF laser with the given parameters.

The processes occurring in a gaseous medium containing a disperse aerosol may be modeled on the assumption of coagulation, precipitation, and (in the presence of electric charges) electrostatic scattering of disperse particles. Aerosol coagulation was described by the lognormal quasi-self-sustaining model of [14], based on an approximation in which a lognormal model [15] and high-current sectional model, conserving the square of the particle volume, [16] are adopted. Here, the particle size distribution is described by the standard lognormal function.

The precipitation of particles was taken into account on the basis of the following model assumptions. The active medium was taken to be ideally mixed, continuous, incompressible, and characterized by a finite viscosity. The particle shape was assumed to be spherical. All these assumptions were to apply the Stokes law to the aerosol precipitation. The concentration of the disperse component remaining in the medium after particle precipitation was calculated from

$$N_{\text{prec}} = N_0 \frac{\exp(-\nu t)}{H}, \tag{5}$$

where H is the transverse size of the active medium of the HF laser, assumed to be equal to the diameter of the larger cavity mirror.

An analysis of the influence of the induced electrostatic charge of fine disperse particles of a conducting aerosol on the rate of its degradation was made within the overall framework of the problem of the lifetime of a two-phase active medium. The formation of a metal aerosol is practically always accompanied by its charging, for example, as a result of thermionic and photoemission of electrons. The concentration of the aerosol remaining after electrostatic scattering of the particles with the same charge and of the same size can be found from expression:

$$N_{\text{esc}} = \frac{N_0}{1 + 4\pi q^2 B N_0 t}, \tag{6}$$

where B is average mobility of the particles.

Specific calculations were carried out for a standard active mixture of the composition $H_2 : F_2 : O_2 : He = 100 : 400 : 40 : 210$ torr, into which an aluminum aerosol was injected in advance. At the initial time $t = 0$, the aerosol was assumed to be quasi-monodisperse with the average initial radius $r_0 = 0.05\,\mu$m and concentration $N_0 = 2 \times 10^9\ \text{cm}^{-3}$.

Fast coagulation, with the constant $\beta = 5 \times 10^{-10}\ \text{cm}^3\text{s}^{-1}$ (Figure 2) and sticking probability practically equal to unity for a submicron aerosol, results in continuous enlargement of the particles with a certain size distribution. The time for the establishment of a self-sustaining lognormal particle size distribution in the investigated two-phase active medium is about 25 seconds. Figure 2 shows the dependencies of coagulation and electrostatic scattering constants on the particle size. We can see that an increase in the average charge alters slightly the coagulation constant; this effect is significant only for small ($r < 0.1\,\mu$m) particles. For an aerosol with $r = 0.05\,\mu$m, the constants of these processes are comparable, and the rate of electrostatic scattering of the charged particles exceeds the rate of coagulation of the neutral particles only if the average charge is $q \geq 1.3e$. Therefore, at the first instant, such electrostatic scattering of submicron-charged particles competes with their coagulation, which results in a rapid loss of the particles from the volume of the active medium.

It follows from Figure 3(a) that during the first 50 seconds, there is a strong fall of the concentration of the charged aerosol with the average unit charge per particle down to the lowest value, $\sim 10^8\ \text{cm}^{-3}$ needed to sustain the laser chemical reaction. An increase in the coagulation time results in continuous spreading of the initial particle size distribution in the direction of larger particles and in a fall of the aerosol, concentration because electrostatic scattering is accelerated by the coagulation and precipitation of the particles.

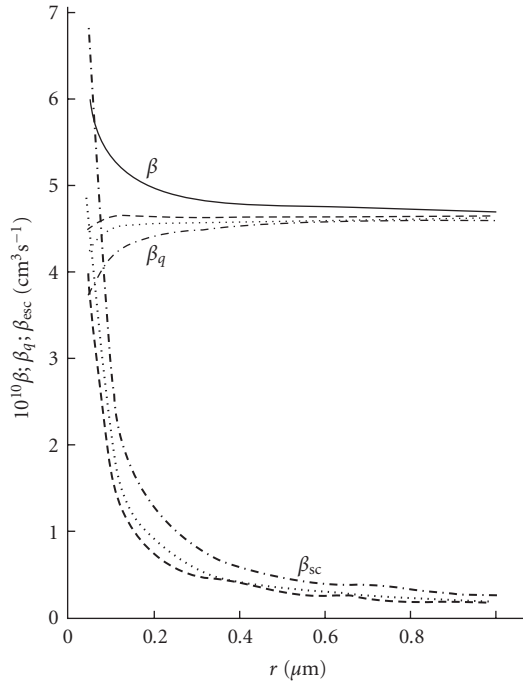

Figure 2: Dependencies of the coagulation constant of neutral particles β, the coagulation constant of charged particles β_q, and aerosol electrostatic scattering constant β_{esc} on the particle radius r for the average induced unipolar charge $q = e$ (dashed curve), $1.1\,e$ (dotted curve) and $1.3\,e$ (chain curve).

