

CTuK63 Fig. 1. Experimental arrangement.

mal disturbances and second, mechanical quenching of the lowest triplet state of the dye molecules. A frequently used and high efficient laser dye solution is Rhodamine 6G in ethylene glycol. Employing a new spectroscopic method, we measured a triplet state lifetime of 5 μ sec in air-saturated solution at room temperature.¹ This is to be compared with the dye molecules, flight time through the active region of a typical cw dye laser. This time is about 2 μ s. Therefore, owing to the motion of the jet, the triplet state population in the active region is reduced sufficiently so that laser action becomes possible. Moreover it was concluded from the triplet data of Rhodamine 6G¹ that no optical gain can be expected in ethylene glycol without mechanical removal of triplet molecules.

The triplet state lifetime can be reduced by the use of other solvents or by an increase of the O₂ concentration of the solution. The question arises if the triplet state lifetime can be reduced to such a degree that cw laser operation can be produced at much smaller dye velocities or even in a stationary dye solution. We did observe cw laser operation in Rhodamine 6G at a flow velocity as low as 0.03 m/s. This was possible because the triplet lifetime could be reduced to less than 1 μ s by using water as the solvent. In order to prevent dimerisation of the dye, 2% Ammonyx LO was added. As shown in Fig. 1 the dye solution (concentration 10⁻⁴ M) was contained in a fused silica cell (1-mm depth, standard spectrometer quality). The dye cell was placed at the focus of a folded three-mirror cavity and oriented at Brewster's angle. The pump source was a Kr-ion laser operating at 531 nm. Threshold for laser operation was 300 mW at flow velocities down to 0.03 m/sec. At velocities smaller than 0.03 m/sec the threshold increased markedly due to thermal inhomogeneities. From the dye flow velocity of 0.03 m/s a flight time through the active region as long as 0.5 ms is calculated.

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A laser system: a Joule exciplex XeCl*-laser-powerful narrow-band dye laser for ecological lidar

T. N. Kopylova, V. B. Kaul',* R. T. Kuznetsova, G. V. Mayer, S. V. Mel'chenko,* A. N. Panchenko,* L. G. Samsonova, V. B. Sukhanov, V. F. Tarasenko,* E. N. Tel'minov, V. D. Kuznetsov Siberian Physical Technical Institute, State University, Tomsk. sq. Revolution 1, 634050, Russia

A long-distance monitor lidar should feature high radiation intensity (of the order of hundreds of millijoules), because a scattered light intensity falls with distance more rapidly than R₂. The laser radiation spectral width should be sufficiently small (of the order of hundreds small or the order of one tenth of an angstrom).

In the present summary of the paper we report the experimental design of a laser system with radiation of hundreds of millijoules at wavelengths allowing one to detect atmospheric impurities of SO₂ and NO₂ without making essential changes in the configuration of the optical scheme. The system includes two laser channels, each consisting of the following system: XeCl-laser oscillator-amplifier and dye laser oscillator-two amplifiers. The wavelength in the first channel is $\lambda_{on} = 3084.5$ Å and in the second one it is $\lambda_{off} = 3077.0$ Å. The radiation energy is up to 250 mJ per pulse and the linewidth is 0.1–0.3 Å. In NO₂ monitoring mode, the XeCl-laser amplifier operating in a free lasing regime pumps the dye laser oscillator-two amplifier system. The first channel is tuned to the wavelength of $\lambda_{on} = 4469$ Å and the other, $\lambda_{off} = 4482$ Å. The radiation energy is about 150 mJ and the linewidth is 0.1 Å in each channel.

The lasers operate at a repetition rate of 5 Hz and are synchronized to provide a time delay of 10 ms for each channel.

Because manufacturing of lasers with characteristics as specified above is a significant problem, we will discuss the laser optical scheme in more detail. The difficulty of obtaining intense narrow-band radiation at the specified wavelengths in the XeCl laser is caused by the following: these wavelengths correspond to the peaks of electronic-vibrational transitions 0–0 and 0–3 with the Frank-Condon factors and hence cross sections and weak signal gains are two times smaller than for 0–1 and 0–2 transitions for these molecules. The use of an original resonator allowed one to obtain the output oscillator energy of 3 mJ for the linewidth of 0.25 Å, and 30 mJ for the linewidth of 0.4 Å at the wavelengths of 3077 and 3084.5 Å. An LIDA-T laser was also used as an amplifier.

