

New highly efficient LGS-KhM erbium-doped glass for uncooled miniature lasers with a high pulse repetition rate

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Abstract. An investigation was made of several relationships governing excitation transfer in Cr–Yb–Er phosphate glasses. The activator concentrations were optimised. A new LGS-KhM erbium-doped laser glass and its modifications characterised by a low thermal expansion coefficient, an enhanced thermomechanical strength, and a high chemical stability were developed. Under free-running conditions the differential efficiency was 2.6% at the threshold pump energy of 5 J reached in active elements 2.5 mm in diameter and 35 mm long. Under *Q*-switching conditions a pulse of 30 ns duration and of 10 mJ energy was emitted when the pump energy was 6–7 J.

1. Introduction

The last few years have seen increasing interest of the developers of new laser systems (for various applications) in erbium-doped glass as a source of radiation in the spectral range safe to the human eye [1, 2]. Intensive investigations are proceeding along the following directions: improvements in the glass matrix and optimisation of the activator concentrations [3], search for efficient materials to be used as passive switches at the 1.54 μm wavelength [4], and replacement of traditional pump sources (xenon flashlamps) with selective sources (semiconductor lasers) [4, 5]. One of the most important applications of erbium light sources is pulsed distance measurements.

In spite of the considerable recent progress in the development of pulsed semiconductor pump sources, emitting in the spectral range of the absorption bands of ytterbium ions (900–1000 nm), in mass-produced erbium-glass devices preference is given to miniature flashlamps with a discharge gap 25–35 mm because of their high reliability and low cost, and also because of the simplicity of design of such systems.

We reported earlier [6, 7] the development of erbium-doped glass, called LGS-Kh, used in a number of minilasers with a record-low threshold pump energy and a high efficiency. This was possible because of introduction of an additional

sensitiser, in the form of Cr^{3+} ions, into the glass. The fullest utilisation of the flashlamp radiation used to pump active elements 2–4 mm in diameter was achieved by introducing into the same glass the maximal concentration, up to $6 \times 10^{19} \text{ cm}^{-3}$, of chromium ions (this corresponded to the absorption coefficient $4.0\text{--}4.3 \text{ cm}^{-1}$ at the 660 nm wavelength). In the single-pulse regime (with the repetition rate not exceeding 1/30 Hz), we obtained the following output parameters for active elements 2.5 mm in diameter and 35 mm long made of LGS-Kh glass and used in a ‘dense packing’ reflector with an 85% output mirror: the threshold lasing energy was 6 J, the differential efficiency was 2% in the free-running regime, the duration of the output pulses did not exceed 30 ns and their energy was 10 mJ when the pump energy was 7–7.5 J (a modulator with frustrated total internal reflection was used). These single-pulse parameters are precisely those required in distance measurements.

However, in the pulse-periodic (0.125–1 Hz) regime the output parameters were somewhat poorer, beginning from the third or fourth pulse, when an ‘empty’ laser head was used. The reason for this was an increase in the rate of quenching of the ytterbium ion luminescence by chromium ions at elevated glass temperatures [8].

With a view of finding a highly efficient glass for minilasers operating at high pulse repetition rates, we carried out a search for a new matrix which would have the following properties: an enhanced thermomechanical strength (a low thermal expansion coefficient), a weak tendency to crystallise when the concentration of ytterbium oxide ions was increased right up to $3 \times 10^{21} \text{ cm}^{-3}$, and the minimal concentration of Cr^{2+} . With this in mind, we synthesised a number of experimental phosphate glasses and studied their characteristics. From several compositions with a low thermal expansion coefficient, we selected that which had good technological properties even when the ytterbium oxide concentration by weight exceeded 30%.

We synthesised the samples with different activator concentrations for investigating the spectral, luminescence, and kinetic properties of the chromium, ytterbium, and erbium ions with the aim of optimising their concentrations. We shall now give the results of this investigation.