A comparison of the change, during the selected time step, in the concentration of the particles remaining in the medium after electrostatic scattering with the change in the total concentration of the particles in the medium undergoing coagulation and precipitation shows (Figure 3(b)) that, beginning from the particle radius $r \sim 0.2\,\mu$m, such coagulation and precipitation in an aerosol predominate strongly over electrostatic scattering, irrespective of the particle charge. When the selected time step is relatively large, $t \geq 1000$ seconds, so that the particles reach micron sizes, and particle precipitation begins to predominate over coagulation, as demonstrated in [13]. Therefore, in describing the gradual degradation of two-phase active media of lasers, it is necessary to take into account all three processes—electrostatic scattering, coagulation, and precipitation—which compete in different ways during different time intervals.

If the criteria for optical transparency of the active medium to the laser radiation are assumed to be $I_{\text{abs}}/I_0 \ll 1$ and $I_{\text{sc}}/I_0 \ll 1$ (I_{abs}, I_{sc}, and I_0 are the intensities of the absorbed, scattered, and incident radiations), we can estimate the maximum permissible lifetime of a two-phase active medium of the investigated laser. The strongest scattering of the $\lambda = 3.3\,\mu$m radiation corresponds to the aluminum particle radius $r = 0.6\,\mu$m and, consequently,

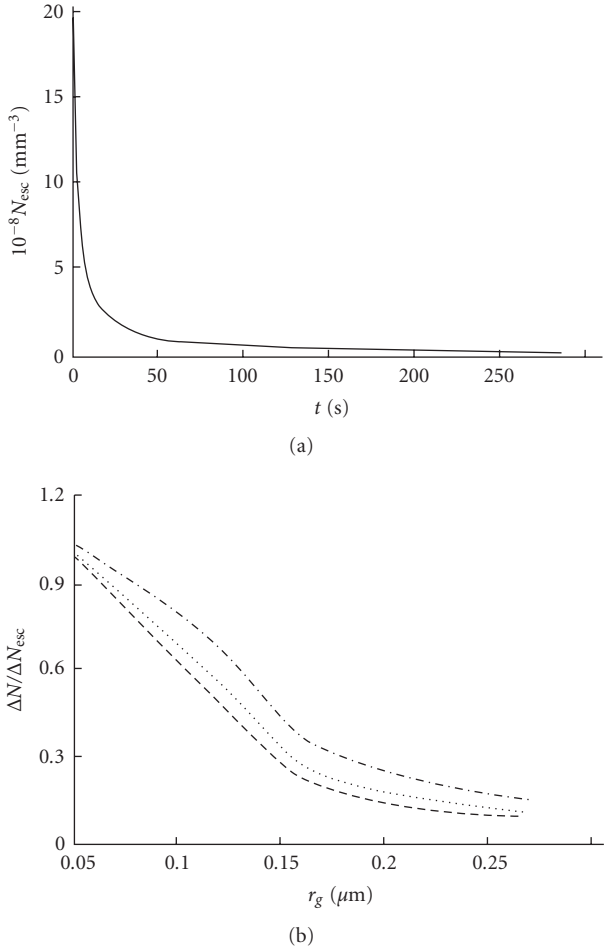

FIGURE 3: Time dependencies of the concentration N_{esc} (a) of a unipolarlycharged aerosol with the average unit elementary charge per particle under electrostatic scattering conditions and (b) of the ratio of the change in the total particle concentration N in the medium, which experiences coagulation and precipitation, to the change in the concentration N_{esc} of the particles which remain in the medium after electrostatic scattering, plotted as a function of the average size r_g of the particles participating in these processes, calculated for $q = e$ (dashed curve), l.l e (dotted curve), and 1.3 e (chain curve).

