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Influence of the cavity losses on the energy and spectral characteristics of a pulsed chemical chain-reaction HF (DF) laser

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Abstract. The dependences of the energy and spectral characteristics of the radiation of a pulsed photoinitiated chemical chain-reaction HF (DF) laser, operating at atmospheric pressure, on the optical cavity parameters were investigated. The maximum specific output energies of the HF and DF chain lasers (53 and 43 J litre^{-1}) and their fullest spectra (47 and 83 lines, respectively) were similar when the unsaturated gain exceeded the threshold by a factor greater than 50. An analysis was made of how the energy and spectrum of the radiation of the HF (DF) laser were influenced by the spatiotemporal inhomogeneity of the initiation and of the course of the chemical chain reaction. There was a marked effect of the CO_2 molecules in the active medium and in the air filling part of the volume of the complex cavity of the DF laser on its efficiency and emission spectrum.

1. Introduction

Excitation of the vibrational – rotational levels of the HF and DF molecules in the chain reaction

$$F + H_2(D_2) = HF(DF) + H(D)\ ,$$

$$H(D) + F_2 = HF(DF) + F, \qquad -\Delta H = 542\,(544)\ \text{kJ mol}^{-1}$$

(where $-\Delta H$ is the heat of reaction) consume approximately equal amounts of the energy released in the chemical reaction [1 – 3]. However, the specific output energies available from HF and DF pulsed chemical lasers (PCLs), based on this chain reaction, usually differ by a factor of 2 – 2.5 [2 – 5]. Various investigators are of the opinion that this is associated with the larger (compared with HF) number of vibrational – rotational levels of the DF molecule (over which the excitation energy is distributed) and with the smaller Einstein coefficients and rate constants of the chain reaction in the DF laser. All this leads to a lower small-signal gain for the majority of the vibrational – rotational transitions in a DF laser, so that the extraction of the laser energy from the active medium of this laser requires a cavity with a higher Q-factor. It has also been suggested that a DF laser cannot provide an output radiation energy exceeding half the output energy of an HF laser because of a significant difference between the energies of the emitted photons and between the vibrational – translational (VT) relaxation rates.

The conditions for the attainment of equality of the maximum energy outputs from the HF and DF molecules have been formulated and partly implemented [6, 8], so that the maximum output radiation energy of a DF laser now exceeds 80% of the maximum output radiation energy of an HF laser.

We shall report a detailed experimental investigation and theoretical calculations of the dependences of the energy and spectral characteristics of the radiation from a photoinitiated chemical atmospheric-pressure HF (DF) laser on the cavity loss factor. This was done over a wide range of variation of this factor (from 0.001 to 40 m^{-1} in the calculation and from 0.1 to 20 m^{-1} in the experiment) in order to determine the conditions for maximising conversion of the chemical energy into the laser output radiation energy.

2. Mathematical model of an HF pulsed chemical laser

A mathematical model, including a description of the chemical reactions, of the vibrational and rotational relaxation processes, and of the spontaneous and stimulated emission was used to investigate the energy and spectrotemporal characteristics of the radiation from a PCL. It was based on the hypothesis of the spatial homogeneity of all the processes occurring in the interior of the cavity. Similar models have been developed by other investigators [9 – 12].

In the mathematical model under consideration, use was made of ordinary differential equations for the concentrations of the active-mixture components, of equations for the photon density, and of the energy balance equation. The constants given in Ref. [11] were mainly employed in the calculations relating to the HF PCLs. The equations for the photon density $\rho_{v,J}$ describe the lasing on the $v, J-1 \rightarrow v-1, J$ transitions with a gain $a_{v,J}$, the coupling-out of the radiation from the interior of the cavity, and the spontaneous emission (the term S_{sp}):

$$\frac{d\rho_{v,J}}{dt} = (c\alpha_{v,J}\rho_{v,J} + S_{sp})\frac{l}{L} - \frac{\rho_{v,J}}{\tau_{ph}}\ , \tag{1}$$

where

$$\tau_{ph} = -\frac{2L}{c\ln(R_{1eff}R_{2eff})} \qquad \text{and} \qquad k_r = \frac{L}{cl\tau_{ph}} \tag{2}$$

are the photon lifetime in the cavity and the cavity loss factor; l is the length of the active medium; L is the distance between the mirrors; R_{1eff} and R_{2eff} are the effective reflection coefficients of the cavity mirrors. The initial parameters of the calculation corresponded to the experimental conditions.

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3. Experimental setup

Modular designs of the chemical reactor and of the initiation system were used in the experiments. The separate reactor module was a metal tube 150 mm in diameter and 1 m long, fitted with connecting flanges. A tubular quartz flashlamp, 22 mm in diameter and with a light-emitting length of 0.9 m per module, was placed at the centre of the reactor along the cavity axis. The brightness temperature of the glow electric discharge in xenon at an initial pressure of 6 – 8 Torr reached 18 kK when the pulse duration was 3 μs at FWHM. The utilisation of the UV radiation from the lamp was improved by coating the inner surface of the reactor with a reflecting polytetrafluoroethylene screen, 5 mm thick. The energy released in the discharge circuit with a single flashlamp was 1.5 kJ. The reactor was filled with a $F_2 - H_2(D_2) - O_2 - He$ gas mixture at a pressure of 0.112 MPa. The mixture composition for the chosen atmospheric pressure laser design had been optimised earlier [13].

