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GENERATION OF PICOSECOND PULSES IN SOLID-STATE LASERS USING NEW ACTIVE MEDIA

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Results are reported of investigations aimed at generating nanosecond radiation pulses in solid-state lasers using new active media having broad gain lines. Passive mode locking is accomplished for the first time in a BeLa:Nd^{3+} laser at a wavelength 1.354 μm, and in a YAG:Nd^{3+} laser on a 1.32-μm transition. The free lasing and mode-locking regimes were investigated in an alexandrite ($BeAl_2O_4$:Cr^{3+}) laser in the 0.72-0.78-μm range and in a synchronously pumped laser on F_2^- centers in LiF in the 1.12-1.24-μm region. The features of nonlinear perception of IR radiation by the eye, using a developed picosecond laser on F_2^- centers, are investigated for the first time.

The interest in picosecond light-pulse sources that are tunable in a wide range of wavelengths is due to the fundamentally new opportunities they offer in high-time-resolution spectroscopy for the investigation of nonstationary processes in solids and of the kinetics of chemical and biological processes, and also in many technical applications such as high luminosity information transmission, radar, etc. [1]. Progress in this field is due mainly to the availability of laser systems that can give reproducible parameters of ultrashort pulses (USP) of picosecond and subpicosecond duration, with the lasing wavelength tunable in a specified band.

At the present time USP are obtained mainly by using active or passive (or a combination of both) locking of the modes of the laser cavity by modulating the loss or gain at a multiple of the intermode frequency. The greatest success in this direction was reached in neodymium lasers with passive mode locking by saturable absorbers, and with synchronous pumping of dye lasers by argon lasers. It is just in the latter case that the record $60 \cdot 10^{-15}$ sec was reached in the cw regime [2], using an active medium with a broad gain line, by introducing into the cavity an additional saturable absorber.

It is known that the pulse duration in the mode-locking regime is inversely proportional to the width of the lasing spectrum, i.e., it depends on the gain linewidth of the active element. There is therefore no doubt that it would be beneficial to find new active media with broad gain lines in various sections of the spectrum, and to find new methods of shortening the UPS duration both with an aim at obtaining femtosecond light pulses and at expanding the active wavelength ranges of such lasers.

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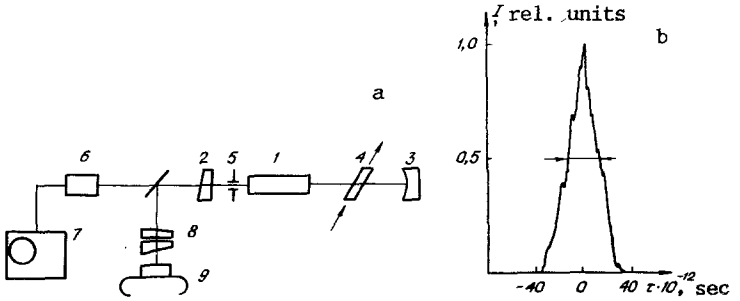


Fig. 1. a) Diagram of experimental setup: 1) active element; 2, 3) mirrors; 4) cell with solution of saturable absorber; 5) diaphragm; 6) coaxial photocell; 7) oscilloscope; 8) Fabry–Perot interferometer; 9) photographic camera. b) Correlation function measured on the basis of vector generation of the second harmonic of 1.32-μm radiation.

We report here our first results of research aimed at obtaining picosecond emission pulses from solid-state lasers with new active media having broad gain lines. These media include lanthanum beryllate with neodymium ($La_2Be_2O_5:Nd^{3+}$) at the wavelength 1.35 μm, yttrium aluminum garnet with neodymium ($YAG:Nd^{3+}$) on the 1.318-μm transition, alexandrite ($BeAl_2O_4:Cr^{3+}$), and lithium fluoride with F_2^- centers.

YAG:Nd^{3+} LASER ON 1.318 μm WAVELENGTH

In our experiments with an $YAG:Nd^{3+}$ laser we accomplished for the first time ever passive mode locking on a transition of wavelength 1.318 μm using a saturable organic dye. This method of obtaining USP in this spectral region turned out to be more effective than the methods previously known, which used synchronous pumping of parametric generators and lasers with dye centers. Mode locking on the $^4F_{3/2}$-$^4I_{13/2}$ transition (transition cross section $\sigma = 1.65 \cdot 10^{-20}$ cm^2, line width $\Delta\nu = 6.5$ cm^{-1}) became possible in a neodymium laser through the use of the new pentacarbocyanin dye 3620u [3]. The previously known type-1057 organic dyes have relaxation times 10^{-8} sec and are suitable only for obtaining giant pulses [4]. Dyes of the pentacarbocyanine series are promising for passive mode locking of lasers in the 1.2-1.4-μm range, since they have intense absorption bands in this spectral range and have relaxation times $t_r \sim 10^{-11}$ sec.