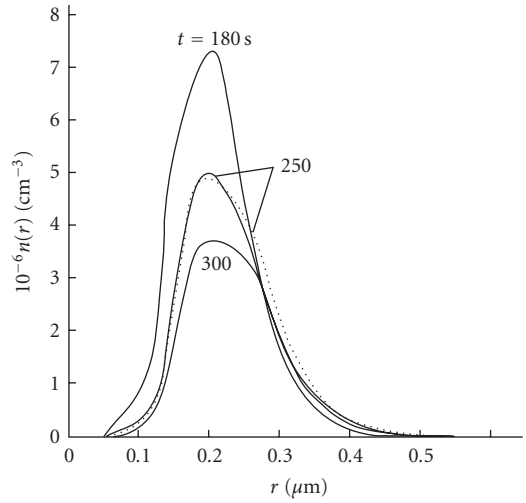

FIGURE 4: Particle size distribution $n(r)$ at different limiting time moments, calculated by taking account of coagulation and precipitation (continuous curves) and ignoring precipitation (dotted curve) of an aluminum aerosol in the active medium of a pulsed HF laser.

the criteria selected by us can be satisfied if the final size distribution $n(r)$ excludes almost completely particles of this radius.

It follows from Figure 4 which gives the distributions $n(r)$ calculated at different instants in time, taking account all the processes resulting in the aging of a neutral aluminum aerosol in the active medium of an HF laser, that the condition $n\ (r = 0.6\,\mu\text{m})/N_0 \ll 1$ is well satisfied for $t =$ 250 seconds. For a longer time interval, for example, $t = 300$ seconds, numerical calculations give an aluminum particle concentration $\sim 3 \times 10^3$ cm^{-3} with the impermissible radius $r = 0.6\,\mu$m. The dashed curve in Figure 4 represents the particle size distribution calculated ignoring precipitation. We can see that the proportion of the large particles in the size distribution increases, and consequently, the lifetime of an aerosol with the specified parameters decreases further by 50 seconds.

Thus, the above analysis of the processes resulting in degradation of the disperse component of two-phase laser active media allows us to draw the following main conclusions.

(i) The lifetime of two-phase active media with specified properties is determined by electrostatic scattering, coagulation, and precipitation of the aerosol when the real particle size distribution function is taken into account.

(ii) An analysis of the optics and laser-chemical kinetics makes it possible to recommend an aluminum aerosol with the parameters $r_0 = 0.09$–$0.4\,\mu$m and $N = 10^7$–10^9 cm^{-3} for a two-phase active medium of the HF laser.

(iii) The calculated lifetime of the HF mixture with disperse Al particles, characterized by the properties given above, is 250 seconds.

2.6. Aerosol-Evaporation Reactor Cavity

In the previous sections, we have determined the aerosol properties satisfying the laser active medium and their dynamics over time. We have considered, also, the first experiments on the formation of a gaseous disperse medium of a pulsed chemical H_2–F_2 laser. However, there remains an unsolved problem of generating a relatively large volume (in excess of 10^3 cm^3) of the active medium of such a laser amplifier and filling this volume homogeneously with a submicron monodisperse metal aerosol which has specified properties. An analysis of the aerosol optics and laser-chemical kinetics of the relevantPBCR shows that the ranges

of the permissible parameters of the disperse component in which oscillation is possible in an HF laser are as follows: the radius of passivated aluminum particles should be r_0 = 0.09–0.4 μm, and their concentration should be $N_0 = 10^9$–10^7 cm^{-3}.

The simplest method for the formation of a two-phase active medium is atomization of a previously prepared fine powder. However, when this method is used, it is impossible to ensure the optimal parameters of the dispersion, size, and concentration of the particles, and also the homogeneity of the filling (because, e.g., of particle agglomeration). Moreover, coagulation, gravitational precipitation, and electrostatic scattering of the injected particles result in continuous variation of the parameters of the disperse component and, consequently, in deterioration of the output characteristics of the laser amplifier or even suppression of lasing. An analysis of the degradation processes in a two-phase active medium of an HF laser shows that, within these ranges of the parameters, the lifetime of the active mixture containing disperse aluminum particles is $\tau_{coag} = 250$ seconds.

