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Investigations of YAG:Er³⁺, Yb³⁺ and YAG:Co²⁺ crystals for laser application

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ABSTRACT

The results of examinations of spectroscopic properties and generation parameters of new active media, i.e., Er³⁺ and Yb³⁺ doped YAG crystals are given. Erbium and ytterbium doped YAG single crystals were obtained by the Czochralski method. The spectral properties and laser characteristics were investigated. Absorption spectra of Er³⁺ and Yb³⁺ - doped active media were measured in the spectral range 190÷5000 nm at room temperature. Excitation and luminescence spectra were also recorded at room temperature with a JOBIN-YVON spectrofluorimeter using a diode laser (POLAROID 4300, 980 nm, 1 W) as an excitation source. The measurements of the lifetime of the Er³⁺ ions in the upper laser level (⁴I_{13/2}) of the samples were made by the direct method with pulse excitation.

We present also the results of investigations on spectroscopic properties and non-linear absorption effect in YAG: Co²⁺ crystal - new absorber used for the systems generating giant-pulse radiation at 1.5 μm.

keywords: eyesafe laser, active media, saturable absorbers, erbium doped YAG, ytterbium doped YAG, cobalt doped YAG

1. INTRODUCTION

There is considerable interest in compact pulsed high peak power laser sources emitting at wavelengths near 1.5 μm. Rangefinders and other applications with free space propagation could be strong benefit of such devices. The wavelength of around 1.5 μm is in the eyesafe regime where significantly higher pulse energies can be used without human eyes damaging¹. For wavelength of 1.55 μm there is the minimum of absorption of quartz optical fibres, within the range of 1.5÷2 μm atmosphere transmission is the highest but absorption of biological media and atmospheric pollution of some type are very strong. Thus, „eye-safe” lasers can be applied for telecommunication, optical radars, distance measurements, remote detection of contaminations, and medicine².

In the first laser systems generating eyesafe radiation, a Raman shifter was used in form of a methane cell in which a conversion of radiation, generated by YAG:Nd³⁺ (λ=1064 nm), into radiation of wavelength of 1.54 μm occurred. Molecular crystals (e.g., Ba(NO₃)₂, CaCO₃, or CaSO₄), in which the effect of forced Raman scattering occurs, are competitive with high-pressure gaseous cells. Because the efficiency of this process is not high, these systems have not found practical applications³. Parametric generation of wavelengths near 1.5 μm in nonlinear crystals was practically not used. The main obstacle in this case is the difficulty to ensure stable laser operation within a wide range of temperatures⁴.

At the beginning of the 90's an erbium-ytterbium-phosphate glass was used for the first time for generation of laser radiation at the wavelength of 1.5 μm⁵. The laser systems including this active material have found many practical applications^{6,7}. The drawback of phosphate glass is its low thermal and mechanical stability. So, new search is carried out for new, higher resistant glass matrices and crystalline media⁸.

Efficient materials generating radiation within "eye-safe" range are Cr⁴⁺ doped crystals. Wide luminescence band of Cr⁴⁺ ions within the range of 1.2÷1.6 μm, enables us to obtain tunable generation in this spectral range but absorption within the range of 900÷1100 nm makes possible to pump with laser diode of 980 nm or neodymium laser of 1064 nm.

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Pumping YAG:Cr⁴⁺ with diode laser gives tunable generation within the range of 1350-1550 with its maximum at 1450 nm⁹. Other crystals used in systems of generation of eye-safe radiation are active media doped with Er³⁺ and Yb³⁺ ions. Yb³⁺ ions are used in Er³⁺ doped crystalline media in order to increase excitation efficiency, especially in diode pumped laser systems¹⁰.