In the experimental setup (Fig. 1a) the active 5.65-mm-diameter element 1, with nonreflecting end faces, was mounted at an angle 3° to the axis of a cavity 60 cm long, consisting of a flat output mirror 2 and a 300-cm rear mirror 3. The transmittances were $T_{1.064} = 0.86$; $T_{1.318} = 0.15$ for mirror 2 and $T_{1.064} = 0.8$; $T_{1.318} = 0.05$ for mirror 3. In such a selective cavity, the 1.064-μm line did not lase even at a tenfold excess of pump above threshold. A solution of dye in dichloroethane flowed at a rate of 10 liter/min through cell 4, 1 mm thick, located 40 mm away from the rear mirror and inclined at the Brewster angle. The choice of the solvent was governed by the requirement imposed on the absorption coefficient and on the nonlinear refractive index at the operating wavelength, viz., $\alpha \sim 0.01$ cm^{-1} and $n_2 < 10^{-12}$ CGS. Diaphragm 5 of 1.5-mm diameter suppressed the nonaxial modes. The durations of the USP were measured by the vector second-harmonic generation technique [5]; the spectrum was recorded with a 1-mm Fabry–Perot interferometer; the form of the pulse train and the type of mode-locking regime was observed on the screen of an S1-75 oscilloscope with 3-nsec resolution. In the experiments we varied the pump intensity, the dye concentration, and the location of the active element in the cavity.

It should be noted that a $YAG:Nd^{3+}$ laser operating at a wavelength 1.318 μm has a tendency to partial mode locking without a nonlinear absorber, but a feature of such a regime would be nonreproducibility of the parameter. A stable USP generation regime was observed when the initial transmission of the cell with absorber was lower than 0.8. The main results were obtained when the ratio of the cross-sectional areas of the beam in the laser medium and in the cell, which was governed by their relative placement in the cavity, was equal to 1.73. The so-called two-threshold locking regime was observed at a pump energy 16-17 J. In this case the pulse-train duration was about 300 nsec, and the average radia-

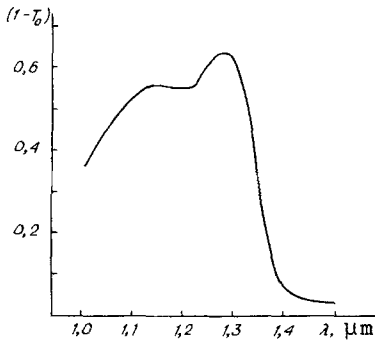


Fig. 2. Absorption of 3620u dye in dichloroethane in the 1.1-1.5-μm region.

tion power at a pulse repetition frequency 12.5 Hz reached 120 mW. Using as the frequency doublet an $LiIO_3$ crystal 10 mm long, the average second-harmonic emission power at 0.659-μm wavelength exceeded 10 mW. The last result can be successfully used for synchronous pumping of dye-center lasers, e.g., to obtain USP of tunable wavelength in an LiF crystal with F_2^+ centers.

When the dye concentration in the absorbing cell was increased to the initial transmittance $T_0 \approx 0.5$ and the pump power was simultaneously increased to 40 J, the duration of the pulse train decreased to 80-100 sec, and the peak pulse power increased to 3.5 MW. Figure 1b shows the correlation curve obtained by the vector second-harmonic generation technique. The average pulse duration at half maximum of this curve, assuming a Gaussian profile, was $(28\text{-}30)\cdot10^{-12}$ sec. The emission spectrum, of width $\Delta\nu \sim 1.5$ cm^{-1}, had a smooth profile, with no self-modulation effects or broadening of the spectrum observed, in view of the small value of n_2 of the active medium in the dichloroethane.

From the data obtained and from the absorption spectrum (Fig. 2) we can estimate certain quantities that describe the 3620u dye used in our experiments. The coefficient of molar extinction at the absorption maximum (1.284 μm) of the solution was $\varepsilon_{1.284} = 14.6\cdot10^4$ liter/(mole·cm). The cross section for dye absorption at 1.318 μm, calculated from the formula $\sigma_d(\lambda) = 2.303\cdot\varepsilon(\lambda)/N_0$, where N_0 is Avogadro's number, with account taken of the absorption line shape, is equal to $48\cdot10^{-17}$ cm^2. Knowing the output radiation power in the two-threshold mode-locking regime and the cavity parameters, we estimated the saturation intensity of the 3620u dye to be $I_{sat} \approx 65$ MW/cm^2. The dye relaxation time, calculated from the formula $\tau_{rel} \approx \hbar\omega/2\sigma_K I_{sat}$, where ω is the frequency of the absorbing transition, turned out to be $3.3\cdot10^{-12}$ sec.

These values enable us to determine the initial dye-solution transmittance needed to achieve the two-threshold regime, using the criterion $\sigma_{1,318}T/\sigma_K\tau_{rel} \geq (\varkappa_0^2/\varkappa_0+\gamma)2/3\cdot\sqrt{3}$, where γ is the linear loss in the cavity, T is the time of passage through the cavity, and $\varkappa_0$ is connected with the initial cell transmittance T_0 by the relation $T_0 = \exp(-1/2\cdot\varkappa_0)$. Therefore at $T = 4.4\cdot10^{-9}$ sec and $\gamma = 1.9$ the two-threshold regime is reached at $T_0 \geq 0.6$, in good agreement with the experimental data. In addition to all the foregoing we note the high photochemical endurance of the employed organic dye — no degradation of the dye was noted during the experiments.

BeLa:Nd^{3+} LASER WITH WAVELENGTH 1.354 μm

Neodymium-activated lanthanum beryllate has been little investigated as a laser material, but even the first reports point to its undisputed superiority to garnet when it comes to generating USP.