Experiments [10] have demonstrated stability of a fluorine-hydrogen mixture with an injected highly disperse component (Al particles with r_0 = 0.2–1 μm and concentration $N_0 = 10^5$ cm^{-3}) against spontaneous self-inflammation and that a chain reaction can be initiated by evaporating such particles by IR laser radiation. On the other hand, these experiments reveal that in order to achieve lasing and a full-scalePBCR, it is necessary to employ improved methods for the formation of a gas-disperse medium with the required parameters and for homogeneous filling of large working volumes in a short time. For example, in a time interval $t < \tau_{coag} = 250$ seconds, it is necessary to form an aerosol with specified properties and to inject a gaseous mixture into the active volume of the laser. The time needed to fill this volume with an H_2–F_2 mixture, and to ensure its homogeneous mixing amounts is , 3-4 minutes in the experiments, and consequently, no more than 10 seconds remain for the formation of the aerosol.

A solution to this problem, proposed below, involves adoption of a fundamentally new method for the preparation of a two-phase active medium of a pulsed HF laser in an aerosol reactor coupled structurally to an unstable telescopic cavity. The use of such a closed HF oscillator-amplifier system, based on aPBCR and containing a device for the generation of a homogeneous aerosol, reduces considerably the time needed for the formation of a two-phase active medium and ensures that the disperse component has the necessary parameters. On the other hand, this system requires an external-pulsed energy source, and it is promising primarily for experimental research on the implementation of aPBCR and investigation of this reaction. The method may be of practical interest also for the improvement of metal vapor lasers and the nascent new aerosol laser technologies [17].

The basic design of the proposed laser amplifier with a two-phase active medium is shown in Figure 5. This laser amplifier consists of an unstable telescopic cavity (1), which is structurally coupled to an aerosol reactor (2). The front cavity mirror has an aperture for coupling to a master oscillator (3). The aerosol reactor is in the form of

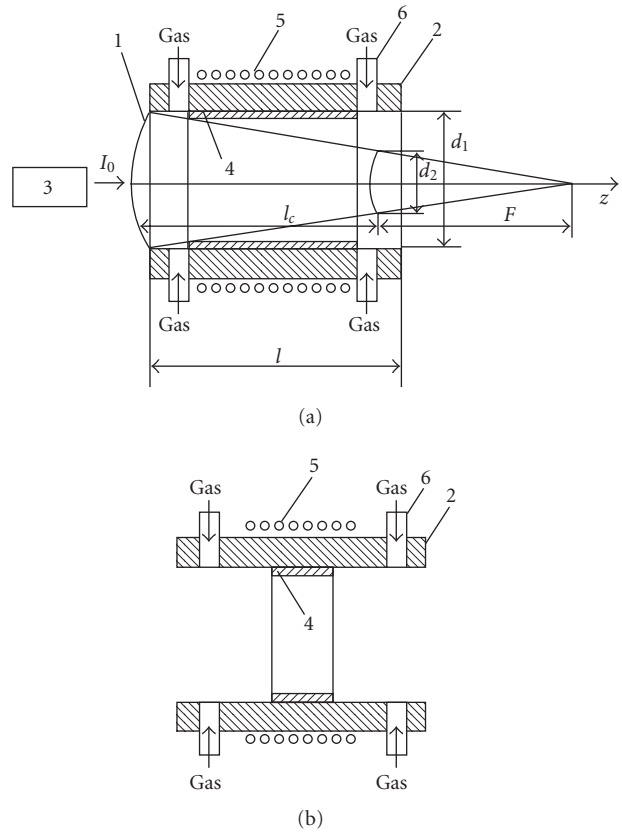

Figure 5: Simplified basic designs of a laser with (a) an aerosole-vaporation reactor and (b) an aerosol reactor with a limited length of the evaporated film : $d_{1,2}$ are the cavity mirror diameters; l_c is the cavity length; F is the focal length of the output mirror; l is the reactor length; (1) unstable telescopic cavity; (2) aerosol reactor; (3) master oscillator; (4) evaporated film; (5) solenoid; (6) gassupply pipes.

a demountable cylindrical chamber made of heat-resistant quartz glass. Plasma deposition or some other method is used to deposit, on the inner wall of this chamber, an aluminum film (4) of the required thickness. This metal film can extend over the whole length of the aerosol reactor (Figure 5(a)), or it may form a closed ring of limited length at the center of the internal wall of the quartz glass chamber (Figure 5(b)). The aerosol chamber variant with a limited length of the evaporated film is preferable from the point of view of preventing contamination of the end walls of the chamber by the metal, which is not permissible when the chamber is used inside the laser cavity to generate a two-phase active medium. The metal film is subjected to a homogeneous electromagnetic field generated in a solenoid (5) and evaporated, so that it mixes with the atmosphere of a low-pressure carrier gas (helium). The reactor contains also a power supply and a capacitor bank. The linear dimensions of the chamber are governed by the cavity parameters.