The chemical reactor was placed inside a plane – plane cavity. The use of different designs of a complex cavity (Fig. 1) and variation of the length of the active medium from 7.6 to 0.2 m made it possible to vary the cavity loss factor over a wide range: from 0.1 to 20 m^{-1}, respectively.

The output energy and its transverse distribution in the beam were recorded with the aid of TPI-2 and TPI-2A calorimetric detectors. The energy distribution in the laser radiation spectrum was determined by a method involving thermal recording of the spectrum on blackened photographic paper. A diffraction grating, a focusing mirror, and a reflecting wedge (employed for self-calibration of the output radiation energy of each spectral line) were used. The energy distribution in the far-field zone was measured with the aid of a reflecting wedge, a spherical mirror, and a blackened photographic-paper screen. The spatiotemporal characteristics of the spectrum and the dynamics of the divergence of the laser radiation were investigated with the aid of a streak camera. In addition, the initiation (FÉU-39 photomultiplier) and emission (Ge – Au photodetector) profiles were recorded. The influence of the atmosphere on the energy of the laser radiation and on its spectrum was eliminated by placing part of the measuring system in a pressure chamber, flush against the cavity output mirror.

In calculation of the energy released by the active medium, account was taken both of the energy coupled-out from the interior of the complex cavity by the mirrors and parallel-plane plates and of the energy lost in the mirrors and windows (owing to the absorption and vignetting of the diverging beam). The energy absorbed by H_2O and CO_2 molecules in the interior of the cavity was also considered.

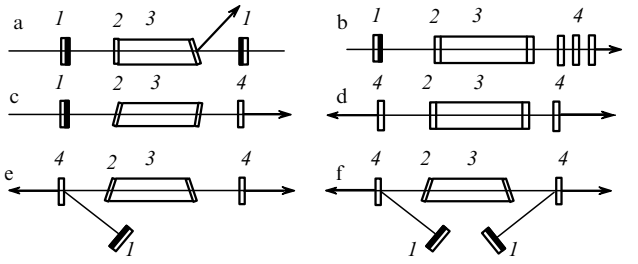

Figure 1. Design of a complex plane – plane cavity: (*1*) copper mirror; (*2*) plane-parallel CaF_2 plate; (*3*) active medium; (*4*) semitransparent mirror.

The effective reflection coefficient of the cavity mirrors was calculated taking into account all the energy losses enumerated above. This was done by means of the formulas

$$R_{\text{eff}} = rT^2 K_a(l_r) K_\theta(l_r)$$

for the left-hand (Fig. 1c) and right-hand (Figs 1a, 1c, and 1e) cavity arms, and

$$R_{\text{eff}} = rR_0^2 T^2 K_a(l_r) K_\theta(l_r)$$

for the left-hand (Figs 1e and 1f) and right-hand (Fig. 1f) cavity arms;

$$R_{\text{eff}} = \frac{[(R - rK_a(l_r)K_\theta(l_r)(R^2 - T^2)]K_\theta(l_{cr})}{1 - rRK_a(l_r)K_\theta(l_r)}$$

for the left-hand (Figs 1a, 1b, and 1d) and right-hand (Figs 1b and 1d) cavity arms. Here, R_0 is the reflection coefficient of the semitransparent mirrors placed in the cavity; R and T are the reflection and transmission coefficients of the reactor windows located in the cavity; r is the reflection coefficient of the external cavity mirrors;

$$K_a(x) = \sum_{i=1}^{N} E_i \exp[-(P_{CO_2} A^i_{CO_2} + P_{H_2O} A^i_{H_2O})x]$$

$$\left(\sum_{i=1}^{N} E_i = 1\right)$$

is the calculated integral coefficient representing the transmission of the laser radiation by the atmosphere on crossing a distance x in the latter; E_i is the relative energy of the ith spectral line; P_{CO_2} and P_{H_2O} are the water vapour and carbon dioxide concentrations in the atmosphere; $A^i_{CO_2}$ and $A^i_{H_2O}$ are the corresponding absorption coefficients of the ith spectral line; N is the number of the spectral lines; $K_\theta(x) = (1 + xD\theta)^{-2}$ is the calculated transmission coefficient of the windows of the working chamber, associated with the vignetting of the diverging laser beams on the apertures of the windows; D is the beam diameter; θ is the divergence of the radiation. For multimode lasing we have [14] $\theta = 0.7(\lambda/L)^{1/2}[\ln(R_1R_2)^{-1}]^{1/3}$, where λ is the radiation wavelength; l_r is the distance between the reactor window and the external cavity mirror; l_{cr} is the distance between the reactor windows.

