

Laser Research Center for Molecular Science

IX-W Developments and Researches of New Laser Materials

Although development of lasers is remarkable, there are no lasers which lase in ultraviolet and far infrared regions. However, it is expected that these kinds of lasers break out a great revolution in not only the molecular science but also in the industrial world.

In this project we research characters of new materials for ultraviolet and far infrared lasers, and develop new lasers by using these laser materials.

IX-W-1 Growth and Scintillation Properties of Yb Doped Aluminate, Vanadate and Silicate Single Crystals

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[*Opt. Mater.* **26**, 529 (2004)]

For the purpose of quick screening for charge transfer (CT) transitions of Yb₃ in various hosts, (Lu_{1-x}Yb_x)₃Al₅O₁₂ (Yb:LuAG) with $x = 0.05, 0.15, 0.30$ and (Y_{1-x}Yb_x)AlO₃ (Yb:YAP) with $x = 0.05, 0.10, 0.30$ were grown by the micro-pulling-down method. (Y,Yb)VO₄ with strong wetting was grown by edge defined film-fed growth method and materials, which require moderate temperature gradient, such as Ca₈(La,Yb)₂(PO₄)₆O₂ and (Gd,Yb)₂SiO₅ were grown by Czochralski method. Strong dependence of the CT luminescence decay time and intensity on temperature was observed for Yb-doped LuAG and YAP. Super fast decay with 0.85 ns decay time was observed in Yb(30%) doped YAP at room temperature. Though the emission intensity is weak at room temperature, it exceeds several times that of PbWO₄. In addition, CT luminescence of Yb:YAP occurs at longer wavelength than in BaF₂, which enables the usage of glass-based photomultiplier for the detection. In addition, higher stopping power will be expected due to the higher density host compared with BaF₂.

IX-W-2 Onset Detection of Solid-State Phase Transition in Estrogen-Like Chemical via Terahertz Transmission Spectroscopy

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[*Appl. Phys. Lett.* **85**, 3914 (2004)]

Solid-state phase transition onset in an endocrine-disrupting estrogen-like chemical (1,4-naphthol) is detected using terahertz transmission spectroscopy. The appearance of two absorption peaks and the sudden

upsurge of terahertz-radiation power at 210 K indicate the onset of the solid-state phase transition. Differential scanning microscopy reveals a first-order phase transition at around 240 K while temperature-dependent x-ray diffraction analysis shows the occurrence of such phenomenon also at around 240 K. This demonstrates the sensitivity of the terahertz spectroscopic technique to phase transition since it provides a signal before such phenomenon actually occurs.

IX-W-3 Design Principle of Wide-Gap Fluoride Hetero-Structures for Deep Ultraviolet Optical Devices

OENZERFI, Riadh; ONO, Shingo; QUEMA, Alex; GOTO, Masahiro; SAKAI, Masahiro; SARUKURA, Nobuhiko; NISHIMATSU, Takeshi¹; TERAKUBO, Noriaki¹; MIZUSEKI, Hiroshi¹; KAWAZOE, Yoshiyuki¹; SATO, Hiroki¹; YOSHIKAWA, Akira¹; FUKUDA, Tsuguo¹
(¹Tohoku Univ.)

[*J. Appl. Phys.* **96**, 7655 (2004)]

The design of fluoride-based optical devices for deep ultraviolet applications is discussed. Variations in the band-gap energy and band structure with respect to composition are investigated for Li_(1-x)K_xBa_(1-y)Mg_yF₃ perovskites. The band-gap energy, lattice constant, and band structure of perovskitelike fluorides are estimated based on *ab initio* calculations within the local-density approximation. The lattice-matched double hetero-structure with direct band-gap compounds (Li_(1-x)K_xBa_(1-y)Mg_yF₃ on either LiBaF₃ or KMgF₃ substrates) is promising for fabrication.

