Liquid-phase epitaxy growth and characterization of Co,Si:YAG thin film saturable absorber

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Received 3 February 2004; accepted 30 May 2004
Available online 21 October 2004

Abstract

Thin films of Co,Si:YAG were grown on YAG and Er,Yb:YAG substrates by means of the isothermal liquid-phase epitaxy (LPE) dipping technique. X-ray diffraction analysis, optical transmission spectra measurements, and passive Q-switching experiments were performed to characterize the obtained layers. Absorption saturation measurements of Co,Si:YAG thin films were carried out at 1.54 μm. Passive Q-switching of the Er:glass laser by use of epitaxial Co,Si:YAG/YAG was observed. We demonstrated experimentally that the Co,Si:YAG layers could be used as an effective saturable absorber for the lasers operating near 1.5 μm.

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PACS: 81.15.Lm; 42.55.Sa; 42.70.Hj; 42.60.Gd; 78.20.e
Keywords: Liquid-phase epitaxy; Saturable absorber; Passive Q-switching; Microchip laser

1. Introduction

The most important application of the oxide crystals doped with tetrahedrally coordinated Co2+ ions is passive Q-switching of solid-state lasers operating in the near-infrared region. A number of crystals such as YAG, LGO, LMA, MALO, ZnSe as well as glass ceramic have been doped with Co2+ ions [1,2].

Fulbert et al. had used for the first time a liquid-phase epitaxy (LPE) to grow thin single crystalline film of Cr4+:YAG saturable absorber [3]. The French researchers have developed the growth of Cr4+:YAG/Nd3+:YAG epitaxial structures designed to fabricate a novel monolithic passive Q-switched microchip laser emitting at 1064 nm [4]. Our previous study concerning LPE growth of rare earth ions doped active garnet thin films for waveguide application indicated LPE technique as a proper method to produce dielectric microchip structures [5]. We have grown successfully a Cr4+ ions doped YAG and GGG layers as a saturable absorber on active Nd:YAG and Nd:GGG substrates, respectively, in order to prepare epitaxial structures for passively Q-switched microchip laser [6]. The obtained microlaser performance (repetition rate, pulse energy and pulse length) have resulted in our interest in epitaxy of Co2+:YAG films.

In this paper, we report for the first time, to our best knowledge, on the growth by LPE a thin saturable absorber layers of Co2+:YAG. Non-linear properties and high optical quality of the obtained Co2+:YAG epitaxial layers allow us to grow Co2+:YAG/Er,Yb:YAG microchip laser structures.

2. Experimental

2.1. Thin films growth

The Co2+:YAG layers were grown from a supercooled molten garnet-flux (PbO-B2O3) high temperature
solution using a standard isothermal LPE dipping technique with reversed axial rotation. The procedure of melt preparation and epitaxy run was similar to that developed for magnetic or waveguide garnet layers. Our first epitaxy experiments have been carried out on \(\{111\}\) oriented, 20mm in diameter, YAG substrates. In order to obtain divalent Co ions it is necessary to compensate the electric charge imbalance by optically inert tetravalent cation, such as Si\(^{4+}\) ion. The Co\(^{2+}\) ions substitute Al\(^{3+}\) ions in tetrahedral positions. The Co\(_3\)Si:YAG films were grown from the melts consisted of Al\(_2\)O\(_3\), Y\(_2\)O\(_3\), Co\(_3\)O\(_4\), SiO\(_2\), PbO–B\(_2\)O\(_3\) mixture. The concentrations (mole fractions) of Co\(_3\)O\(_4\) were changed in the range from 0.0002 to 0.0054 while the mole concentration ratio of Co\(_3\)O\(_4\):SiO\(_2\) equals 2 was kept constant for all used melt compositions. The growth temperature for all epitaxy run was held relatively high (>1000°C) in order to avoid Pb ions incorporation. Substitution of aluminum ions by cobalt and silicon ions caused an essential change of film lattice constant.