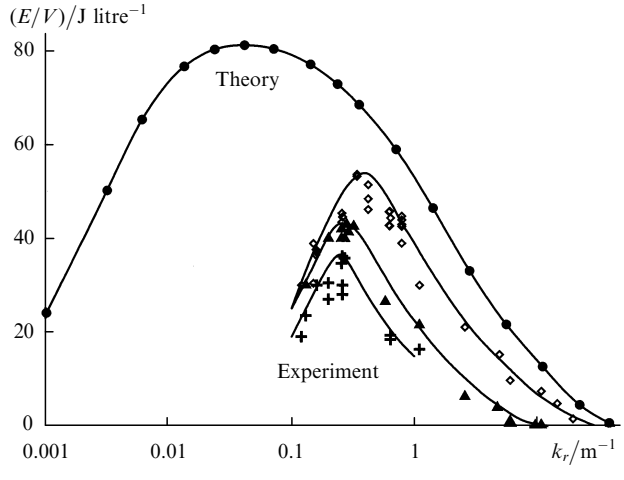

Figure 2. Dependence of the specific output radiation energy of HF (●, ◇) and DF (+, ▲) lasers on the cavity loss factor for $[CO_2] > 0.1\%$ (+) and $[CO_2] < 0.1\%$ (▲).

4. Results of a study of an HF (DF) pulsed chemical laser and discussion

The experimental and calculated dependences of the specific laser output energy on the cavity loss factor (Fig. 2) differed from the linear dependence [15, 16] usually employed for solid-state and gas lasers emitting radiation as a result of a small number of active transitions. With decrease in k_r, an accelerated near-exponential rise of the laser energy was observed in these PCLs for a relatively small change in the lasing pulse duration (2 – 3 μs). A large 'excess parameter' ($M > 50$), defined — in the present case of a multicomponent spectrum — as the ratio of the small-signal gain for the strongest line to the cavity loss factor k_r, is essential for such lasers in order to attain the maximum output radiation energy.

The rise of the specific output radiation energy was accelerated by the fact that, on reduction in k_r from its threshold value, an increasing number of lines with ever decreasing small-signal gains are involved in the lasing process. For example, for $k_r = 20$ m^{-1} only four lines belonging to the 2 – 1 vibrational band were present in the HF-laser radiation spectrum, for $k_r = 6.2$ m^{-1} about 20 lines in four bands were involved in lasing, and for $k_r = 0.1$ m^{-1} the lasing was based on 47 transitions in seven bands in the wavelength range 2.7 – 3.5 μm (Fig. 4). The nature of the variation of the calculated spectrum (Fig. 3) agreed well with the experimental variation. For an HF laser, the maximum specific energy of 53 J litre^{-1}