IX-W-4 Terahertz Time-Domain Spectroscopy of Amino Acids and Polypeptides

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[*Biophys. J.* **L22** (2005)]

Frequency-dependent absorption coefficients and refractive indices of amino acids (glycine and L-alanine) and polypeptides (polyglycine and poly-L-alanine) in the wavenumber region from 7 to 55 cm⁻¹

were measured by terahertz timedomain spectroscopy. A vibrational band was observed at 45.5 cm^{-1} for polyglycine, which was assigned as an interchain mode. The reduced absorption cross sections of the amino acids and polypeptides show power-law behavior. The exponents are different between the monomers and polymers, and those of the two polypeptides suggest that the time dependences of the total dipole moments are similar in the timescale of subpico- to picoseconds.

IX-X Development and Research of Advanced Tunable Solid State Lasers

Diode-pumped solid-state lasers can provide excellent spatial mode quality and narrow linewidths. The high spectral power brightness of these lasers has allowed high efficiency frequency extension by nonlinear frequency conversion. Moreover, the availability of new and improved nonlinear optical crystals makes these techniques more practical. Additionally, quasi phase matching (QPM) is a new technique instead of conventional birefringent phase matching for compensating phase velocity dispersion in frequency conversion. These kinds of advanced tunable solid-state light sources, so to speak "Chroma Chip Lasers," will assist the research of molecular science.

In this projects we are developing Chroma Chip Lasers based on diode-pumped-microchip-solid-state lasers and advanced nonlinear frequency conversion technique.

IX-X-1 Spectroscopic Properties of Yb:GdVO₄ Single Crystal: Stark Levels, Selection Rules, and Polarized Cross Sections

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[OSA Topical Meeting of Advanced Solid State Photonics MF8 (2005)]

Spectroscopic properties Yb:GdVO₄ single crystals were investigated. We determined the stark levels and discussed about the selection rules. We found that the 1030.7-nm transition from the lowest level in upper manifold to the highest level in lower manifold was forbidden transition in π -polarization. The absorption cross section, stimulated emission cross section, and radiative lifetime were also evaluated. σ_{ab} and σ_{em} at 948-nm were 6.1 and 6.7×10^{-20} -cm², respectively. The calculated radiative lifetime of Yb:GdVO₄ was 313- μ s, and was consistent to the reported fluorescence lifetime of 320- μ s. From the wave-profiles of σ_{em} and polarized decay rate, the averaged wavelength of fluorescence was estimated to be 991-nm. If the pumping wavelength is set to 1000-nm, the absorption coefficient of 15 at.% Yb:GdVO₄ is still 9.8-cm⁻¹, and laser cooling effect should be observed. Obtained spectroscopic parameters in this work were consistent with the previous report of laser oscillation. Authors also indicate that Yb:GdVO₄ has enough absorption at pumping wavelength for laser cooling.

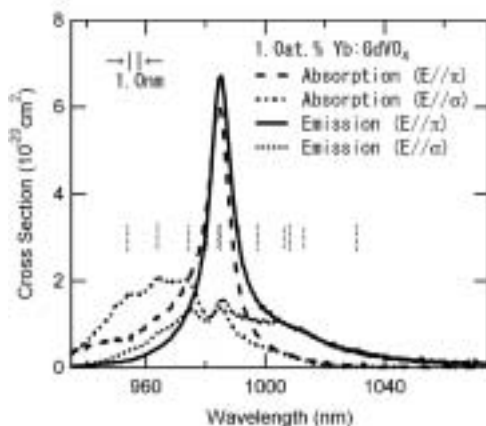


Figure 1. Polarized absorption and stimulated emission cross sections of Yb:GdVO₄.

IX-X-2 Spectroscopic Properties of All-Ceramic Composite with Layer-by-Layer of Nd:Y₃Al₅O₁₂ and Nd:Y₃ScAl₄O₁₂

SATO, Yoichi; TAIRA, Takunori; IKESUE, Akio¹
(¹JFCC)

[Conf. Lasers Electro-Optics JTuC28 (2005)]

The ceramic laser materials have been greatly interested because of their possibility for advanced lasers. The technique of composite ceramics has been especially considered to be the most important in the ceramics. Recently, this new ceramics technology about composite ceramics has been realized. In this work, we fabricated the all-ceramic composite with layer-by-layer of Nd:Y₃ScAl₄O₁₂ (YSAG) and Nd:Y₃Al₅O₁₂ (YAG).