2. Spectral properties of the chromium ions

One of the most important parameters, which has a decisive influence on the efficiency of Cr–Yb–Er laser glasses, is the rate of back transfer of energy in the Cr–Yb ion pair. As demonstrated by us earlier [6], this rate depends strongly on the composition of the glass and on the conditions during its fabrication. It was established for the first time in Ref. [9]

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that a phosphate glass contains not only Cr^{3+} but also Cr^{2+} ions. The latter are the main agent quenching the luminescence of ytterbium ions and they give rise to additional inactive losses in the wavelength range 0.9–1.5 μm . Depending on the composition of a glass and the conditions during its fabrication, the Cr^{2+} content ranged from 0.1% to 20% of the total amount of Cr. The Cr^{2+} concentration in the LGS-Kh glass could be minimised in order to reduce significantly the rate of the back-transfer process: the reduction was approximately by a factor of 5 compared with the familiar LGSS-0135/01 glass [6]. Consequently, an increase in the contribution of the Cr^{3+} ions to the sensitisation of the erbium luminescence in LGS-Kh was observed up to extremely high chromium ion concentrations, amounting to $(7-8) \times 10^{19} \text{ cm}^{-3}$. As pointed out above, this was extremely important for the fullest utilisation of the flashlamp radiation when small active elements were used.

Measurements carried out on a sample with the ytterbium ion concentration of $1.6 \times 10^{21} \text{ cm}^{-3}$ and with various chromium ion concentrations showed that the rate W_{YbCr} of the back transfer of energy in the Cr–Yb ion pair increased linearly with the concentration N_{Cr} of the chromium ions and the ratio $W_{\text{YbCr}}/N_{\text{Cr}} = 6 \times 10^{-18} \text{ cm}^3 \text{ s}^{-1}$ was 1.5 times less than the corresponding ratio for LGS-Kh. Therefore, the new composition is preferable for coactivation with chromium ions, even compared with LGS-Kh.

The quantum efficiency of sensitisation of the ytterbium ion luminescence by the chromium ions was found to be 0.6 for chromium ion concentrations $(1-9) \times 10^{19} \text{ cm}^{-3}$, confirming the conclusion reached in Ref. [10] that the sensitisation efficiency is independent of the glass composition.

3. Optimisation of the Yb^{3+} concentration

An increase in the Yb^{3+} concentration is desirable for two reasons. When the ytterbium ion concentration is raised up to $N_{\text{Yb}} = 2.5 \times 10^{21} \text{ cm}^{-3}$, the rate of transfer of the excitation energy in the Yb–Er pair increases [11] and so does the absorbed pump energy. In our case, there was no danger that an increase in the ytterbium concentration would give rise to nonuniform pumping of the active element across its diameter, since this diameter was small (2–4 mm). For the glass with the ytterbium ion concentration $2.3 \times 10^{21} \text{ cm}^{-3}$ the average absorption coefficient k was always 7 cm^{-1} , with the exception of a very narrow peak at the 977 nm wavelength. Therefore, for active elements $d = 4 \text{ mm}$ in diameter, the dimensionless coefficient representing the degree of uniformity of the distribution of the pump radiation over the diameter was $kd/2 = 1.4$. The most uniform distribution was obtained when this coefficient was equal to the refractive index of the active medium [12], which in our case was 1.5.

We investigated the dependence of the rate of the back transfer of energy on the parachor ytterbium ion concentration. At ytterbium concentrations up to $1.3 \times 10^{21} \text{ cm}^{-3}$ the rate of transfer in the Yb–Er pair is proportional to the square of the concentration [11]. Since the back-transfer process in the Cr–Yb pair [10], like the forward transfer in the Yb–Er pair, can be described within the framework of the dipole–dipole interaction, one would expect a similar dependence for the rate of the back-transfer process in the Cr–Yb pair. This hypothesis was checked by preparing samples of a new composition with the same concentration of Cr^{3+} , equal to $9 \times 10^{19} \text{ cm}^{-3}$, but with various ytterbium ion concentrations. The kinetics of decay of the ytterbium ion luminescence

after excitation with a pulse from a semiconductor laser ($\lambda = 970 \text{ nm}$) was exponential over the intensity range amounting to two orders of magnitude. We determined the duration τ_{Yb} of the ytterbium ion luminescence at room temperature ($20 \pm 2 \text{ }^\circ\text{C}$). The rate W_{YbCr} was found from

$$W_{\text{YbCr}} = W_{\text{Yb}} - W_{\text{Yb}}^0, \quad (1)$$

where W_{Yb}^0 and $W_{\text{Yb}} = 1/\tau_{\text{Yb}}$ are the rates of decay of the ytterbium ion luminescence in a glass free of chromium ions and in the investigated glass. The rate $W_{\text{Yb}}^0 = 0.7 \times 10^3 \text{ s}^{-1}$ was independent of temperature.