Searching of non-linear absorbers for laser generating radiation at 1.5 μm are focused on materials doped with the following ions; U²⁺, Co²⁺, and Er³⁺¹¹⁻¹⁴. The most advantageous spectroscopic parameters [active cross section and saturation energy (intensity)] have media doped with cobalt; LaMgAl₁₁O₁₉:Co²⁺ (LMA:Co) and SiO₂-ZnO-Al₂O₃:Co²⁺ ceramics. Absorbers with bivalent cobalt ions in tetrahedral sites are characterised by various spectroscopic parameters. YAG:Co²⁺ and YSGG:Co²⁺ crystals are so-called "fast absorbers" with relaxation time of about 1 ns¹¹ and ZnSe:Co²⁺ is "slow absorber" with relaxation time of 290 μs¹⁴.

2. CRYSTAL GROWTH

The crystals were obtained by the Czochralski method, using iridium crucibles of external dimensions φ 50×50×1.5 mm. Insulating housing of the crucible was made of alundum ceramics and granular zirconium ceramics stabilised with hafnium (ZrO₂:HfO₂) that filled the space between the crucible, the tube, and alundum base.

The crystallisation processes were carried out in nitrogen atmosphere containing small amount (a fraction of percentage) of oxygen. High purity oxides Y₂O₃(5N) and Al₂O₃(5N) from J.M. & PROD., Er₂O₃(5N), Yb₂O₃(5N) and CoO(4N) from ALDRICH were used as initial materials.

Crystals of a diameter of about 25 mm and a length of 60 mm with following compositions were obtained:

YAG:Er³⁺ (1.5% Er³⁺)

YAG:Er³⁺, Yb³⁺ (0.9% Er³⁺, 9% Yb³⁺)

YAG:Er³⁺, Yb³⁺ (0.5% Er³⁺, 5% Yb³⁺)

YAG:Co²⁺ (2% Co²⁺)

In Fig.1 typical YAG:Er³⁺,Yb³⁺ and YAG:Co²⁺ single crystal grown by Czochralski method is presented. In all obtained crystals the core area of the characteristic threefold symmetry was found. The core has clear, sharp boundaries and its area has a diameter of about 2 mm.