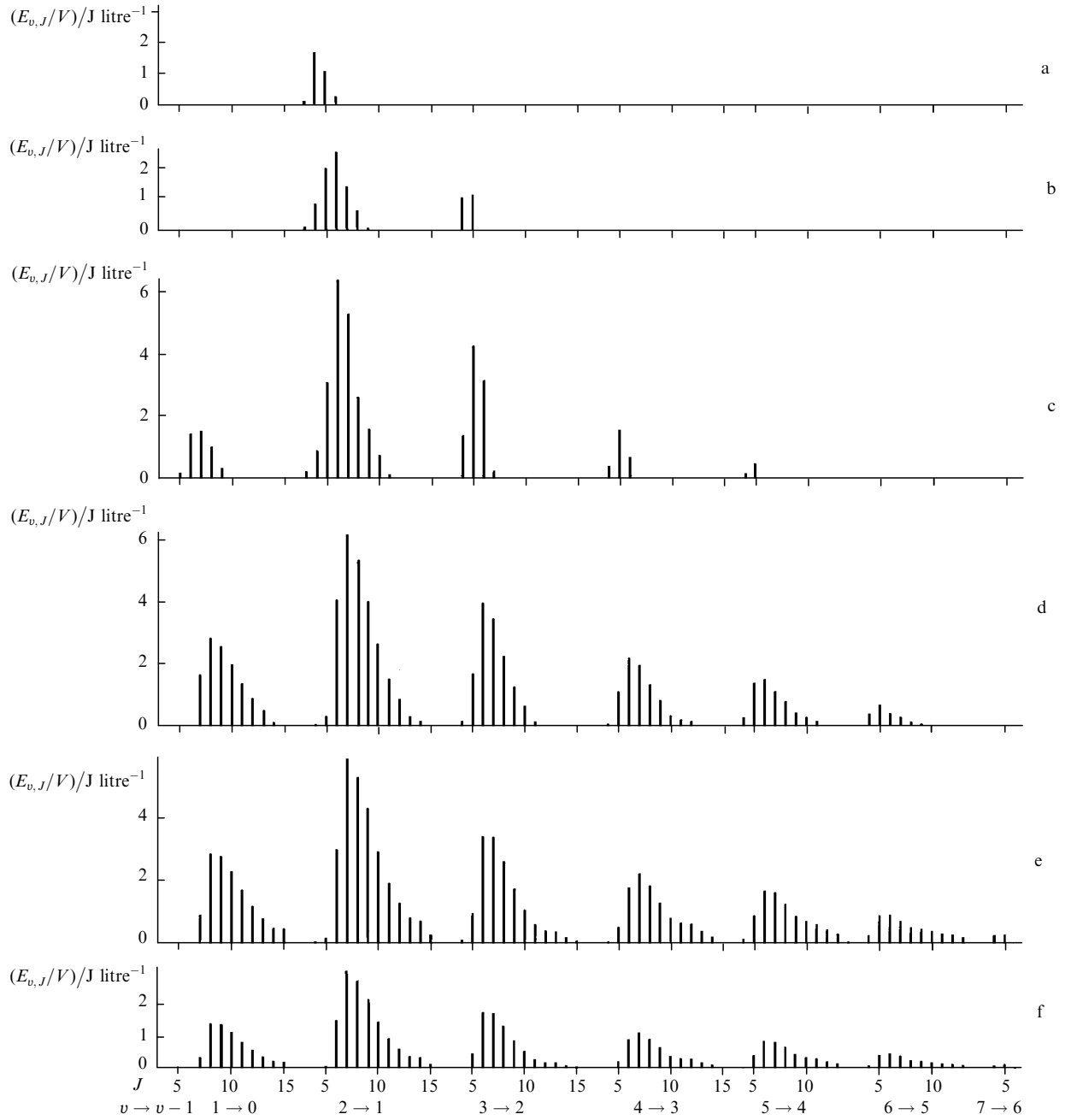

Figure 3. Variation of the calculated emission spectrum of the HF pulsed chemical laser as a function of the cavity loss factor; $k_r = 20$ m^{-1} (a), 7.8 m^{-1} (b), 2.8 m^{-1} (c), 0.35 m^{-1} (d), 0.041 m^{-1} (e), and 0.0031 m^{-1} (f).

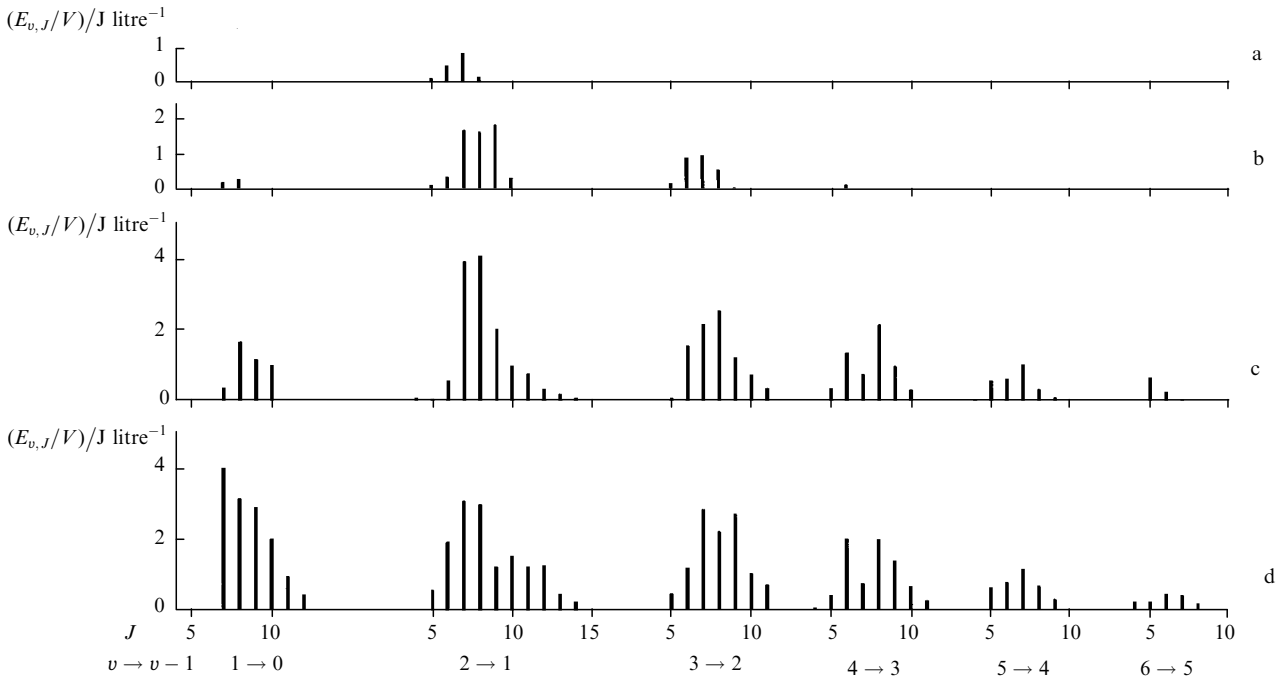

Figure 4. Variation of the experimentally recorded emission spectrum of the HF pulsed chemical laser as a function of the cavity loss factor: $k_r = 19.9$ m^{-1} (a), 6.2 m^{-1} (b), 0.59 m^{-1} (c), and 0.34 m^{-1} (d).