Let us now consider the formation of a two-phase laser active medium in the aerosol reactor. A bank of capacitors with a calculated capacitance C is discharged over a short time interval by closing the circuit. The discharge excites

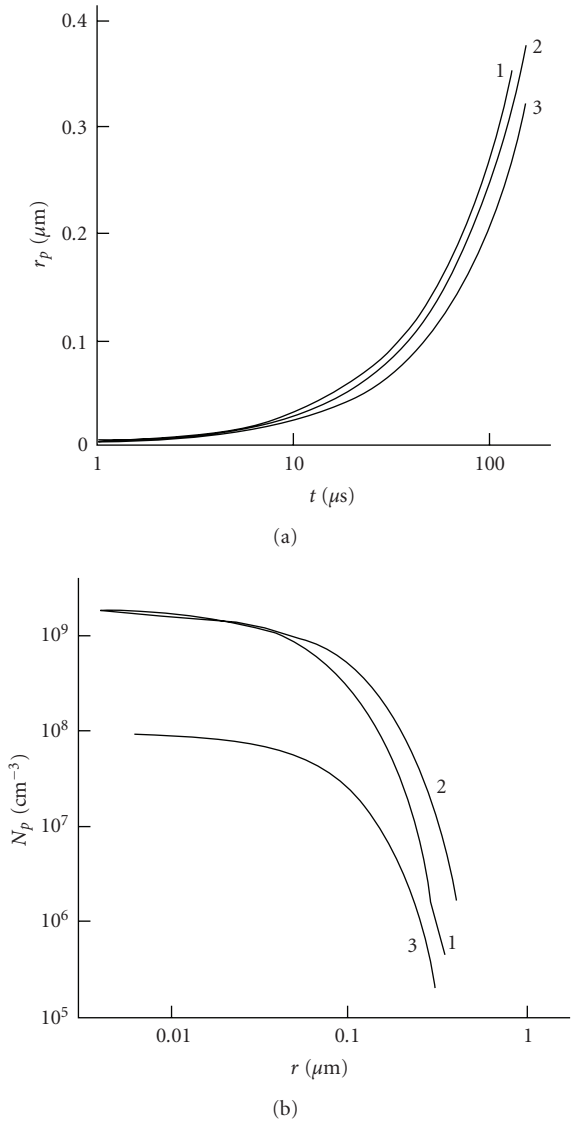

FIGURE 6: (a) Evolution of clusters in time and (b) distribution of the concentration of the particles formed by the condensation of Al vapor, described in terms of the particle size $N_p(r_p)$, calculated for helium buffer (carrier) gas pressures $P_{\text{He}} = 600$ torr (1), 400 torr (2), and 300 torr (3).

the solenoid, which has the following parameters: its length is l_L, the number of turns is N_L, the wire cross-section is S_L, and the resistance of the primary winding is R_1. A homogeneous magnetic flux Φ is generated inside a closed region surrounded by the primary winding whose inductance is L_1. This flux is sufficient for the evaporation, over a short time interval, of the metal film, where the film acts also as a secondary winding whose resistance is R_2. The metal vapor spreads in the surrounding cold gaseous He, then condenses to submicron metal particles. The metal can be prevented from reaching the end faces of the chamber, and the region with aluminum vapor can be filled homogeneously if the film is evaporated under the conditions of laminar flow of the carrier gas through pipes (6) placed at the ends of the chamber (2). A small amount of oxidant (O_2) injected into the reactor after condensation of the metal vapor produces an oxide film on the surfaces of the aluminum particles. Such an oxide film is 5–10 nm thick and prevents spontaneous self-inflammation of the fluorine-hydrogen working mixture of the laser. The necessary average size of the resultant particles and their concentration are ensured by varying the thickness of the initial metal film and by suitable selection of the electric parameters of the apparatus. The aerosol chamber variant with a limited length of the evaporated film (Figure 5(b)) has an additional advantage that it is possible to vary the aerosol parameters by altering the evaporated film length.