The dependence plotted in Fig. 1 shows that the rate of the back-transfer process in the Cr–Yb pair increases, as expected, with the ytterbium ion concentration, but the increase is not linear throughout the investigated range, $(0.4-2.3) \times 10^{21} \text{ cm}^{-3}$, of the ytterbium ion concentrations. This is most probably associated with the smaller microparameter C_{da} for the Yb–Cr pair, corresponding to the analogous parameter for the Yb–Er pair, which means that the critical concentration of the donor (ytterbium) N_{cr} is reached earlier (at this concentration the migration-accelerated quenching stage changes over to the kinetic stage). In accordance with the ‘hopping’ model, N_{cr} is given by the formula [13]

$$N_{\text{cr}} = \left(\frac{2\pi}{3} \right)^{-1.5} \frac{(C_{\text{da}}/C_{\text{dd}})^{0.5}}{R_{\text{min}}^3}, \quad (2)$$

where C_{da} and C_{dd} are the microparameters of the interaction in the Yb–Cr and Yb–Yb pairs, respectively, related to the excitation transfer rates W_{da} and W_{dd} for these pairs by the expressions $C_{\text{da}} = W_{\text{da}}R_{\text{da}}^6$, $C_{\text{dd}} = W_{\text{dd}}R_{\text{dd}}^6$; R_{min} is the minimum distance of approach between the Cr (acceptor) and Yb (donor) ions.

It can be seen from Fig. 1 that an increase in the ytterbium ion concentration from $1.6 \times 10^{21} \text{ cm}^{-3}$ (which is the concentration in the LGS-Kh glass) to $2.3 \times 10^{21} \text{ cm}^{-3}$ increases the rate of back transfer of energy in the Cr–Yb pair by a factor of just 1.25, whereas when the new glass matrix was used, an overall reduction in the rate of the back transfer process was by a factor of 1.5. Therefore, the ytterbium ion concentration in the new glass was 1.5 times higher compared with LGS-Kh, but this did not increase the overall rate of the back transfer of energy in the Cr–Yb pair. Therefore, it should be possible to maximise the concentrations of the chromium and ytterbium ions. This would make it feasible to develop low-threshold highly efficient radiation sources generating pulses at a low repetition rate.

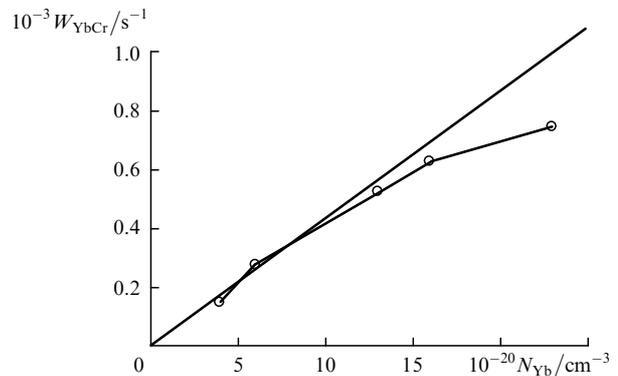


Figure 1. Dependence of the rate of back transfer of energy in the Cr–Yb pair on the ytterbium ion concentration at $T = 20 \text{ }^\circ\text{C}$.

4. Dependence of the back-transfer rate in the Cr–Yb pair on the temperature of a medium

As pointed out above, the rate of the back transfer of energy in the Cr–Yb pair depends on the temperature of the ambient medium and this reduces the energy parameters of the Cr–Yb–Er glass lasers operating in the pulse-periodic regime. In the ‘close packing’ configuration and in the absence of cooling, the active element is heated strongly by the temperature field of the flashlamp. Our measurements indicated that the temperature of the active element with a diameter of 2.5 mm and 35 mm long, pumped by an INP2-3/35 flashlamp at a pulse repetition rate 1/4 Hz to deliver an energy of 8 J per pulse, exceeded the room temperature by 50 °C–60 °C when steady-state operation was reached (5 min after switching on). The steady temperature was almost independent of the chromium ion concentration in the glass active element.