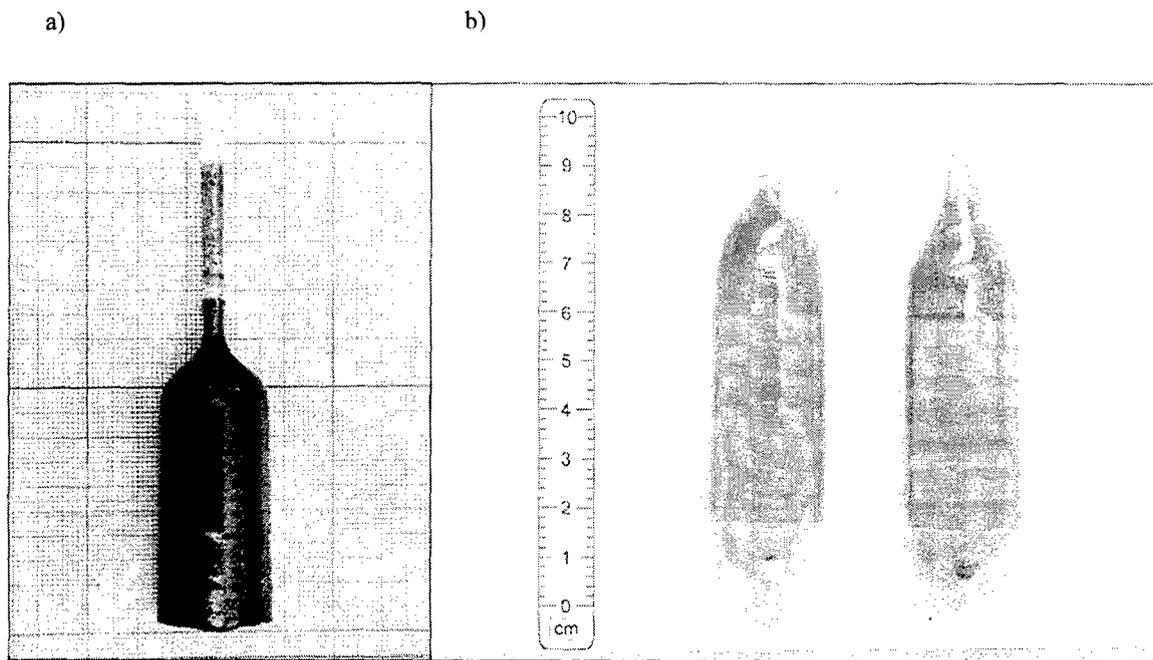


Fig. 1. As-grown YAG:Co²⁺ (a) and YAG:Er³⁺,Yb³⁺ (b) single crystal.

On the basis of the investigations carried out with a Mach-Zender interferometer it was stated that the crystals in the core area are characterised by an inhomogeneity of the refractive index due to defects and stresses. Out of the core area all of crystals were optically homogeneous. The stress in the crystals was examined using the elastoscopic method and registering the picture obtained for crystal, placed between crossed polarisers.

On the basis of the interferometric and elastoscopic investigations the areas of the crystal cuts were determined in order to perform optical elements for the investigations of the spectroscopic characteristics.

3. SPECTROSCOPIC INVESTIGATIONS

From the obtained crystals plane-parallel plates of a thickness of 1 mm were cut, granded, and polished for spectral measurements. In order to determine the dependence of the absorption coefficient on wavelength $k(\lambda)$, the transmission was measured as a function of the wavelength. The measurements were carried out within the spectral range of 200÷3000 nm ($\Delta\lambda=1$ nm) using a LAMBDA900 PERKIN ELMER spectrophotometer and within the range of 1.5÷25 μm ($\Delta 1/\lambda=1$ cm^{-1}) using a Fourier PERKIN ELMER spectrophotometer 1725-X FT-IR. The spectra of the absorption coefficient for Co^{2+} doped YAG crystals are shown in Fig. 2.