The basic beryllate and garnet characteristics of interest for laser systems are listed in Table 1. We must call attention to the following features: First, there is a substantial difference between the gain lines, making the beryllate a preferred active medium for USP generation [7-9]. This applies primarily to the $^4E_{3/2}$-$^4I_{13/2}$ transition, for which the beryllate cross section is not smaller than that of the garnet. Second, the optical endurance of the BeLa:Nd^{3+} crystal exceeds substantially that of all known laser materials, a fact of particular importance when operating in the mode-locking regime. A final im-

TABLE 1

Laser material	Transition	Emission wavelength	Luminescence linewidth	Transition cross section	Excitation threshold
		μm	cm^{-1}	$cm^2 \cdot 10^{-19}$	J
BeLa:Nd^{3+}	$^4F_{3/2}-^4I_{11/2}$	1,069	30	2,1	1,8
BeLa:Nd^{3+}	$^4F_{3/2}-^4I_{11/2}$	1,079	38	1,5	3,0
BeLa:Nd^{3+}	$^4F_{3/2}-^4I_{13/2}$	1,354	36	0,22	2,5
YAG:Nd^{3+}	$^4F_{3/2}-^4I_{11/2}$	1,064	6	7,9	1,0
YAG:Nd^{3+}	$^4F_{3/2}-^4I_{13/2}$	1,318	6.5	0,15	1,2

portant factor is the nonlinear refractive index, which determines the onset of undesirable nonlinear effects, such as self-focusing of the radiation, phase self-modulation, and others. It can be seen that even with respect to this property the beryllate is superior to the garnet.

We shall describe here the results of an investigation of passive mode locking at a wavelength 1.354 μm using the saturable dye 3620u. The experimental setup is quite similar to that shown in Fig. 1a. An $La_2Be_2O_5:Nd^{3+}$ crystal with an approximate neodymium density 2 wt. %, used as the active element, was 6 mm in diameter 70 mm long, with E‖x. The cavity 0.5 m long consisted of flat output window 7 with transmittances $T_{1.07} = 0.86$; $T_{1.35} = 0.15$ and a spherical 2-m mirror 3 with $T_{1.07} = 0.8$; $T_{1.35} \leqslant 0.005$. The solution of the dye in trichloroethane was drawn through the cell 4.

As already noted, it is important to choose correctly the initial transmittance of the filter and the ratio of the beam diameters in the active element and the filter, $\delta = \omega_f/\omega_a$. In the geometry chosen, this parameter was $\sigma \sim 3.4$. The dye transmittance optimal for USP formation was estimated in accordance with the criterion [6] $\sigma_{1.35}T/\sigma_f\tau_{rel} \geqslant \frac{x_0^2 2}{(x_0+\gamma)3\sqrt{3}}$, where $\sigma_{1.35}$ and σ_f are the cross sections for the transitions of the active medium and the absorber, respectively, while T and τ_{rel} are the temporal characteristics of these media. For the active elements used in our experiments, the coefficient T_0 (the initial transmittance of the cell) should exceed 0.5, as was convincingly confirmed by all our experiments.

At a 1.354-μm wavelength and at an initial saturable-filter transmittance $T \approx 0.75$, we obtained a train of pulses with total duration about 100 nsec and average picosecond-pulse duration measured by a correlation procedure $\tau \sim 15 \cdot 10^{-12}$ sec (Fig. 5). The diffuse spectrum observed was $\Delta\lambda \sim 1.2$ nm wide, so that $\Delta\nu\tau \sim 2$. The overestimate of the last parameter is due to the need for working at an initial filter transmittance higher than optimal, since the optical endurance of the interference coatings of the cavity elements is limited.

Measurements using single-stage amplification of the USP emission were performed with other less concentrated crystals on a transition with 1.069-μm wavelength. The best results were obtained in a cell containing a solution of 3274u dye in ethyl alcohol at an initial transmittance $T_0 \sim 0.7$. In the near-threshold regime, the USP trains had a 40-nsec duration at an average power 40 mW at a 12.5-Hz frequency and an individual pulse duration $(13\text{-}18)\cdot 10^{-12}$ sec. This corresponds to a single pulse energy 0.6 mJ and a $4\cdot 10^7$-W peak power. When amplified by a second analogous element the average train power reached 230 mW, corresponding to a single-pulse power $2\cdot 10^8$ W.

The gain curve (Fig. 4) can be described by the expression $E_{out} = E_{sat}\ln[1 + G_0(\exp E_{in}/E_{sat}-I)]$ at $E_{in} \ll E_{sat}$, where G_0 is the weak-signal gain and E_{sat} it the saturation-energy density. It can be seen from Fig. 4 that no saturation is reached, so that $E_{out} = 0.0026E_{in}$.

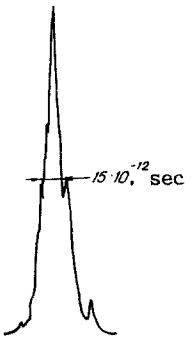


Fig. 3

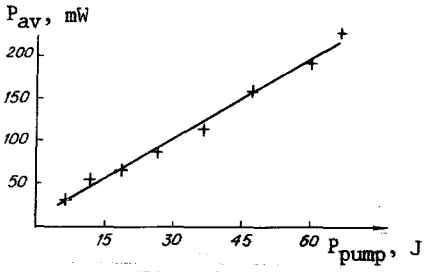


Fig. 4

Fig. 3. Correlation function, from vector second-harmonic generation method, of 1.345-μm radiation in a BeLa:Nd^{3+} laser.