The temperature dependence of W_{YbCr} was determined for samples with the ytterbium ion concentration $2.3 \times 10^{21} \text{ cm}^{-3}$ and with various chromium ion concentrations. Samples were placed in a thermostat and the duration of the ytterbium ion luminescence was measured at various temperatures. At each temperature we determined, in accordance with formula (1), the rate W_{YbCr} . The dependences obtained are plotted in Fig. 2.

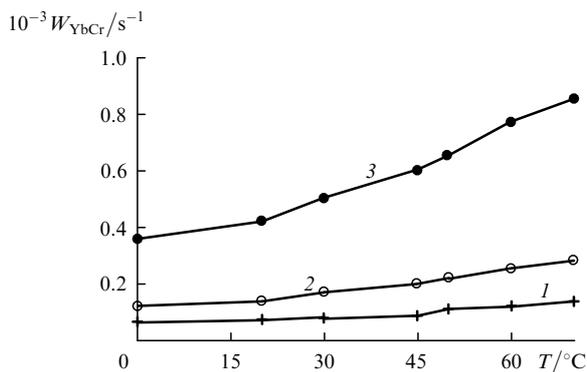


Figure 2. Temperature dependences of the rate of back transfer of energy in the Cr–Yb pair, obtained for $N_{Cr} = 10^{19} \text{ cm}^{-3}$ (1), $2 \times 10^{19} \text{ cm}^{-3}$ (2), and $6 \times 10^{19} \text{ cm}^{-3}$ (3); $N_{Yb} = 2.3 \times 10^{21} \text{ cm}^{-3}$.

A simple calculation utilising familiar relationships governing the quantum efficiency of sensitisation η_s in the Yb–Er pair [14] shows that, when the erbium ion concentration is $1.6 \times 10^{19} \text{ cm}^{-3}$ in a glass with the chromium ion concentration $6 \times 10^{19} \text{ cm}^{-3}$, the initial value of η_s decreases from 0.86 to 0.83 when temperature is increased from 20 °C to 60 °C and the value of η_s corresponding to the lasing threshold falls from 0.75 to 0.68 (when the reflection coefficient of the output mirror is 85%). Since η_s is the decisive factor governing the efficiency of storage of an inverted energy in erbium glass, its decrease reduces the energy parameters of the laser.

We thus found that an increase in the back-transfer rate in the Cr–Yb pair with increase in temperature was the only factor which restricted the chromium ion concentration in the new glass to be used in pulse-periodic lasers.

5. Selection of the erbium ion concentration

In a three-level lasing scheme the erbium ion concentration should be as low as possible. Calculations show that, if the output mirror has the reflection coefficient $R > 80\%$ and the length of the active element is 25–45 mm, the lowest threshold pump energy corresponds to the erbium ion concentration $(0.5–0.6) \times 10^{19} \text{ cm}^{-3}$. However, we should bear in mind that the rate of sensitisation of the erbium ions by the ytterbium ions is proportional to the erbium ion concentration and, consequently, at low values of these concentrations the quantum efficiency of energy transfer to erbium is low, which has a negative effect on the differential efficiency of free running and on the inversion energy in the Q-switched regime. For example, when the erbium ion concentration was reduced from $1.6 \times 10^{19} \text{ cm}^{-3}$ to $5 \times 10^{18} \text{ cm}^{-3}$ the differential efficiency in the free-running regime decreased by a factor larger than 1.6.

Selection of the erbium ion concentration is thus a very important task and, as shown in practice, for the majority of flashlamp-pumped lasers the optimal glass has the erbium ion content $(1.0–2.0) \times 10^{19} \text{ cm}^{-3}$ and the necessary output parameters are reached by altering the volume of the active element.