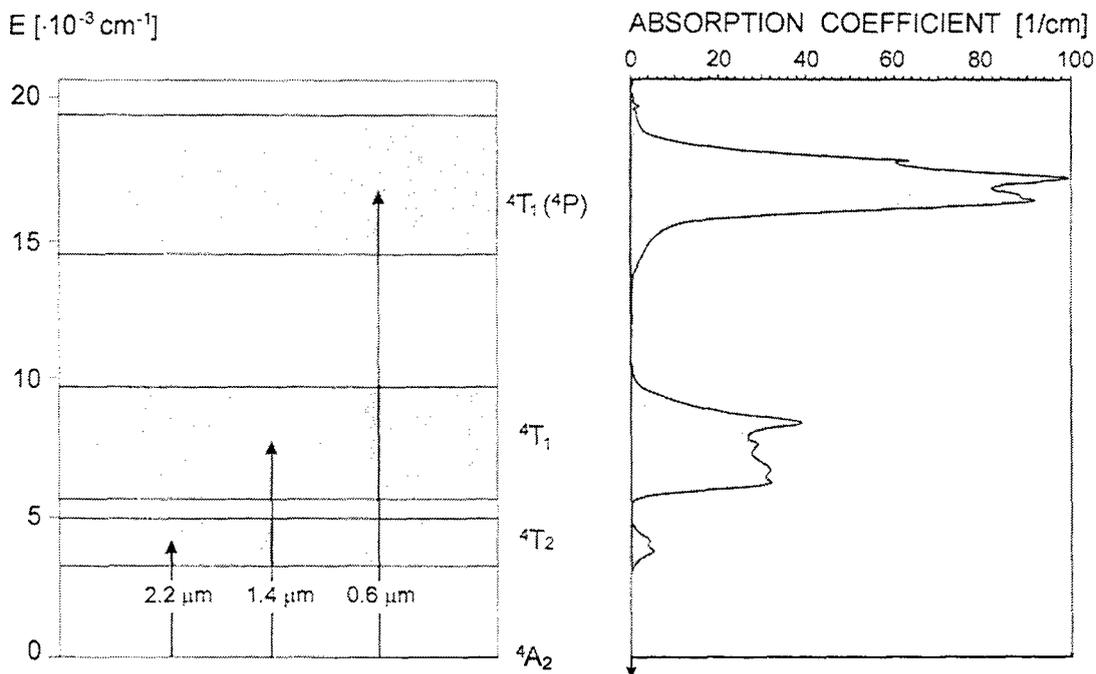


Fig. 2. Energy levels for Co^{2+} in tetrahedral sites and room temperature $\text{YAG}:\text{Co}^{2+}$ (2 at.% Co^{2+}) absorption spectra

The measurements of the luminescence spectra have been performed with a H20 JOBIN YVON monochromator (focal length 200 mm). For excitation a laser diode emitting at 970 nm was used. The luminescence, was registered by means of a LOCK-IN (STANFORD RESEARCH SR510) system with a thermoelectrically cooled InGaAs detector.

The measurements of the lifetime of the ions in the upper laser level (${}^4I_{11/2}$) of the samples doped with Er^{3+} and Yb^{3+} were made by the direct method with pulse excitation. The crystal was excited with a radiation pulse significantly shorter than the lifetime τ of the excited level. After excitation the fluorescence decay can be observed.

As source of the exciting pulses of wavelength 970 nm (excitation of Er^{3+} and Yb^{3+} ions) a POLAROID 4300 laser diode was used. The laser was supplied from a power supply SDL800, controlled by a pulsed generator and generated pulses of about 8 μs duration at a frequency of about 0.66 kHz. In the detection channel, perpendicular to the excitation channel, a germanium photodiode was used and the fluorescence decay was registered with a digital oscilloscope LeCROY 9350AM (500 MHz). The fluorescence decay times of the investigated samples are listed in Table 1.

Tab. 1. The fluorescence decay times (level $^4I_{11/2}$) of the investigated YAG:Er³⁺ and YAG:Er³⁺,Yb³⁺ crystals

Crystal	Fluorescence decay time
YAG:Er ³⁺ (1.5% Er ³⁺)	5.6 ± 0.1 ms
YAG:Er ³⁺ ,Yb ³⁺ (0.9% Er ³⁺ , 9% Yb ³⁺)	4.3 ± 0.1 ms
YAG:Er ³⁺ ,Yb ³⁺ (0.5% Er ³⁺ , 5% Yb ³⁺)	3.5 ± 0.1 ms

4. NON-LINEAR ABSORPTION INVESTIGATIONS

Investigations of non-linear absorption of YAG:Co²⁺ crystals of various concentrations of Co²⁺ ions were carried out on the basis of analysis of bleaching process dynamics.

Non-linear absorber is characterised by molecular and macroscopic parameters. The molecular parameters are as follows: σ is the absorption cross-section [cm²], τ is the lifetime at the excitation level [s], N_0 is the concentration of absorption centres [cm⁻³], $\Delta N = N_2 - N_1$ is the density of population inversion.

The macroscopic parameters: d is the absorber thickness [cm], T is the absorber transmission for the intensity I , I_s is the saturation intensity [W/cm²].

If the relaxation time of absorber is $\tau \ll t_n$, where t_n is the duration of diagnostic pulse, an absorber is of „fast” type and the dependence $T(I)$ describes the equation formulated by Keyes¹⁵ and Hercher¹⁶.

$$(1 - T)^{-1} \ln \frac{T}{T_0} = \frac{I}{I_s} \quad (1)$$

where T_0 is the initial transmission (for small signals), I is the density of incident radiation, I_s is the density of saturation power determined as

$$I_s = \frac{h\nu}{\sigma\tau} \quad (2)$$

where τ is the lifetime of excited absorber.