(50 J litre^{-1} is the specific output radiation energy and 3 J litre^{-1} represents the intracavity losses) was observed experimentally for $k_r = 0.4$ m^{-1}, whereas in the calculation the maximum specific energy of 83 J litre^{-1} was reached for $k_r = 0.04$ m^{-1}. Furthermore, an additional factor accelerating the rise in the specific output energy as k_r diminishes is activation of the transitions between high excited vibrational levels, which initiates a cascade of transitions (coupled to the former transitions) between lower vibrational levels.

In an experimental study of a DF laser it was found that the energy distribution in the emission spectrum of the DF molecule was influenced strongly by the selective losses in the CO_2 impurity, present in the cavity interior and having a strong absorption band in the wavelength range 4.2 – 4.3 μm. The removal of CO_2 from the cavity led to the appearance of lasing and to an increase in the intensity of the radiation emitted as a result of a large number of vibrational – rotational transitions [6, 7]. Two series of experiments were therefore carried out in the study of the dependences of the energy and spectral characteristics on the cavity loss factor. They differed in measures being taken in the second series to remove CO_2 from the cavity (the mirrors of a portable plane – plane cavity were flush against the reactor windows and an additional purification was carried out to remove the CO_2 impurity from fluorine by condensing the latter and filtering it off at liquid nitrogen temperature).

The specific output radiation energy in the region of its maximum increased approximately by 30% in the second series of experiments (Fig. 2). On decrease in k_r, lasing of the DF molecule appeared at $k_r = 12.5$ m^{-1}. For $k_r = 11$ m^{-1}, about 10 lines of the lower 2 – 1 and 3 – 2 bands were present in the spectrum. With $k_r = 2.6$ m^{-1}, lasing occurred on 33 lines in the first five bands, and with $k_r < 0.2$ m^{-1}, lasing was observed for over 80 spectral lines in ten bands in the wavelength range 3.5 – 5.0 μm (Fig. 5).

The results (Fig. 2) clearly demonstrate a tendency towards a gradual mutual approach of the specific output radiation energies of the HF and DF lasers on reduction in the cavity losses. The maximum specific energy of 43 J litre^{-1} was obtained from the DF laser for the cavity loss factor $k_r = 0.25$ m^{-1}; this amounted to 95% of the specific energy of the HF laser for the same values of k_r and to 80% for $k_r = 0.35$ m^{-1}.

Our calculated and experimental dependences of the specific output radiation energies of the HF and DF lasers on the cavity loss factor were similar. However, there were also significant differences: the maximum lasing energy was predicted for $M \approx 1000$ in the calculation and found at $M \approx 50 - 100$ in the experiment.

The calculated energy efficiency of the HF laser is half its maximum value when the loss factor is reduced to 0.002 m^{-1}. The principal cause of deterioration of the calculated energy characteristics at low values of k_r (< 0.04 m^{-1}) is the fact that the lifetime of the population inversion for separate transitions becomes comparable with the cavity photon lifetime τ_{ph}. Then the photons formed during the lasing process do not manage to escape from the cavity interior up to the lasing termination moment when the laser medium becomes strongly absorbing as a consequence of its being heated and because of accumulation of molecules in the lower states.

An appreciable reduction in the specific output radiation energy was observed in the experiment at appreciably higher cavity loss factors despite a further rise in the number of lasing transitions in the emission spectrum of the HF (DF) laser. A twofold reduction in the specific output energy was observed for the HF laser with the cavity loss factor $k_r \approx 0.1$ m^{-1} and for the DF laser with $k_r \approx 0.08$ m^{-1}. The small differences in the fall of the total output radiation energy of the HF and DF lasers are associated with the fact that, for a large number of transitions in the DF molecule, the small-signal gain is comparable with the loss factor of the active medium. This was confirmed by the dynamics of changes in the emission spectra of the HF and DF lasers: on reduction in the loss factor k_r from 0.35 m^{-1} (which corre-