The wide emission bandwidth of 5.5 nm in Nd:YSAG is composed of three transitions (R_1 - Y_1 , R_1 - Y_2 , R_2 - Y_3), and contains the valley in its profile. The ratio of peak-to-valley is almost 50%, which reaches 93% in Nd:YAG. Fortunately, the valley in the emission band of Nd:YSAG overlaps the peak in the emission band of Nd:YAG. When we pumped from YSAG side with 809.0 nm pumping source, this peak-to-valley improved to 43%. If we fabricate the three-layered composite of YAG, YSAG, and Y₃Sc₂Al₃O₁₂, this specimen will perform more flat emission band. These profiles of fluorescence in Figure 1 are related to the effective emission cross-section, thus we can design the gain profile of this layered composite material. Even in the case of diode-pumping, we can control the spectroscopic properties by tuning the temperature of laser diode.

As a result, we found that the tuning of the pumping wavelength made the spectral profile of fluorescence from the composite ceramics more even than that from single-layered Nd:Y₃Sc_xAl_{5-x}O₁₂ ceramics. The technique of composite ceramics is proved to be useful because it can provide not only the compositional tuning but also a new controllability by the selection of the pump source.

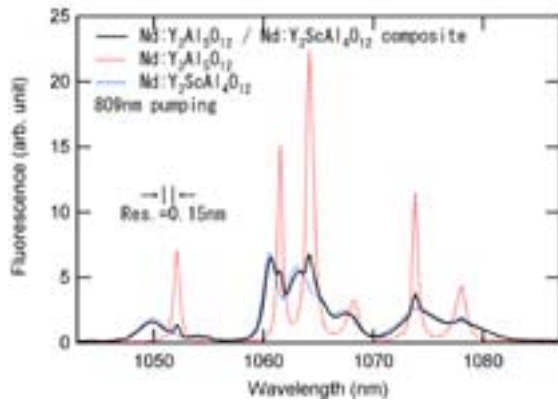


Figure 1. Fluorescence profiles of 1.0at.% Nd:YAG, Nd:YSAG, and their composite.

IX-X-3 Hybrid Process for Measurement of Spectroscopic Properties of Nd:GdVO₄

SATO, Yoichi; TAIRA, Takunori

[*Pac. Rim Conf. Lasers Electro-Optics CWI2-5* (2005)]

In order to construct highly performing solid-state lasers, laser materials should be able to absorb efficiently the pump radiation. The request of high pump absorption efficiency is more important for the case in microchip laser scheme, or under direct pumping into the emitting levels. Rare-earth doped vanadate crystals are the promised candidates solid-state lasers with high-performance because of their desirable properties of high optical absorption. However, there are many reports for an absorption cross section of the transition between $^4I_{9/2}$ and $^4F_{5/2}$ in Nd:GdVO₄. In this paper, authors discussed how to evaluate the spectroscopic properties of Nd- and Yb-doped GdVO₄ crystals.

We propose the “Hybrid Process” which is shown in Figure 1 as a technique for the spectroscopic evaluation in order to avoid overlooking the influences of emission in absorption and the influences of absorption in emission by simultaneous evaluation. This process also gives the radiative quantum efficiency (η_r). We studied Nd:GdVO₄ (Shandong Newphotonics) and Nd:YVO₄ (ITI) according to the Hybrid Process.

As a result, rare-earth doped vanadate crystals are the promised candidates solid-state lasers due to their large cross sections and relatively high thermal conductivity. In order to perform these superior characteristics, we have to notice that heavy heat generation under non-lasing condition and the difference in the thermal conductivity depending on the crystal-axis.

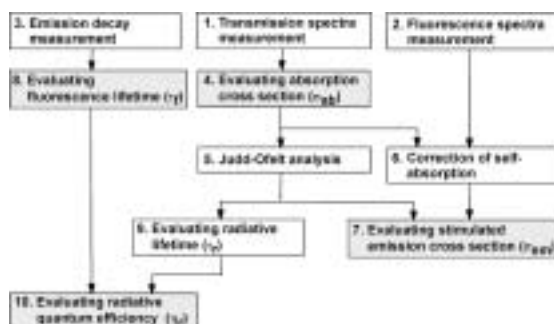


Figure 1. The flow-chart of “Hybrid Process” for measurement of the spectroscopic properties in Nd:vanadate.