Relationship (1) can be solved numerically¹⁷ or analytically using special *LambertW* $\equiv W(x)$ function¹⁸⁻²⁰ which is defined as a solution of equation

$$W(x) \cdot \exp(W(x)) = x \quad (3)$$

Analytical solution of equation (1) is dependence

$$T = \frac{I_s}{I} \cdot \text{LambertW} \left(T_0 \cdot \frac{I}{I_s} \cdot \exp \left(\frac{I}{I_s} \right) \right) \quad (4)$$

The changes of transmission of the examined samples as a function of power density of the passing radiation, emitted by the giant-pulse KIGRE MR-253 erbium laser were determined. Laser generated the pulses of 8 mJ energy with duration 25 ns. The power density of a diagnostic pulse was changed within the range of 1÷600 MW/cm². Figure 3 presents the example of application of *LambertW* function for approximation of measurement data of transmission dependence as a function of power density of incident laser radiation wavelength of 1535 nm obtained for YAG:Co²⁺ sample of initial transmission of 24.9%.

As it can be seen from the presented results of analyses, special function *LambertW* allows to correct description of „fast” non-linear absorbers for wide range of their initial transmissions. The results of performed analyses were compared with theoretical models for „fast” absorber as well as with experimental results obtained for non-linear YAG:Co²⁺ absorbers.

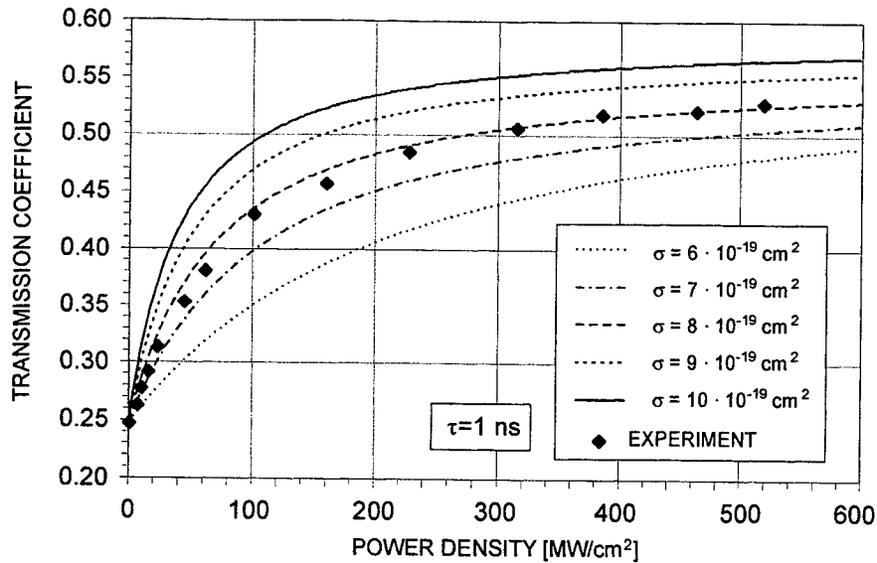


Fig. 3. *LambertW* function applied for approximation of measurement results of transmission as a function of power density of incident laser radiation obtained for YAG:Co²⁺ non-linear absorber

5. LASER ACTION IN YAG:Er³⁺,Yb³⁺

Investigations of longitudinally pumped microlasers generating at 1535 nm made of YAG:Er³⁺,Yb³⁺ were carried out. A schematic of the laser cavity is shown in Fig. 4. The results for YAG:Er³⁺,Yb³⁺ (0.9% Er³⁺, 9% Yb³⁺) are presented in Fig. 5.