Fig. 4. Average power of amplified USP in BeLa:Nd^{3+} vs the pump energy at a 12.5-Hz repetition frequency.

ALEXANDRITE LASER OPERATING ON VIBRONIC TRANSITION

Alexandrite – $BeAl_2O_4:Cr^{3+}$ – holds a special position among the recently synthesized laser materials. Chrysoberyl as a matrix in which the Al^{3+} ions are replaced by Cr^{3+} ions is the third crystal, after sapphire and garnet, in which lasing by chromium ions was obtained. Lasing was achieved in alexandrite not only on the R line but also, most importantly, on the vibronic transition $^4T_2$-$^4A_2+\hbar\omega$. This, first, uncovered a possibility of smooth tuning the frequency over a wide spectral amplification band, 0.7-0.8 μm. Second, the laser, operating in a four-level scheme, has a relatively low pump threshold and a high efficiency at room temperature. The efficiency is increased when the temperature is raised to 70°C, and this facilitates the development of alexandrite laser systems operating in the cw regime or at high pulse-repetition frequency.

In our study we investigated the free-lasing and USP regimes with passive mode locking. The level scheme of the Cr^{3+} ions in alexandrite (Fig. 5) shows a broad band of the laser vibronic transition $^4T_2$-$^4A_2$, whose cross section at room temperature is $\sigma \sim 7\cdot10^{-21}$ cm^2 and increases to $2\cdot10^{-20}$ cm^2 at 475°K. On the R line (^2E-$^4A_2$ transition) the transition cross section $3\cdot10^{-19}$ cm^2 is considerably higher than in ruby.

Chrysoberyl has an orthorhombic structure, the gain depends on the crystal orientation, and the laser emission is linearly polarized without the use of any polarizers in the cavity.

The experimental setup is perfectly similar to that shown in Fig. 1a. The active element of 6-mm diameter and 70-mm length, with chromium density $1.6\cdot10^{19}$ cm^{-3}, had nonreflecting end faces; the INP 5/60 pumping flash lamp was placed in a single-block quartz illuminator. The ultraviolet part of the pump, shorter than 350 nm, which is in the inactive-absorption region of alexandrite and lowers the laser efficiency because of parasitic photochemical and thermal processes in the active element, was cut off by an aqueous solution of sodium nitrate and a thin Pyrex tube. Experience has shown that the use of such a filter protects the working crystal from radiation damage by the pump light.

The axis of the active-element rod coincided with the [001] crystallographic direction of alexandrite; the stimulated emission was polarized along the [010] direction. In the first experiments we used a laser cavity made up of a nontransmitting mirror with curvature radius 300 cm and a flat output mirror with transmittance 0.03 (each mirror had a 3° wedge). The distance between mirrors was 500 cm. In the free-lasing regime the pump threshold in the nondispersive cavity was 48 J, the same value as in [11]. The lasing spectrum was smooth and had a width 5-6 nm centered close to 750 nm. An average output power 1.1 W was obtained at pump energy and pulse duration 80 J and 250 μsec, pulse repetition frequency 12.5 Hz, and output mirror transmittance 0.08.

Tuning in the 718-780-nm range in a TEM_{00} mode generation regime was made possible by placing in the cavity a Lyot filter (Fig. 6).

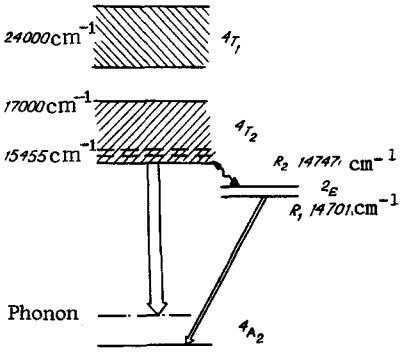


Fig. 5

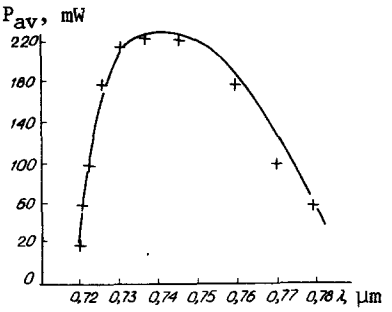


Fig. 6

Fig. 5. Level scheme of Cr^{3+} ions in alexandrite ($BeAl_2O_4$). Zero-phonon lines R_1 ($\sigma = 3\cdot10^{-19}$ cm^2) and R_2 ($\sigma = 6.5\cdot10^{-20}$ cm^2) at $\tau = 1.5\cdot10^{-3}$ sec. Laser electron–phonon transition $^4T_2$-$^4A_2$ ($\sigma = 7\cdot10^{-21}$ cm^2 at 300°K, $\tau = 250\cdot10^{-6}$ sec).

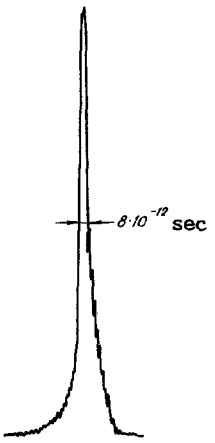
Fig. 6. Average output power vs lasing wavelength in an alexandrite laser at a 12.5-Hz repetition frequency.