The evaporation of an aluminum film with a given mass $m_0 = 0.1$ g requires an energy $\Delta E = L_{\text{ev}} m_0 \approx 1088$ J. Calculations show that a solenoid with a primary winding made of copper wire 4 mm in diameter and containing $N_L = 50$ turns, in combination with a capacitor of capacitance $C = 10\,\mu$F subjected to a voltage $U_c = 50$ kV, can ensure evaporation of this film over a time $\tau_{\text{ev}} = 1/(2\gamma) \approx 0.217$ millisecond. The same results can be obtained for a lower voltage across the capacitor, for example, $U_c = 18$ kV, if the selection is made of a solenoid with a primary winding consisting of wires with a small diameter (1.5 mm) when the number of turns is $N_L = 4$. The energy stored in the capacitor is then $E_C = 1620$ J, which is sufficient to evaporate the film and to compensate for the energy lost in heating the wires in the primary winding and for the heat lost to the ambient gas and to the chamber walls. The variant with a small number of primary winding turns in the solenoid corresponds to an aerosol reactor with an evaporated film of finite length (Figure 5(b)), which is preferable for intracavity use.

Discharge of a capacitor which stores this energy results in slight heat dissipation in the primary winding of the solenoid as well. Consequently, the primary winding temperature rises by $\Delta T = E_C/C_p m_{Cu} \approx 40^\circ$C, where C_p and m_{cu} are the specific heat and mass of a copper wire in the primary winding. Film evaporation is accompanied also by the loss of heat to the ambient gas and to the quartz-glass chamber walls. Let us ignore the energy lost as a result of heat conduction in the gas and estimate the upper limit to the heat lost to the chamber walls from the film being evaporated. The depth Δh of penetration of heat into quartz glass over the course of diffusive heat exchange with the heated film during the total evaporation time $\tau_{\text{ev}} \approx 0.217$ millisecond is $\Delta h = \sqrt{2\chi\tau_{\text{ev}}} \approx 2 \times 10^{-5}$ m, where χ is the thermal diffusivity of quartz glass. The energy lost through the chamber walls in the case of a four-turn solenoid does not exceed $\Delta Q = \Delta m_1 C_{pl} \Delta T_1 \approx 500$ J, where Δm_1 is the mass of the heated chamber wall, C_{gl} is the specific heat of quartz glass, and ΔT_1 is the temperature drop at the film-glass interface. The newly-formed aluminum vapor expands adiabatically inside the reactor and experiences a condensation "jump" accompanied by the formation of clusters with critical radius r_{cr} and high concentration (Table 2) in a short time of $\tau \approx 1$ microsecond. The evolution of these clusters at various ambient gas pressures is illustrated in Figure 6 and Table 2. It follows from our

Table 2: Results of numerical calculations of the formation of a two-phase active medium and of the output characteristics of an HF laser. Here, P_{He} is the buffer (carrier) gas pressure; r_{cr} and r_0 are the critical and final radii of the condensed particles; N_0 is the particle concentration; t^* is the time needed for the formation of particles with the final radius r_0; E_{out} and E_{sp} are the total and specific energies of the output radiation pulses; P_{max} is the maximum output power; τ_p is the duration of the output pulse; and k_{amp} is the energy gain.

P_{He}/torr	$10^4\, r_{cr}/\mu$m	r_0/μm	$10^{-7}\, N_0/\text{cm}^{-3}$	t^*/μs	E_{out} /J	E_{sp}/J L^{-1}	$10^{-10}\, P_{max}$/W	τ_p/ns	$10^{-4}\, k_{amp}$
300	9.4	0.276	5	134	1076	141.23	1.9	736	7.1
400	9.5	0.198	10	80.5	1465	192.26	2.0	716	9.7
600	11	0.158	13	57.6	1485	194.92	1.5	737.6	9.9

calculations that the time t^* needed for the formation, over the course of condensation, of particles with final radius r_0 is ~50–150 microseconds, which corresponds to the time of attainment of the balance between the masses of the evaporated film on one hand and of the clusters and vapor on the other. The growth of these particles in the range of radii from r_{cr} to $r_0 \approx 0.1\,\mu$m occurs at a practically constant and maximum concentration N_0 (Figure 6(b)), which makes it easier to satisfy the condition on r_0 for a two-phase active medium of an HF laser. For large particles, $0.1\,\mu\text{m} < r_0 \leq r_{max} = 0.5\,\mu$m, the particle (cluster) concentration N_0 rapidly falls to $\sim 10^5\,\text{cm}^{-3}$, and the attenuation of the generated IR radiation is then slight. It follows that condensation of the aluminum vapor in 50–150 microseconds produces a polydisperse aerosol with a distribution of the particle radii close to a "step" (Figure 6(b)). The width of this step matches the particle radius range of $r_{cr} \approx 1\,\text{nm} \leq r_0 \leq 0.1\,\mu$m.