IV-X-4 Absorption, Emission Spectrum Properties and Efficient Laser Performances of Yb:Y₃ScAl₄O₁₂ Ceramics

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(¹JFCC)

[*Appl. Phys. Lett.* **85**, 1898 (2004)]

We report on the continuous-wave laser performances of Yb³⁺-doped disordered Y₃Al₅O₁₂/Y₃Sc₂Al₄O₁₂ (YAG/YSAG) ceramics fabricated by sintering method. These new materials exhibit relatively low minimum pump intensity (I_{\min}) and broad emission bandwidth even in the yttrium aluminum garnet systems. The value of I_{\min} in the Yb:Y₃ScAl₄O₁₂ ceramics was found to be 2/3 compared with the Yb:YAG single crystal under 970-nm zero-line pumping. Efficient laser oscillation of 72% slope efficiency was obtained for input pump power. These materials attract great interest for high power femtosecond microchip lasers and amplifier applications.

IX-X-5 Passive Mode Locking of a Mixed Garnet Yb:Y₃ScAl₄O₁₂ Ceramic Laser

SAIKAWA, Jiro; SATO, Yoichi; TAIRA, Takunori; IKESUE, Akio¹
(¹JFCC)

[*Appl. Phys. Lett.* **85**, 5845 (2004)]

We have demonstrated a passively mode-locked Yb³⁺-doped Y₃ScAl₄O₁₂ ceramic laser, which is a solid solution of Y₃Al₅O₁₂ (YAG) and Y₃Sc₂Al₄O₁₂ (YSAG), by using a semiconductor saturable-absorber mirror (SESAM). The maximum average output power was as high as 150 mW with a pulse duration of 500 fs. With a 0.5% output coupler, pulses as short as 280 fs having an average output power of 62 mW at 1035.8 nm were obtained.

IX-X-6 High-Power Operation of Diode Edge-Pumped, Glue-Bonded, Composite Yb:Y₃Al₅O₁₂ Microchip Laser with Ceramic, Undoped YAG Pump Light-Guide

TSUNEKANE, Masaki; TAIRA, Takunori

[*Jpn. J. Appl. Phys.* in press]

The high-power operation of a diode edge-pumped, composite Yb³⁺-doped Y₃Al₅O₁₂ (Yb:YAG) microchip laser has been demonstrated. The novel composite microchip has a 5-mm-diameter, 300- μ m-thickness, single-crystal Yb:YAG core and a transparent ceramic, undoped YAG pump light-guide surrounding the core with the same thickness. The high-reflection coated face of the microchip was bonded to a water-cooled heat-sink by a thermally conductive glue for efficient con-

ductive cooling. The pump light from a fourfold diode stack directly coupled to the light-guide and maximum output powers of 410 W (peak) and 285 W have been successfully obtained at an input pump power of 866 W in quasi-continuous-wave (10 ms, 10 Hz) and continuous-wave operations, respectively, at room temperature.

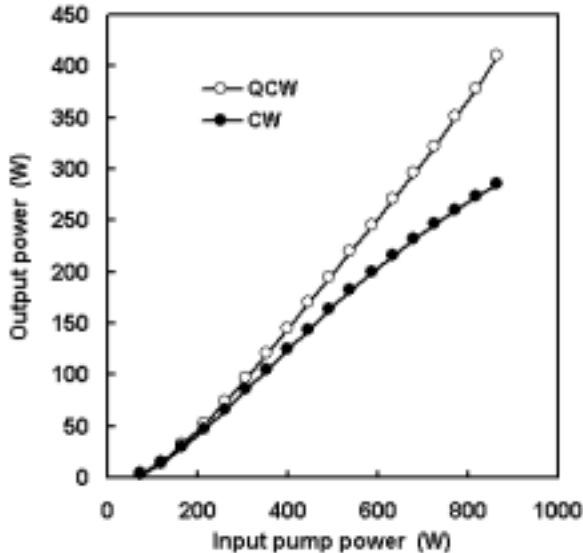


Figure 1. Input pump power versus laser output power of diode edge-pumped, composite Yb:YAG microchip laser in QCW and CW operations at room temperature.