It should be noted that at high excitation levels there are produced in the active element appreciable thermal stresses that produce a thermal lens. The focal length of this lens was measured with a telescope-collimated He–Ne laser beam, recorded on photographic film by a 1-m lens at various pump levels. The dependence of the effective focal length of the thermal lens on the average excitation power is approximated in our case by the expression $F(m) = 1.5/P_{cp}^{1}{}^{2}(kW)$. The induced thermal birefringence can be neglected, since it is less than the natural one, $\Delta n \sim 0.002$, as verified by polarization interference in crossed polarizers. To compensate for the thermal effect the cavity construction was modified by replacing the concave mirror with 300-cm curvature radius by a convex one with 200-cm radius. The laser output power increased to 2.5 W.

Mode locking in the alexandrite laser was effected by two saturable absorbers DS1 and DTTS, whose relaxation times in ethanol solutions were $22\cdot10^{-12}$ sec and $130\cdot10^{-12}$ sec, respectively [1]. The DS1 abosrption band, with half-width 40 nm, has a maximum near 709 nm if alcohol is used as the solvent; this makes its use as a nonlinear absorber difficult. The absorption contour of DS1 could be shifted by 8 nm to the long-wave region by replacing the ethanol by dimethylsulfoxide, thus making mode locking possible in the 700-750-nm band. For absorption at wavelengths exceeding 750-nm one can use with success the DTTS dye in ethyl alcohol, even though it has a relatively long relaxation time.

The cell with the saturable dye, oriented at the Brewster angle, was placed ahead of the nontransmitting mirror; the dye solution was drawn at a rate of 10 liter/min. The lasing wavelength was tuned with a Lyot filter, and the TEM_{00} modes were separated with a diaphragm. The end faces of the alexandrite crystal were tilted 2-3° relative to the cavity axis. The degree of the mode-locking regime was set by varying the density of the dye solution and the pumping level. The envelope of the pulse train and the locking regime were monitored by an FÉK-09 coaxial photocell and an oscilloscope with a common time resolution 3 nsec. The average duration of the USP in the train was measured, as in all our other investigations, by the vector second-harmonic generation technique. In the two-threshold regime with DS1 in dimethylsulfoxide we obtained a train duration 200-300 nm with average pulse duration 0.02 nsec. In the near-self-stabilization regime [12], using a somewhat modified cavity geometry, when the USP were produced by a sequence of free-lasing spikes, the average USP was decreased to 8 psec (Fig. 7). Appreciable modulation of the train envelope was observed in this case. The tuning range achieved with the DS 1 absorber was 725-745 nm at a spectrum width 0.2 nm.

When a DTTS solution in ethanol was used, USP of 0.09-nsec duration were generated in the 750-765-nm region. The average output power of a laser with DS1 was 20 mW at 735-nm wavelength. Raising the crystal temperature to 50°C (as monitored by a differential thermocouple on the rod) made the mode-locking regime more stable in view of the increased gain of the active medium.



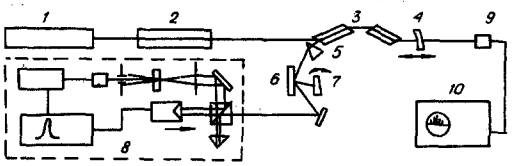


Fig. 7 Fig. 8

Fig. 7. Correlation function, from the vector second-harmonic generation method, of alexandrite-laser emission at 730 nm.

Fig. 8. Diagram of laser operating on F_2^- centers in LiF and synchronously pumped by YAG:Nd^{3+} and BeLa:Nd^{3+} lasers: 1) laser; 2) amplifier; 3) crystals; 4, 7) mirrors; 6) diffraction grating; 9) coaxial photocell; 10) oscilloscope; 8) correlimeter.

Doubling the laser frequency may be of interest. When the emission of an alexandrite laser operating in the free-lasing regime was focused in an $LiIO_3$ crystal (ℓ = 10 mm) the average emission power at 375-nm wavelength exceeded 5 mW.

By measuring the dependence of the laser output power on the pump energy at various transmittances of the output mirror we were able to calculate the lasing losses, which amounted to $\rho \sim 0.008$ cm^{-1}. The estimated maximum gain is 0.01 cm^{-1}. Clearly, further research into alexandrite lasers calls for a crystal with considerably higher chromium-ion density.

LASER ON F_2^- CENTERS IN AN LiF CRYSTAL, SYNCHRONOUSLY PUMPED BY A YAG:Nd^{3+} LASER

The only lasers of tunable wavelength capable of generating picosecond pulses in the near-infrared were until recently dye lasers [13] and parametric generators [14]. The advent of F-center lasers led to the production of a number of tunable USP lasers operating in the wavelength bands 1.24-1.45 μm ($KF:F_2^+$) [15], 2.5-2.8 μm (KCl:Li-F_A(II)) [16] and 0.88-0.98 μm ($LiF:F_2^+$) [17].

We report in this section investigations of picosecond pulses in the spectral region 1.1-1.25 μm in an LiF(F_2^-) laser synchronously pumped by a YAG:Nd^{3+} or BeLa:Nd^{3+} laser operating in the USP regime at 1.06-nm wavelength.

A diagram of the setup is shown in Fig. 8. The emission of a neodymium laser in the form of a train of USP of 40 nsec duration and pulse-repetition frequency 12.5 Hz was fed to a single-stage amplifier. The output energy was 45-50 mJ. Mode locking in the laser was by using a solution of 3955 dye in ethyl alcohol. The average duration of the train pulses was 0.032 nsec. The contrast of the picosecond pulses exceeded 1000.