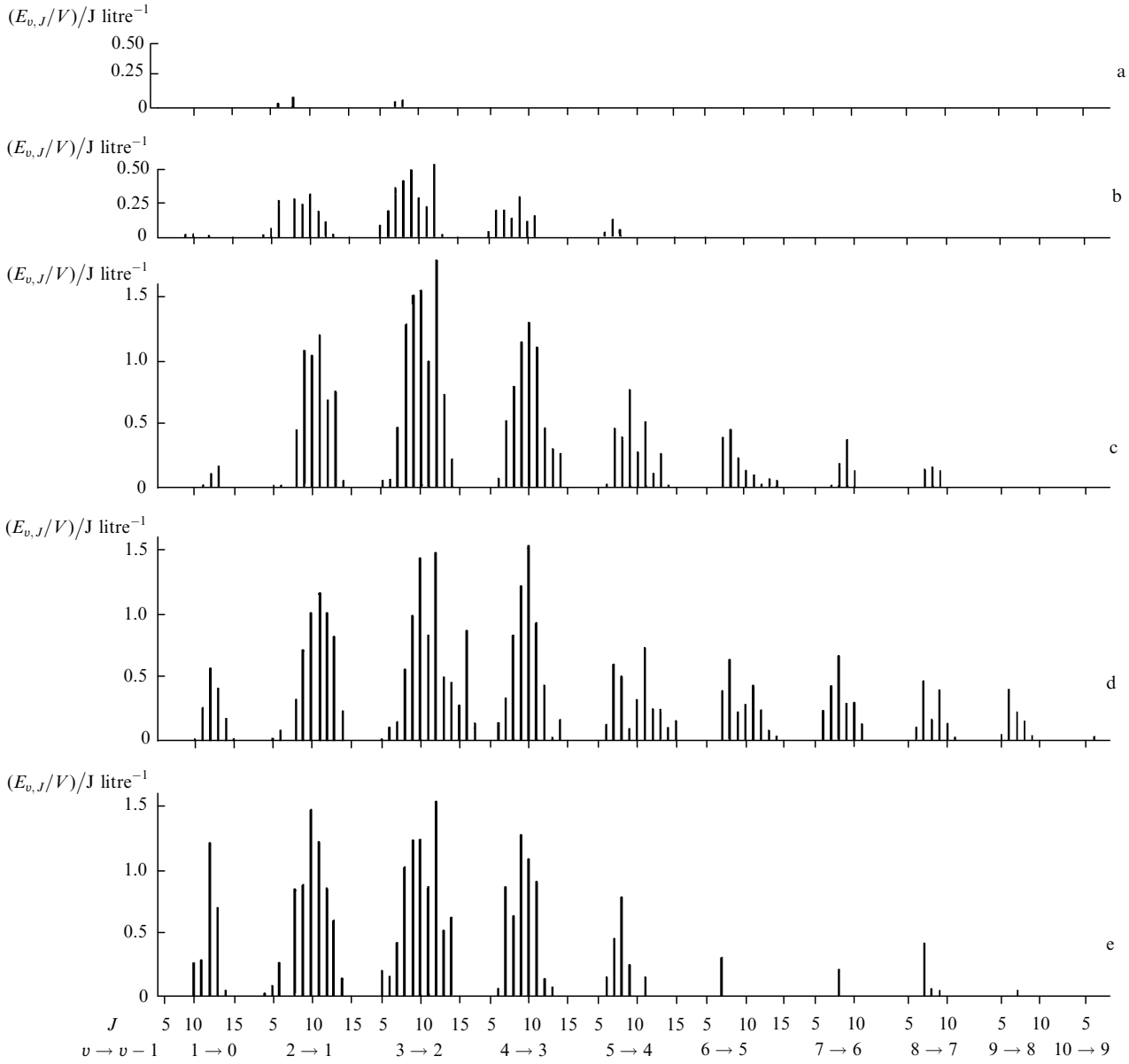

Figure 5. Variation of the experimentally recorded emission spectrum of the DF pulsed chemical laser as a function of the cavity loss factor: $k_r = 11$ m^{-1} (a), 2.6 m^{-1} (b), 0.58 m^{-1} (c), and 0.16 m^{-1} (d) for $[CO_2] < 0.1\%$, and $k_r = 0.20$ m^{-1} for $[CO_2] > 0.1\%$ (e).

sponds to the maximum of the HF-laser emission) to 0.1 m^{-1}, the output radiation energy decreased by a factor of 2 for the HF laser and by a factor of 1.5 for the DF laser, whereas the number of spectral lines increased by $\sim$5 and $\sim$20 lines, respectively, for the same lasers.

The principal cause of the differences between the calculated and experimental dependences for the HF laser is the neglect, in the calculation model, of the energy losses arising in the active medium. These losses appear following the development of a spatiotemporal inhomogeneity in the course of the chain reaction under the conditions of inhomogeneous illumination of the active volume. Another cause of the diffrences is the failure to take into account the possible losses on absorbing impurities (similar, for example, to CO_2 in the DF laser).

The influence of the inhomogeneity of the initiation of the initial mixture on the radiation characteristics can be seen clearly when the results of the experimental study of the spatiotemporal characteristics of the radiation beam are examined.

The transverse inhomogeneity of the initiation of the reaction in the active mixture leads to a marked spatiotemporal inhomogeneity of the emission spectrum of a pulsed chemical laser (Fig. 6). In the parts of the active medium adjoining the flashlamp, the initiation intensity is at its maximum, while at the periphery of the reactor it falls by a factor of 3 because of the cylindrical geometry of the illumination of the active volume and the absorption by fluorine of the UV radiation from the flashlamp (Fig. 7). The delays of the onset and termination of the lasing are unambiguously related to the intensity of the initiation of the active mixture: they increase with decrease in the illumination intensity and with increase in the rotational quantum number J.