A fast reaction of aluminum with oxygen added to helium occurs on the surfaces of the growing clusters: $4Al + 3O_2 \rightarrow 2Al_2O_3$. The result is a dense protective Al_2O_3 film, 5–0 nm thick, on the particle surfaces. This film prevents spontaneous self-inflammation of the working mixture of the laser during the dark reactions. It was demonstrated experimentally in [10] that mixing of passivated aluminum particles—~0.5 μm in diameter and with a concentration ~10^5 cm—with a laser-active mixture of the composition $H_2 : F_2 : O_2 : He = 50 : 114 : 11 : 635$ torr does not increase the rate of accumulation of the HF molecules, and such a mixture has the required long-term stability.

2.7. Beam Stability of a Chemically-Active Aerosol

The off-resonant initiation mechanism of a PBCR by laser evaporation of ultradisperse metal particles injected into a laser active medium imposes restrictions from below and above on the radiation intensity of the master oscillator. On the one hand, the intensity of the input radiation should be sufficient for effective evaporation of submicron metal particles, and on the other hand, it should be below the threshold intensity of the optical breakdown of an H_2–F_2 laser gas-disperse active medium. The kinetics of plasma formation in the laser infrared radiation field has been studied for gas-disperse fluorine containing media in [8].

The limiting factors for maximum values of the output energy of HF/DF laser based on a PBCR are only the beam stability of the laser active medium and the cavity mirrors. The thresholds for optical breakdown are as follows.

(i) Beam stability of metallic mirrors from molybdenum and copper, at a pulse duration of 100 nanoseconds, $E_{thr} \sim 100\,\text{J/cm}^2$, that is, $I_{thr} \sim 10^9\,\text{W/cm}^2$ (E_{thr} and I_{thr} are threshold energy and intensity).

(ii) Optical breakdown threshold of the laser active medium is $I_{thr} \sim 10^{10}\,\text{W/cm}^2$ ($\tau_{thr} \sim 10$ nanoseconds).

(iii) Optical breakdown threshold of raw air is $I_{thr} \sim 2 \cdot 10^9\,\text{W/cm}^2$.

(iv) Optical breakdown threshold of purified air is $I_{thr} \geq 10^{10}\,\text{W/cm}^2$.

3. Conclusions

The results presented in this paper lead to the following main conclusions.

(i) An analysis of the aerosol optics, based on the exact Mie diffraction theory, and the laser chemical kinetics of a PBCR have been used to determine the range of the maximal permissible parameters of the disperse component, which is capable of supporting the operation of a pulsed chemical HF laser with a two-phase active medium, with particle radius $r_0 = 0.09$–$0.4\,\mu$m and particle concentration $N_0 = 10^9$–$10^7\,\text{cm}^{-3}$.

(ii) In this range of aerosol parameters, the lifetime of the active medium of the HF laser with the disperse Al particles is governed by the coagulation, precipitation, and electrostatic scattering of particles occurring in the time range of ~10^3 seconds.

(iii) The first experiments demonstrated the stability of a fluorine-hydrogen mixture with an injected fine disperse component (Al particles of radius $r_0 = 0.2$–$1\,\mu$m with concentration $N_0 = 10^5\,\text{cm}^{-3}$) under the conditions when spontaneous combustion is possible. It was also shown that a chain reaction could be initiated by the evaporation of particles under the action of IR radiation. The corresponding numerical calculations let us conclude that in some experiments, conducted in [13], the photon-branched chain reaction has been observed for the first time. For a full-scale demonstration of the effect of the photon-branched chain reaction, it is necessary to improve the technique of preparing the gas-disperse medium as discussed above.

(iv) The optical breakdown of a chemically active aerosol at the wavelength of the generated radiation (λ_g = 3.3 μm) is characterized by the threshold intensity I_{thr} = 1–3 $\times$ 10^{10} W/cm^2.

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