A problem of intense UV radiation (with energy ≥ 1 J per pulse) conversion in dye lasers was solved by the original design of dye-cells, such as a cell-prism of total internal reflection. A cell-prism is made of quartz with a cavity for dye solution oriented in a special way. Due to the phenomenon of total internal reflection, pump radiation excites the dye volume uniformly by four light beams. This allows one to produce a uniform laser output beam with a sufficiently small diver-

gence (<3 mrad). A high conversion efficiency (of about 25%) is possible for flow power fluency ≥ 2 MW/cm². An optical setup required to obtain high energy parameters of narrow-band radiation (linewidth of about 0.1 Å) involves a high-power MZHL-03 dye laser. The design also includes a master generator and two amplifiers.

*Institute of Strong-Current Electronics, Siberian Department of the Russian Academy of Sciences, Tomsk, ave. Akademicheskii 4, 634055, Russia

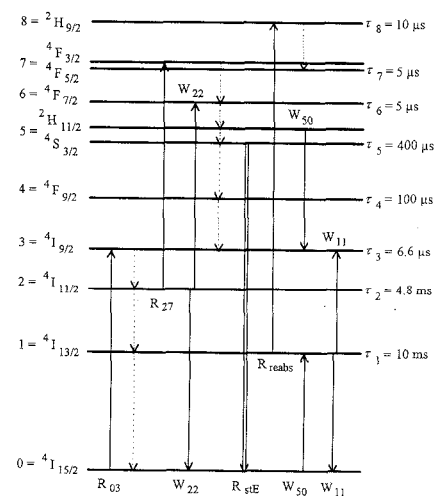
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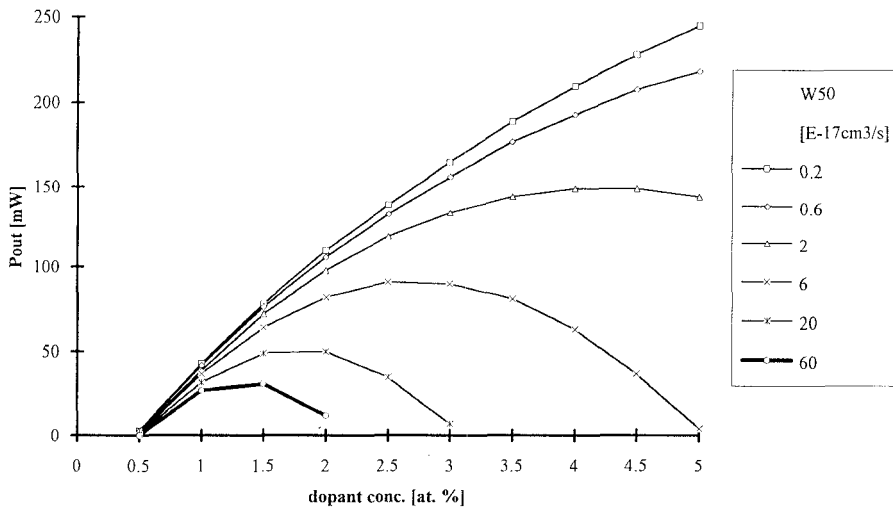
Population mechanisms of the room-temperature 551 nm Er³⁺:LiYF₄ laser

M. Pollnau, E. Heumann,* F. Heine,* T. Danger,* W. Lüthy, G. Huber,* H. P. Weber, Institute of Applied Physics, University of Bern, Sidlerstr. 5, CH-3012 Bern, Switzerland