The emission of radiation on separate vibrational – rotational transitions occurs layer-by-layer in the case of a plane – plane cavity: it appears and vanishes most quickly

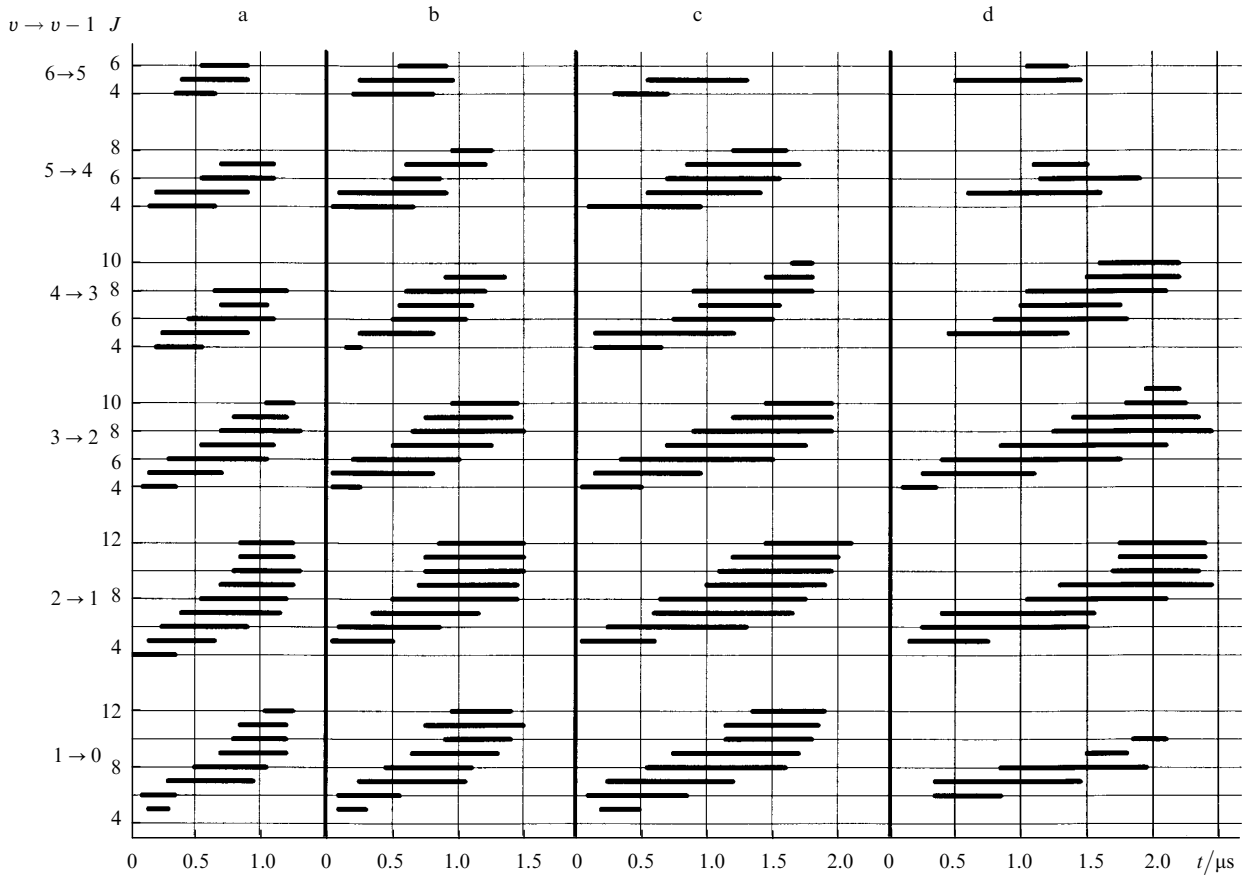

Figure 6. Evolution of the emission spectrum of the wide-aperture HF laser as a function of time for four regions of the active medium located at distances of 35 mm (a), 50 mm (b), 70 mm (c), and 95 mm (d) from the flashlamp surface.

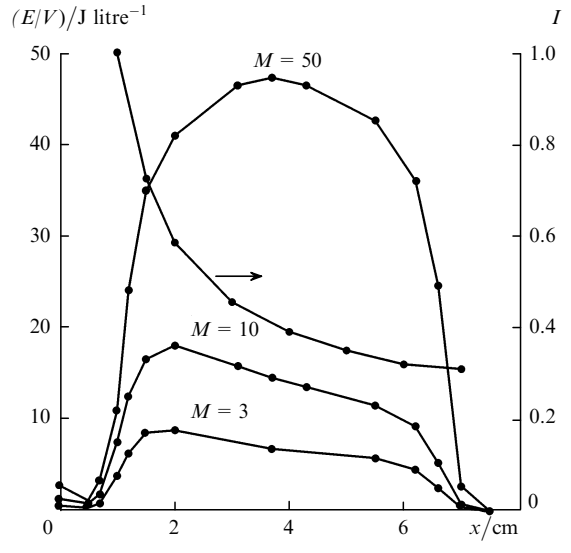

Figure 7. Distribution of the specific energy in the near-field zone of the radiation beam (at a distance of 6 m from the cavity mirror) for different 'excess parameters' M and the distribution of the relative initiation intensity I (x is the distance from the centre of the flashlamp).

in the layers of the active medium adjoining the flashlamp and then spreads to the periphery. The geometrical dimensions of the annular region of the radiation, migrating from the flashlamp to the periphery, decrease with increase in the rotational quantum number and for $J > 9$ these dimensions are smaller than the aperture of the cavity. The narrowing of the lasing zone and the appearance and growth of the refractive index gradient in the active medium during the development of a chain chemical reaction [17, 18] lead to additional radiation energy losses. Such losses arise owing to refraction of the radiation into the layers of the active medium which have already passed the lasing stage and in which the gain becomes negative.