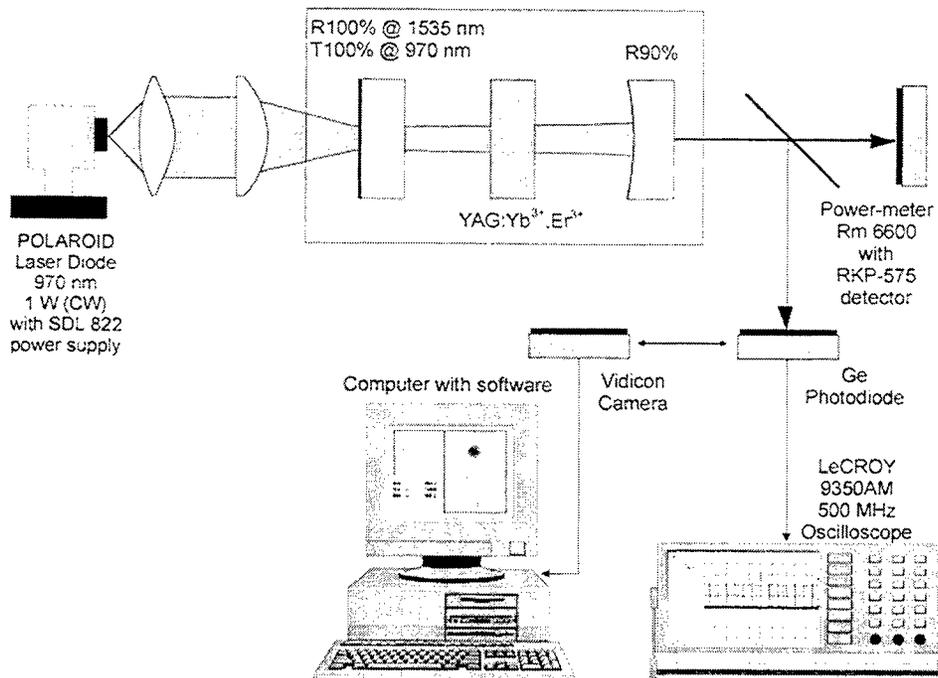


Fig. 4. Experimental setup for the diode pumped YAG:Er³⁺,Yb³⁺ laser

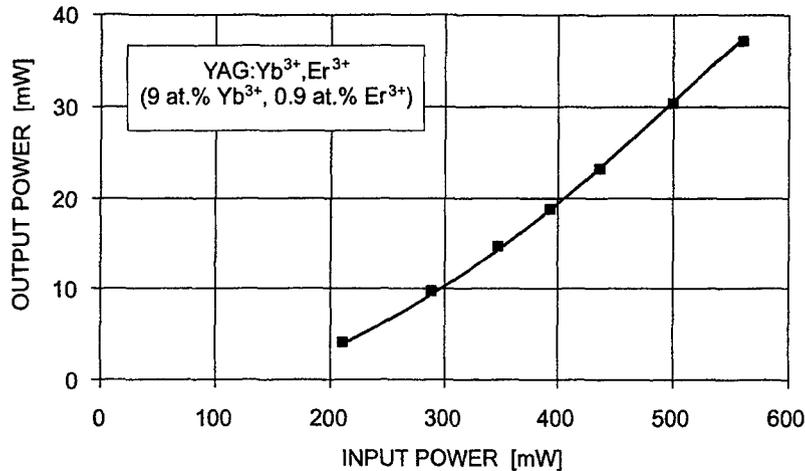


Fig. 5. Generation characteristics of YAG:Er³⁺,Yb³⁺ diode pumped laser (output mirror T=10%, r=40 mm)

6. CONCLUSIONS

The conditions of growing YAG:Er³⁺, YAG:Er³⁺,Yb³⁺ and YAG:Co²⁺ single crystals of various concentrations, devoted to lasers generating eyesafe radiation, were determined. The carried out investigations of the optical and spectroscopic properties of the obtained crystals showed their good optical quality, so they can be applied in laser systems.

Due to the advantageous spectroscopic parameters of YAG:Er³⁺, YAG:Er³⁺, Yb³⁺ and YAG:Co²⁺ crystals and their high thermal and mechanical stability characteristic for garnets, those crystals can be used in laser technology as active materials of lasers generating eyesafe radiation.

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