One of the interests in solid-state laser physics has focused on diode-pumped blue and green upconversion lasers. At 551 nm upconversion-pumped laser operation in Er³⁺:LiYF₄ at cryogenic temperatures has been reported, see e.g., Ref. 1. Recently room-temperature lasing under different excitation schemes has been achieved.^{2,3} However, the population and deexcitation processes of the upper laser level as well as the influence of reabsorption on the laser wavelength⁴ have not yet been quantitatively understood. In this paper the results of a computer simulation of the Er³⁺:LiYF₄ laser are reported. The rate-equation scheme considers all excited levels up to ²H_{9/2} ground-state depletion, excited-state absorption (ESA) on the pump and laser wavelength, three upconversion processes, stimulated emission, and a realistic resonator design. From the computer simulation the population mechanisms of the laser system are ana-



CTuK65 Fig. 1. Energy level scheme of Er³⁺:LiYF₄ indicating the processes that are relevant for the population and deexcitation of the ⁴S_{3/2} upper laser level.



CTuK65 Fig. 2. Calculated output power versus dopant concentration for different values of the cross relaxation parameter W_{50} . The experimental results of Ref. 3 correspond to the curve with $W_{50} = 6 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$.

lyzed and suggestions for a better laser performance are derived.

The parameters of the system were taken from Refs. 3 and 5. The experimental results for the room-temperature 551 nm cw laser³ were reproduced in the simulation with a laser-beam waist of 30 μm . The upper laser level is populated from $^4I_{11/2}$ via ESA $R_{27} \ ^4I_{11/2} \rightarrow \ ^4F_{3/2}$ and interionic upconversion $W_{22} \ (^4I_{11/2}, \ ^4I_{11/2}) \rightarrow \ (^4I_{15/2}, \ ^4F_{7/2})$ (see Fig. 1). The calculated rates for $P_{\text{abs}} = 400 \text{ mW}$ at 810 nm are $R_{27} = 81\%$, $W_{22} = 19\%$. This shows that inversion is achieved mainly via ESA. Excited-state reabsorption on the laser wavelength $R_{\text{reabs}} \ ^4I_{13/2} \rightarrow \ ^2H_{9/2}$ has a small cross section in $\text{Er}^{3+}:\text{LiYF}_4$. Only 10% of the green emission reabsorbed, if a cross section $\sigma_{18} = 2 \times 10^{-21} \text{ cm}^2$ is assumed. Also ground-state reabsorption is weak because the laser terminates in the highest Stark level of $^4I_{15/2}$ and the ground state is efficiently bleached.

The intrinsic lifetime of the $^4S_{3/2}$ upper laser level is concentration-quenched by the cross relaxation $W_{30} \ (^2H_{11/2}, \ ^4I_{15/2}) \rightarrow \ (^4I_{9/2}, \ ^4I_{13/2})$, because $^2H_{11/2}$ is thermally coupled with $^4S_{3/2}$ and Boltzmann-populated from this level. Experiments show that the laser has the highest output power at approximately 1% dopant concentration.³ By variation of the parameter W_{50} (Fig. 2) and comparison with the experimental result the value of W_{50} can be estimated: $W_{50} = 6 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$. Owing to the strong cross relaxation W_{50} energy is upconverted via $W_{50} \ ^4I_{15/2} \rightarrow \ ^4I_{13/2}$, $W_{11} \ ^4I_{13/2} \rightarrow \ ^4I_{9/2}$, relaxation $^4I_{9/2} \rightarrow \ ^4I_{11/2}$, and $R_{27} \ ^4I_{11/2} \rightarrow \ ^4F_{3/2}$. The laser is thus supported by an avalanche process. The processes W_{50} and W_{11} seem to be more efficient than ground-state absorption R_{03} , which has a very small absorption coefficient.

In conclusion we have shown that the strong cross relaxation from the upper laser level is responsible for the quenching of the laser output with dopant concentration. The corresponding parameter has been estimated. Crystals with a smaller cross-relaxation parameter would allow higher dopant concentrations and absorption coefficients. An avalanche effect

appears to be an important excitation mechanism for the green erbium laser. This effect can be optimized by using other pump wavelengths with higher ESA cross sections.