The cavity of the F_2^- laser comprised a 2-m dichroic mirror with transmittance $T_{1.064}$ = 0.8, a prism, a 600-cm^{-1} diffraction grating, and a flat nontransmitting mirror. Both mirrors had a reflectance 0.9 in the 1.1-1.3-μm range. The LiF(F_2^-) crystals with Brewster-angle end faces had a total length 60 or 160 mm (two variants). Pumping was in a noncollinear geometry with the beams intersecting at an angle 3-4°. The F_2^- laser wavelength was tuned by rotating mirror 7, and the emission was extracted in the zeroth order of the diffraction grating. The temporal characteristics of the radiation were measured by the procedure and apparatus described above. The emission spectrum was investigated with a diffraction spectrograph having a resolution 0.35 nm/mm.

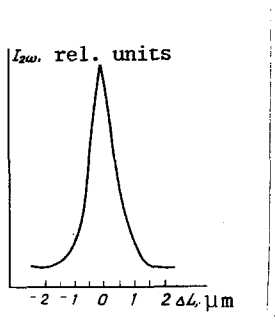


Fig. 9

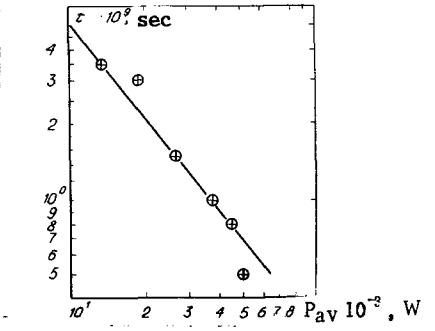


Fig. 10

Fig. 9. Average lasing power of F_2^- laser in LiF vs deviation $\Delta l = \pm(L_F - L_p)$ of cavity length.

Fig. 10. Delay of lasing by F_2^- centers vs average pump-laser power (pulse repetition frequency 12.5 Hz).

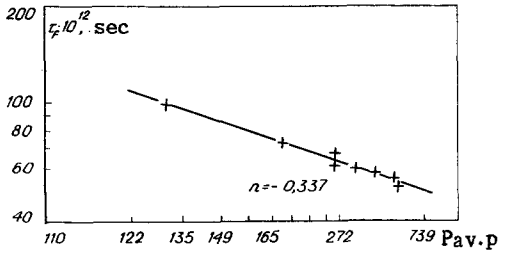


Fig. 11. USP duration in an F_2^--center laser vs the average pump-laser power (repetition frequency 12.5 Hz).

When the F_2^- laser gain was modulated at a frequency equal to or a multiple of the intermode beat frequency, the laser and pump pulses were synchronized by smooth displacement of mirror 4 along the cavity axis. Two ratios of the lengths L_p and L_F of the pump-laser and F_2^- laser cavities were investigated, $L_p = L_F$ and $L_p = 2L_F$. One and two pulses generated on the axial period in the first and second case, respectively, as monitored by the oscilloscope. The onset of locking when the cavity lengths were matched was revealed by a "power resonance" of the F_2^- laser emission (Fig. 9) on doubling of the frequency in the $LiIO_3$ crystal [18].

The appreciable width of the "resonance" is due to the limited number of pump pulses and to the limited lasing evolution time. Just as in [19], a delay of the F_2^- laser pulses relative to the pump train was observed for both cavity-length ratios. According to our measurements the delay time τ_d depends on the excess of the pump power above threshold. The observed dependence is described by the formula $\tau_d \approx P_{av.p}^{-0.54}$ (Fig. 10).

It is known that in the case of synchronous pumping the train-pulse duration depends on the gain spectrum width and modulation depth as well on the number of passes through the cavity during the train evolution time. At fixed gain-spectrum width and pump-pulse duration, the duration of the lasing pulse is given by [20]: $\tau_F^3 = \tau_p/(\Delta\nu)^2 \ln(g_0/\alpha)$, where τ_p is the pump-pulse duration, $\Delta\nu$ the gain linewidth, g_0 the gain, and α the loss. The experimental results (Fig. 11) agree well with the theoretical ones and show that for our case of nonstationary lasing the functional dependence is of the form $\tau_F \sim (\ln P_{av\cdot p})^n$, where $n = -0.337$ at a lasing spectrum width $\Delta\nu_g = 8$ cm^{-1}. The dependence of τ_F on the spectrum width was not investigated by us more thoroughly, since it is obviously necessary to increase the duration of the pump-pulse train to obtain a deeper modulation of the F_2^- laser gain.

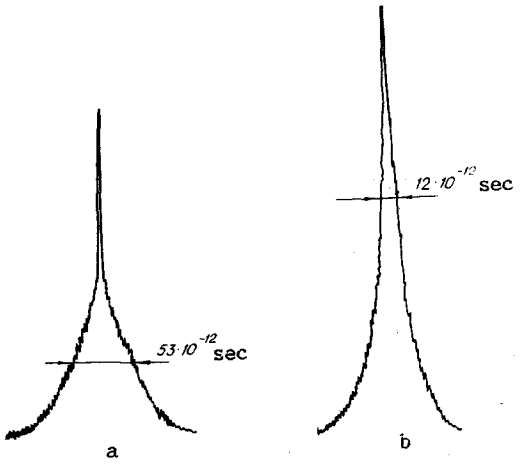


Fig. 12

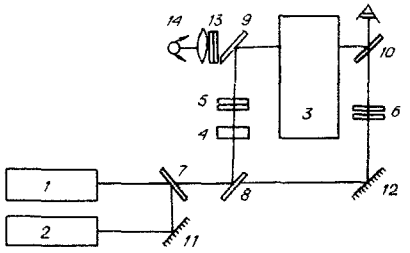


Fig. 13

Fig. 12. Second-harmonic-generation correlation curves for partial (a) and total (b) mode locking of a laser based on F_2^- centers in LiF.