The longitudinal inhomogeneity of the initiation of the reaction, associated with the nonsimultaneity of the triggering of the flashlamps and the inhomogeneity of the light emitted by them, can also reduce the laser radiation energy. This reduction is the result of the absorption of the radiation (generated in the regions of the active medium with a relatively low initiation level) by the regions of the active medium with a higher degree of initiation in which lasing terminates sooner. Evidently, the influence of the longitudinal and transverse initiation inhomogeneities should have the greatest effect in the regions of the active medium adjoining the flashlamp. In these regions the lasing pulse duration is minimal and the changes in the refractive index gradient and in the gain, as well as in the rate of these changes are maximal, as confirmed by measurements of the energy distribution over the beam cross section.

When the cavity loss factor was altered, a redistribution of the energy in the near-field zone of the beam cross section was observed (Fig. 7). At high values $k_r = 5-20\ m^{-1}$ (and for small 'excess parameters' $M \approx 1-4$), the energy distribution corresponded fully to the chemical-reaction initiation intensity distribution: the maximum in the radiation energy

density was observed at a distance from the flashlamp smaller than 1 cm. For low values $k_r < 0.2$ m^{-1} ($M \approx 50$), the maximum radiation density was recorded closer to the periphery of the radiation beam at $x \approx 3$ cm. An energy redistribution over the beam cross section with changes in k_r and all anomalous dependence of the specific output radiation energy on the initiation constant at low values of k_r were observed. They were consequences of different transverse variations of the distributed losses associated with deflection of the radiation beam onto the flashlamp wall and the absorption in the regions of the active medium which have passed through the lasing stage.

The experimental data presented above indicate a strong influence of the spatiotemporal inhomogeneity in the course of the chain process on the energy, spectral, and spatiotemporal characteristics of the radiation beam. A comparison was made of the observed dependences of the energy characteristics on the cavity loss factor for a homogeneous model of a PCL (calculation) and for an inhomogeneously initiated HF (DF) laser (experiment). This comparison indicated a possibility of a further rise of the specific energy output in the experiment as a consequence of the use of initiation systems ensuring a more homogeneous course of the chain process in the laser medium. An increase in the specific output energy owing to an increase in the initiation intensity demands the fulfillment of ever more stringent conditions as regards the spatial and temporal homogeneities of the initiation of the active medium owing to an increase in the absolute optical inhomogeneities.

5. Conclusions

Computational and experimental studies were made of the spectral characteristics of the HF and DF lasers over a wide range of the cavity loss factors (from 0.001 to 40 m^{-1}). It was found that, in order to obtain the highest energy characteristics of a PCL operating on the basis of a chain reaction, it is necessary to employ a cavity with a high Q-factor ensuring that the unsaturated gain exceeds the threshold gain by a factor greater than 50.

Evidently, for such high 'excess parameters', it is necessary to ensure sufficiently low intracavity losses. According to the calculated data, obtained for a homogeneous model of a PCL, the factor representing the unwanted losses in the active medium amounted to 0.002 m^{-1}. In the experiment, this factor reached 0.1 m^{-1}. The principal causes of the losses are associated, on the one hand, with the development of optical inhomogeneities of the active medium due to the spatiotemporal inhomogeneity both of the initiation and of the course of the chain reaction. On the other hand, there are losses associated with the presence of absorbing impurities (for example, of CO_2 for the DF laser) in the laser medium. For the most efficient conversion of the energy stored in the active medium into the laser radiation energy, it is therefore necessary, first, to employ an initiation system ensuring a high homogeneity in the course of the chemical reactions and, second, to use especially pure components for the preparation of the active media. Special studies are being made [6 – 8] in order to formulate the requirements which must be met by the purity of the gases employed and to determine the influence of various types of the spatiotemporal initiation inhomogeneity on the energy characteristics of the laser radiation.

We note that the use of a cavity with a high Q-factor and an increase in the length of the active medium to several metres made it possible to observe for the first time near-maximal specific energy outputs from the active media of pulsed chemical HF and DF lasers (53 and 45 J litre^{-1}). Moreover, this cavity enabled us to observe the fullest possible spectra of their radiation, incorporating high vibrational bands (up to the 7 – 6 and 10 – 9 bands), emitted in the wavelength ranges 2.6 – 3.4 μm and 3.5 – 5.0 μm, respectively.

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