### 2.2. Characterization

Diffraction measurements were performed using X-ray quasi-parallel double-crystal arrangement with 400 reflection on a Ge monochromator and 444 YAG reflection of CuK\(_\alpha\) radiation [7].

The optical transmission spectra of epitaxial structures Co\(_3\)Si:YAG/YAG and Co\(_3\)Si:YAG/Er,Yb:YAG were measured by Perkin-Elmer Lambda 900 spectrophotometer.

Absorption saturation measurements were performed at 1.54\(\mu\)m using the KIGRE MR-253 laser head with rotating prism mechanical Q-switch. Passive Q-switching experiment was realized using 1.54\(\mu\)m Er:glass laser and Co\(^{2+}\):YAG/YAG structures as saturable absorber.

### 3. Results and discussion

The growth of high quality thick garnet epitaxial layers is limited by lattice mismatch between the film and substrate. In the case of good film lattice match to the substrate, the recorded XRD patterns indicate one single peak (Fig. 1(curve 1)). For film and substrate lattice difference, two peaks appear in the XRD rocking curve (Fig. 1(curve 2)).

At first, we have established growth conditions in order to obtain a sufficient absorption assigned to \(^{4}A_2\rightarrow^{4}T_1\) (\(^{4}F\)) transition of tetrahedrally coordinated Co\(^{2+}\) ions. Fig. 2 presents the transmission spectra showing the effect of increasing Co\(^{2+}\) ions concentration in the layer. Intensity of the absorption band between 1200 and 1600nm increases in proportion to Co\(_3\)O\(_4\) mole fraction in the melt. Additionally, the best lattice match was obtained for the highest Co\(_3\)O\(_4\) mole fraction. So, the Co,Si: YAG epitaxial films of the thickness up to 150\(\mu\)m on both substrate sides, were grown. Layer thickness was calculated with \(\pm 5\%\) accuracy from a difference of substrate weight before and after the layer growth. We have assumed the uniformity of the layer thickness on both substrate surfaces. The observed transmission spectra of epitaxial structure grown from the melt contained the highest amount of Co\(_3\)O\(_4\) is presented in Fig. 2(curve 3). The layers thickness in this sample reached 80\(\mu\)m.

A saturable absorber layer of Co, Si:YAG has been grown also on the active Er,Yb:YAG substrate in order to obtain an epitaxial structure, suitable for passive Q-switching of microchip laser operating at 1535nm. The concentration of erbium and ytterbium ions in the substrate was about 1 and 10at.\%, respectively. At this dop-
The investigations of changes in transmission, as a function of power density of transmitted 1535 nm radiation for Co, Si:YAG thin films with different concentration of Co\(^{2+}\) ions, have been carried out. KIGRE MR-253 laser provided probe pulses of 8 mJ of output energy and 25 ns of duration (FWHM). The laser pulse fluence (energy density) at the sample was varied in the range of 0.001–10 J/cm\(^2\) by changing the laser output, or by moving the samples along the propagation axis of the laser beam. Fig. 4 presents measurement results of transmission changes as a function of energy density of radiation at wavelength of 1535 nm for the sample of initial transmission \(T_0 = 82.3\%\).

A special \textit{LambertW} function was used for description of, so-called, “fast” non-linear absorber. Dynamics of non-linear bleaching, i.e., type of relationship between transmission and intensity of incident radiation depends on relationship between relaxation time of absorber and duration time of diagnostic pulse. If the relaxation time is \(\tau \ll t_n\) where \(t_n\) is the duration of diagnostic pulse, an absorber is of “fast” type (for Co:YAG, \(\tau \approx 1\) ns [1]) and the dependence \(T(I)\) describes the equation formulated by Keyes [8] and Hercher [9]:

\[
(1 - T)^{-1} \ln \frac{T}{T_0} = \frac{I}{I_S}
\]