Fig. 13. Experimental setup for the investigation of nonlinear perception of laser IR emission: 1) F_2^- center laser; 2) spectrograph; 3) monochromator; 5, 6, 13) filters; 14) incandescent lamp; 4) frequency doubling crystal ($LiIO_3$).

At a sixfold pump excess over threshold, the pulse durations obtained were: $\tau_F = 23\cdot10^{-12}$ sec ($L_F = L_p$, $\Delta\nu_g = 1.5$ cm^{-1}); $\tau_F = 5.3\cdot10^{-12}$ sec ($2L_F = L_p$, $\Delta\nu_g = 8.0$ cm^{-1}); $\tau_F = 12\cdot10^{-12}$ sec ($2L_F = L_p$, $\Delta\nu_g = 2.0$ cm^{-1}).

Figure 12 shows the second-harmonic-generation correlation curves at $P_{av.p} = 490$ mW (sixfold excess above threshold) for $\Delta\nu_g = 8$ cm^{-1} (partial locking) and $\Delta\nu_g = 2$ cm^{-1} (total). We note that, judging from the correlation curve in Fig. 12a, the "chirp" effect observed in the lanthanum beryllate laser did not occur in our active medium at the pump levels employed.

We have performed experiments with mode-locked pumping of an F_2^- laser by a BeLa:Nd^{3+} laser operating in the passive mode-locked regime. As described above, the pumping laser produced in this case train-pulse durations 15-18 nsec at a total train duration 40 nsec and approximate energy 250 mW. These pump parameters ensured a fivefold excess above threshold in the F_2^- laser, and shortened the pulses to 7-8 nsec at a lasing spectrum width 2 cm^{-1}.

A very important fact is that in the course of operation we observed no photodisintegration of the F centers at pump-power densities up to $2\cdot10^9$ W/cm^2. The time stability of the color centers ensured a constant F_2^- laser efficiency on the order of 2% during all the experiments. Thus, the high optical and thermal endurance, as well as the temporal stability of the F centers, create the necessary prerequisites for the development of highly stable picosecond pulse generators in the range 1.1-1.25 μm on the basis of F_2^- centers in an LiF crystal.

USE OF PICOSECOND LiF(F_2^-) LASER TO STUDY NONLINEAR VISUAL PERCEPTION OF INFRARED RADIATION

The advent of a picosecond-pulse source with wavelengths tunable in the near infrared makes possible a relatively simple but very interesting experiment on nonlinear perception of radiation by the human eye. The first nonlinear perception of infrared radiation from a laser with "unaided" eye was reported in [21]. Similar investigations were made by now for cw as well as microsecond and nanosecond pulsed lasers. The most accurate measurements were made in [22] with a pulsed nanosecond-band laser tunable in the range 0.9-1.3 μm.

It was of interest to ascertain whether a change to picosecond pulses would lead to any new properties. The use of an LiF (F_2^-) laser with mode-locked pumping by an YAG:Nd^{3+} laser made it possible to operate in two regimes: a) with total mode locking, when the

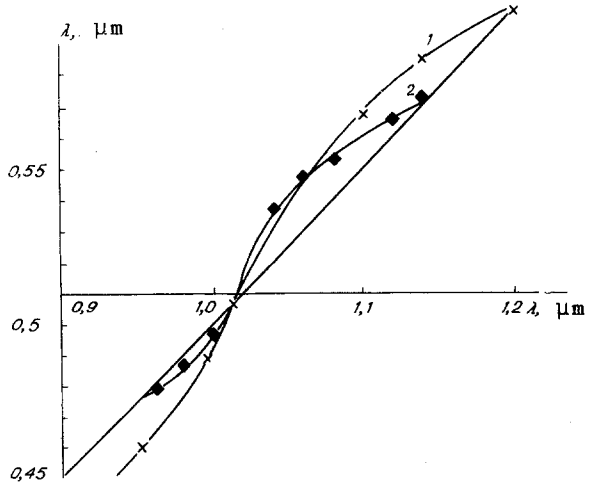


Fig. 14. Linear perception vs wavelength of IR radiation (curve 1); linear perception recalculated for IR radiation, at a tenfold change of the radiation intensity (curve 2).

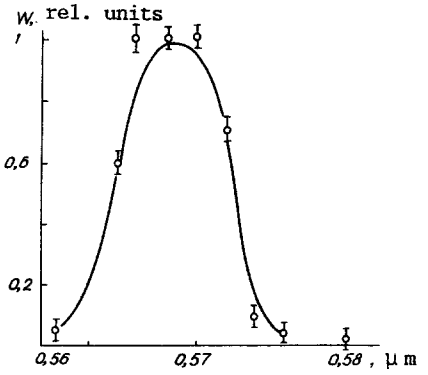


Fig. 15. Sensitivity curve of nonlinear perception of IR radiation.

laser produced in the USP regime pulses of duration $15 \cdot 10^{-12}$ sec, and 2) with partial mode locking when in the short-pulse regime pulses of duration $(200\text{-}500) \cdot 10^{-12}$ sec were produced in the subnanosecond range.