**Institute of Laser-Physics, University of Hamburg, Jungiusstr. 11, D-20355 Hamburg, Germany*

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New data on induced-emission cross section in solid-state laser materials

I. F. Balashov, B. G. Berezin, V. N. Ivanov, *S. I. Vavilov Optical Institute, Babushkina St. 36, St. Petersburg 193171 Russia*

The values for induced-emission cross section of laser glasses and crystals determined from standard absorption and emission measurements and Judd-Ofelt calculations are not sufficient to describe the generation properties of laser materials. A more successful way to determine effective values for laser material cross section is based on mathematical approximation of output to input energy characteristics realized in lasers with known resonator parameters.

The measurements were made by using pulse lasers. Active rods from materials to be measured were located in the pumping cavity of the laser. It was necessary to make some tricks to avoid the influence on the results of the measurements by such phenomena as pumping system nonlinearity, uncertainty in irra-

diate losses in the resonator, and non-homogeneity of pumping light in active rod. After realizing these tricks measuring values for effective cross section became independent on the parameters of the laser and on laser construction.

An essential difference was found between the values for effective cross section received in our experiments and values determined with standard spectroscopic calculations. It turned out that the effective cross sections in similar laser materials were dependent on the concentration of the activator. Effective cross section in neodymium-doped phosphate glasses changes from $3.6 \times 10^{-20} \text{ cm}^2$ for concentration $1.4 \times 10^{20} \text{ cm}^{-3}$ to $1.9 \times 10^{-20} \text{ cm}^2$ for $12.2 \times 10^{20} \text{ cm}^{-3}$. Moreover the values for effective cross section in these glasses are linear dependent on quantum efficiency of luminescence. The Judd-Ofelt cross section for all these glasses is about $3 \times 10^{-20} \text{ cm}^2$ and is independent of concentration.

The values for efficient cross section in neodymium silicate glasses were found to be $3.1 \times 10^{-20} \text{ cm}^2$ for concentration $1.9 \times 10^{20} \text{ cm}^{-3}$ and 2×10^{-20} for $4.6 \times 10^{20} \text{ cm}^{-3}$ while the spectroscopic value for these glasses is only $1.7 \times 10^{-20} \text{ cm}^2$.

The measurements were made also for some neodymium-doped crystals. The efficient cross section in YAG:Nd at 1.06 micron was $1.7 \times 10^{-19} \text{ cm}^2$ for concentration of Nd 1.1 wt. percent and about $3 \times 10^{-19} \text{ cm}^2$ for 0.5 wt. percent. The Judd-Ofelt cross section in effective laser crystals KGd $(\text{WO}_4)_2$:Nd ($4.3 \times 10^{19} \text{ cm}^{-3}$) is more than in YAG:Nd ($3.6 \times 10^{19} \text{ cm}^{-3}$) but its effective cross section is $1.25 \times 10^{-19} \text{ cm}^2$ for concentration 7 wt. percent and $1.9 \times 10^{-19} \text{ cm}^2$ for 3 wt. percent. The greater difference between effective and spectroscopic cross sections in KGd $(\text{WO}_4)_2$ than in YAG can be explained by the greater concentration of neodymium in KGd $(\text{WO}_4)_2$.

The method for measuring the effective cross section can be used not only for neodymium-doped laser materials. We began to investigate laser solid-state materials for other generation wavelengths.

The results of the investigations show that our method for measuring effective cross section has given new opportunity to evaluate laser properties of active solid state materials.

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Spectrum stabilizing in Cr:Tm:Er:YAG laser at 2.69 μm

I. V. Mochalov, G. T. Petrovskii, A. V. Sandulenko, V. A. Sandulenko, V. S. Terpugov, *S. I. Vavilov Optical Institute, Babushkina St. 36, St. Petersburg 193171 Russia*

It is of much difficulty to provide efficient lasing on YAG:Er³⁺ crystal at the shorter wavelength transition (2.69 μm) of the 3 μm -lasing spectrum of Er³⁺ due to high value of $I_{13/2}$ level lifetime and because of cross-relaxation. Codoping the crystal with Cr³⁺ ions as sensitizers and Tm³⁺ ions disactivating $^4I_{13/2}$ level, situation changes. However the tendency of Cr:Tm:Er:YAG (CTE-YAG) laser spectrum to longer wave shifting still remains. In this work, the possibilities of stabilizing the