6. Composition of the new LGS-KhM glass

Our investigation led to the development of a new phosphate glass, which we called LGS-KhM. The base activator concentrations in the new glass are $[Yb^{3+}] = 2.3 \times 10^{21} \text{ cm}^{-3}$, $[Er^{3+}] = 1.6 \times 10^{19} \text{ cm}^{-3}$, and $[Cr^{3+}] = 2.2 \times 10^{19} \text{ cm}^{-3}$. The main characteristics of the LGS-KhM glass are as follows:

Emission wavelength (nm)	1535
Luminescence duration (ms)	8.5
Stimulated-transition cross section (10^{-20} cm^2)	0.75
Refractive index n_d	1.543
Refractive index at $\lambda = 1535 \text{ nm}$	1.532
Softening temperature (°C)	480
Thermal expansion coefficient	
in the range 20 °C–40 °C ($^{\circ}\text{C}^{-1}$)	87×10^{-7}
Density (g cm^{-3})	3.01
Solubility in water (weight loss) at $T = 100 \text{ }^{\circ}\text{C}$ after 1 h (%)	0.024

We also produced two modifications of this glass, differing from the base composition by the concentration of the Cr^{3+} ions: LGS-KhM/CR0.9 ($N_{Cr} = 0.9 \times 10^{19} \text{ cm}^{-3}$) and LGS-KhM/CR5.6 ($N_{Cr} = 5.6 \times 10^{19} \text{ cm}^{-3}$). The thermal expansion coefficients of these glasses are $87 \times 10^{-7} \text{ }^{\circ}\text{C}^{-1}$ (in the range 20 °C–40 °C), compared with $127 \times 10^{-7} \text{ }^{\circ}\text{C}^{-1}$ for LGS-Kh. This reduction in the thermal expansion coefficient should increase the thermomechanical strength of glass by a factor of about 2.5 [15].

7. Lasing characteristics of the LGS-KhM glass

In the lasing tests we used active elements 2.5 mm in diameter and 35 mm long made of LGS-KhM and of its modifications LGS-KhM/CR0.9 and LGS-KhM/CR5.6. The tests on these elements were made under the following conditions. A cavity, with the base length 100 mm, was formed by a plane output mirror with the reflection coefficient 84% and a nontransmitting spherical mirror with a radius of curvature $R = 50 \text{ cm}$

located in such a way that the distance between the latter mirror and the laser head made it possible to insert an intracavity modulator with frustrated total internal reflection.

The laser head was a cylindrical quartz tube 14 mm in diameter, silvered on the outside, and containing an INP2-3 flashlamp 35 mm long and an active element. For convenience of its replacement, the active element was placed loosely inside a thin-walled (outer diameter 4 mm) quartz tube kept at a fixed position in the laser head. The mass of the laser head containing the active element, the flashlamp, and the supports was about 15 g; there was no cooling, except through the surface of the head. Pumping was provided by a PS1210OP power supply, which maintained a constant voltage across a capacitor with an error of 1% and which fed a discharge circuit assembled in such a way that the light pulse emitted by the flashlamp was dome-shaped with the duration 1 ms at 0.1 of the amplitude. The output energy was measured with an IMO-2N power meter.

Fig. 3 gives the energy characteristics of the active elements operating in the free-running regime and also in the Q -switched single-pulse regime. The new LGS-KhM glass was superior to LGS-Kh in respect of the output energy (lasing threshold 6 J, differential efficiency 2.2%) in spite of the 2.5 times higher chromium ion concentration. A further reduction in the chromium ion concentration increased the threshold energy of lasing and reduced the laser efficiency, so that for LGS-KhM/CR0.9 the lasing threshold was 6.6 J and the differential efficiency was 1.9%. In the single-pulse regime it would be preferable to use the glass with the higher chromium ion concentration (LGS-KhM/CR5.6 modification) which had a record-low lasing threshold of 5 J and a differential efficiency of 2.6%.