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 \(I_S = \frac{hv}{\sigma_T}\) and \(\tau\) is the lifetime of excited absorber. Relationship (1) can be solved numerically or analytically using special \textit{LambertW} function [10] which is defined as a solution of equation:

\[
W(x) \exp(W(x)) = x
\]

Analytical solution of Eq. (1) is dependence

\[
T = \frac{I_S}{I} \text{LambertW} \left( \frac{T_0 I}{I_S} \exp \left( \frac{I}{I_S} \right) \right)
\]

On the basis of approximation of measurements data and performed analysis, the absorption cross-sections \(\sigma_{GSA}, \sigma_{ESA}\), and concentration of the absorption centers \(N_0\) were determined. The best fits to the experimental data were obtained with calculated values of the ground-state absorption cross-section \(\sigma_{GSA}\) that varied between \(3 \times 10^{-19}\) and \(8 \times 10^{-19}\) cm\(^2\) for different samples. Doping level of Co\(^{2+}\):YAG thin films was...
calculated to be $2 - 6 \times 10^{19} \text{cm}^{-3}$, showing a good agreement with the nominal concentration. At the same time the optimal ratio of the excited-state absorption cross-section $\sigma_{\text{ESA}}$ to the ground-state absorption cross-section $\sigma_{\text{GSA}}$ for different samples were found to be between 0.1 and 0.4.

Using a sample of Co$^{2+}$:YAG/YAG structure with the layer of non-linear absorber thickness of 150 $\mu$m from both sides of YAG substrate, a passive Q-switching of 1.54 $\mu$m flash lamp pumped Er:glass laser was tested. The resonator length was 30cm and consisted of a concave rear mirror (radius of curvature, $r = 200\text{cm}$) and flat output mirror with reflectivity at 1.54 $\mu$m, $R = 88\%$. The Co$^{2+}$:YAG saturable absorber (without any antireflection coating) with initial transmission $T_0 = 69.3\%$ was placed between the rod (QE 7S KIGRE) and the output mirror. The passively Q-switched 1.54 $\mu$m laser produced diminishing “pulse train” with a few mJ per pulse. Such a “pulse train” generation was also observed for 1.3m Nd:YAG diode pumped laser with V$^{3+}$:YAG as a saturable absorber [11]. In Fig. 5, the results of theoretical modelling for the case of V$^{3+}$:YAG and oscilloscope traces for Co$^{2+}$:YAG/YAG saturable absorbers were presented. The main difference between these results was the output pulses length, which was up to a few hundred of ns for 1.54 $\mu$m laser, and a few tens of ns for 1.3 $\mu$m laser. Theoretical modelling of Q-switching regime for Co$^{2+}$:YAG and experiments with a diode pumped passively Q-switched Co$^{2+}$:YAG/Er,Yb:YAG microchip lasers for high peak power pulse generation will be done and reported in the nearest future.

4. Conclusions

By means of the liquid-phase epitaxy the Co$^{2+}$:YAG films were grown successfully on YAG and Er,Yb:YAG substrates up to 150 $\mu$m in thickness. According to spectroscopic and transmission saturation measurements it can be concluded that the obtained films fulfil the materials requirements for Q-switching application. The experiments with a lamp pumped 1.5 $\mu$m laser show the possibility of using Co$^{2+}$:YAG layers for eye safe Q-switched lasers and microchip lasers.

Acknowledgments

The authors would like to thank Professor J. Jabczynski for helpful discussions, and to Mrs. K. Mazur and Dr. J. Sass for X-ray diffraction measurements. Financial support by the grant no. 7T11B 042 20 of the Polish State Committee for Scientific Research is gratefully acknowledged.

References


Fig. 5. (a) The diminishing “pulse train” as a results of theoretical model for V$^{3+}$:YAG saturable absorber [11]. (b) Oscilloscope traces of a multiple output pulses from a passively Q-switched 1.54 $\mu$m laser on the background of pumping radiation (the upper traces).