To assess the influence of an incandescent lamp on the nonlinear perception of the visible band (Fig. 13), the second harmonic of an IR laser beam was passed through a monochromator in parallel with incandescent light. Comparison of the perception of the second harmonic and of the incandescent lamp has shown that the perceived color hue of the lamp depends to a considerable degree on the intensity and can lead to substantial errors in the determination of the wavelength as perceived by the procedure of [22, 23]. A detailed investigation of the dependence of the linear perception on the intensity was carried out back in the 1930s [24], when a change of the linear perception was observed for a tenfold change in intensity; the plot of [24] is close in character to that of [22]. The two plots are shown for comparison in Fig. 14. In the linear perception there are three points that are independent of intensity: 0.507, 0.575, 0.470 μm. In the nonlinear perception, the 1.014-μm wavelength was taken by the author of [22] to be a second harmonic (of 0.507 μm) and was in good agreement with the corresponding linear-perception point. The second wavelength 1.2 μm, regarded as a second harmonic, does not coincide with a special point of the linear perception, although the general character of the deviation in the radiation perception is similar for both plots. This is a very interesting fact and its interpretation must be approached with caution.

In our experiment we passed through the monochromator the second-harmonic radiation, besides the incandescent-lamp light and the laser IR emission. All three signals were ad-

justed to equal intensity perception and to equality of the linear perception of the second harmonic and the lamp, after which the second-harmonic beam was blocked off and the perceptions of the IR radiation and of the lamp were compared. The level of the energy incident on the cornea was varied, using neutral filters, in the range from $2 \cdot 10^{-6}$ to $7 \cdot 10^{-4}$ J/cm^2. The upper limit was dictated by laser safety. Both in the USP and in the short-pulse regime, the wavelength perception differed in the mean by 5-7 nm in the long-wave region at an energy density $7 \cdot 10^{-4}$ J/cm^2 and 1.14-μm wavelength. With increasing radiation power, the perceived wavelength shifted to the long-wave region by 10-12 nm at an energy $9 \cdot 10^{-6}$ J/cm^2. Although the shift is negligible when the power is changed by two orders of magnitude, with accuracy ±5 nm a shift was typically observed in all the measurements. In the USP regime, the radiation perception was somewhat closer to the second-harmonic wavelength at the maximum permissible energies. The threshold sensitivites for the USP and short-pulse regimes turned out to be close and we estimate them to be $(1.45\text{-}0.7) \cdot 10^{-6}$ J/cm^2, although for the USP regime we expected a higher threshold sensitivity of the eye for the two-photon mechanism of IR radiation perception.

The dependence of the perceived wavelength on the intensity (Fig. 14) can be treated in two ways: 1) as a nonlinear analog of the Purkyně effect [24] – the shift of perception wavelength as a function of intensity; 2) as a superposition of the linear Purkyně effect on the nonlinear-perception data. The spectral dependence of the perception in the 1.1-1.25-μm band for short and ultrashort pulses agrees with the results of [22].

Investigation of the IR-sensitivity perception curve has shown that in the 1.1-1.25-μm range there is readily observed a susceptibility peak with a maximum at 1.13 μm (Fig. 15), in agreement with the results of [22] in which, in addition to this peak, a structure consisting of seven peaks was recorded. These peaks are attributed by the authors to a manifestation of the vibrational structure of the pigment molecules in two-photon absorption. A vibrational spectrum, not resolvable in the linear-absorption spectra (T = 300°K), was observed [25] in the two-photon absorption (TPA) spectra. Further confirmation of the connection between the structure in the perception sensitivity and the vibrational spectrum, due to the TPA mechanism, is of great importance for the understanding of the processes underlying vision, and calls for direct verification on iodopsin molecules.

According to present notions, color vision is due to the retinal cones in which light produces photoisomerization of three types of visual pigment molecules contained in the iodopsin. There are no data on the rates of photoisomerization of these pigments. Using the data on rhodopsin (the black-and-white vision pigment), the rate of isomerization is determined by the time $6 \cdot 10^{-12}$ sec [1], a fact attributed to relaxation of electronic excitation of the rhodopsin molecule.

The absence of a clear-cut difference in the perception of IR radiation at a pulse duration $(15\text{-}20) \cdot 10^{-12}$ sec compared with nanosecond pulses indicates that the rate of photoisomerization of iodopsin molecules is in all likelihood also in a range amounting to several picoseconds.

CONCLUSION

Thus, even the first results of an investigation of USP lasers using certain new active materials demonstrate the possibility of using them for various purposes, including for mode-locked laser pumping. Examples are BeLa:Nd^{3+} lasers on the 1.069-μm transition for pumping lasers on F_2^- color centers in LiF; lasers on impurity centers such as MgF_2:Ni(Co) on the 1.354-μm transition for pumping lasers emitting in the 1.6-1.7-μm band; the alexandrite laser for excitation of USP generation by lasers on F_2^+, F_3^- and other color centers in LiF and NaF crystals; and second harmonic pumping of USP dye lasers.

It must be specially emphasized that alexandrite is a promising active medium for continuously operating picosecond lasers tunable in the 0.7-0.8-μm band.

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