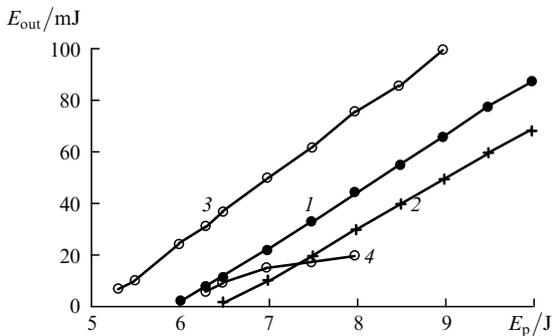


Figure 3. Dependences of the output energy E_{out} on the pump energy E_p for the LGS-KhM glass (1) and for its modifications LGS-KhM/CR0.9 (2) and LGS-KhM/CR5.6 (3), obtained for a single pulse under free-running conditions, and also for the LGS-KhM glass under Q -switching conditions (4).

Tests on the active elements were made in the pulse-periodic free-running regime at pump energies needed to generate a single pulse with an energy of about 15 mJ. The results are presented in Fig. 4. Three series of results were obtained at repetition rates 0.125–0.5 Hz and five series of measurements were made at 1–2 Hz. The interval at which a series was repeated was selected to ensure that the active element cooled to room temperature after each series. The dependences obtained demonstrated clearly the advantages of the new LGS-KhM glass in the pulse-periodic regime. For example, when the pulse repetition rate was 1 Hz, the fall of the laser output energy by the tenth pulse was 20%

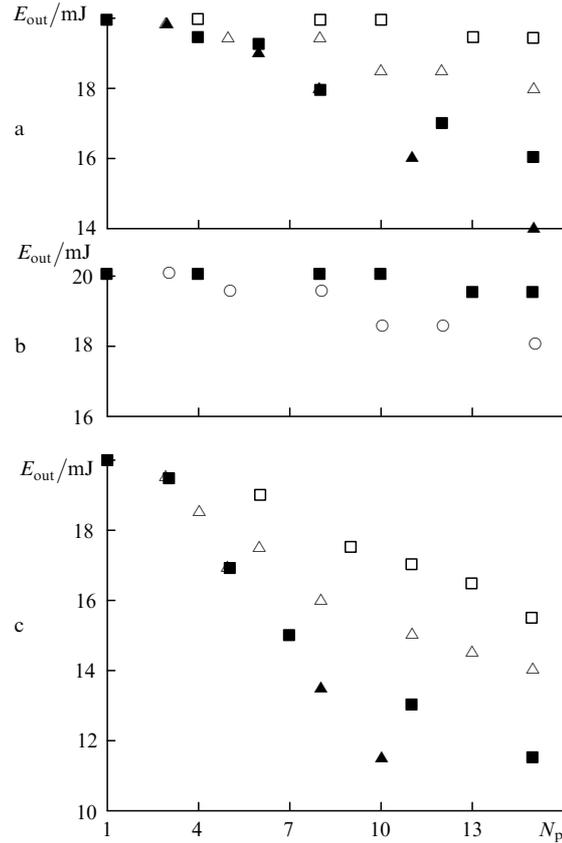


Figure 4. Dependences of the output energy E_{out} on the consecutive number N_p of a pulse in a series, obtained for the LGS-KhM glass (a) and for its modifications LGS-KhM/CR0.9 (b) and LGS-KhM/CR5.6 (c) at the following pulse repetition rates (Hz): 0.125 (\square), 0.25 (\triangle), 0.5 (\blacksquare), 1 (\blacktriangle), and 2 (\circ).

for LGS-KhM (Fig. 4b), whereas for LGS-Kh (and also for LGS-KhM/CR5.6) the fall was 50% (Fig. 4c). However, for the glass with the lower chromium ion concentration (LGS-KhM/CR0.9, see Fig. 4a) at the repetition rate up to 2 Hz there was no fall in the output energy for a series limited to 10 pulses. At pump energies exceeding the threshold by a factor of 1.5 the product Dq (D is the beam diameter and q is the divergence angle) was ~ 10 mm mrad and it was nearly independent of the pulse repetition rate for all the glasses.

The properties of the new erbium-doped phosphate glass LGS-KhM developed at our Institute — characterised by a low thermal expansion coefficient, an enhanced thermo-mechanical strength, and good chemical stability in combination with its excellent laser characteristics — should make this glass very suitable for devices with forced cooling operating at high repetition rates (tens of hertz) and output powers (tens of watts). Investigations of the feasibility of such operation are proceeding at present and the results obtained will be published separately.

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