IX-X-7 Continuous-Wave Deep Blue Generation in a Periodically Poled MgO:LiNbO₃ Crystal by Single-Pass Frequency Doubling of a 912-nm Nd:GdVO₄ Laser

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[*Jpn. J. Appl. Phys.* **43**, L1293 (2004)]

We report blue emission at 456 nm by single-pass second-harmonic generation in a bulk periodically-poled MgO:LiNbO₃ crystal, for the first time to our best knowledge. Using as a pumping source a continuous-wave Nd:GdVO₄ laser operating at 912 nm, blue power of 167 mW with 8.3% infrared-to-blue conversion efficiency and 4.2%/W normalized conversion efficiency was obtained. The present data, combined with our previous results on green generation, supports the prospect of obtaining compact and high power visible sources based on bulk periodically poled MgO:LiNbO₃.

IX-X-8 Continuous-Wave Ultraviolet Generation at 354-nm in a Periodically Poled MgO:LiNbO₃ by Frequency Tripling of a Diode End-Pumped Nd:GdVO₄ Microlaser

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[*Appl. Phys. Lett.* **85**, 3959 (2004)]

Irregular side spread of polarization inversion was suppressed in PPMgLN up to 2-mm thickness by using a multipulse poling method. Cascaded THG was performed by employing a first-order SHG-QPM PPMgLN followed by a SFG-QPM PPMgLN stage, and a 1063 nm Nd:GdVO₄ laser as a pumping source. CW power of 47 mW at 354 nm with 3.9%/W/cm normalized conversion efficiency was obtained. To our knowledge this is the first demonstration in PPMgLN of such a system and the highest UV power at 354 nm reported by CW single-pass process in a QPM structure. The present result supports the prospect of obtaining compact and high power UV source based on bulk periodically poled MgO:LiNbO₃.

IX-X-9 High-Power Continuous-Wave Intracavity Frequency-Doubled Nd:GdVO₄-LBO Laser under Diode Pumping into the Emitting Level

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(¹IAP-NILPRP, Romania)

[*IEEE J. Sel. Top. Quantum Electron.* **11**, 631 (2005)]

Continuous-wave emission in Nd:GdVO₄ at the 1- μ m fundamental wavelength and 531-nm second harmonic, which was obtained by intracavity frequency-doubling with LBO, is investigated under end-pumping with high-power diode laser at 808 and 879 nm. The maximum green power of 5.1 W with 0.31 overall optical-to-optical efficiency in a beam of $M^2 = 1.46$ was obtained under 879-nm pumping, directly into the ⁴F_{3/2} emitting level. To the authors best knowledge this is the highest CW green power obtained for the Nd:GdVO₄ laser material.

IX-X-10 Deep Blue Generation at 456 nm in a Periodically Poled MgO:LiNbO₃ Ridge-Type Waveguide by Single-Pass Frequency Doubling of a Nd:GdVO₄ Micro-Laser

PAVEL, Nicolaie¹; TAIRA, Takunori; IWAI, Makato²; YOSHINO, Takeshi²; IMAEDA, Minoru²
(¹IAP-NILPRP, Romania; ²NGK)

[*Conf. Lasers Electro-Optics CTuC30* (2005)]

This paper reports on our work toward scaling deep blue light by SHG-QPM process in a PPMgLN ridge-type waveguide: 93 mW power at 456 nm was achieved by single-pass SHG of a 912-nm Nd:GdVO₄ laser.

IX-X-11 Efficient 1.06 and 1.34- μ m Laser Emission of Highly-Doped Nd:YAG under 885-nm Diode Pumping into the Emitting Level

PAVEL, Nicolaie¹; TAIRA, Takunori
(¹IAP-NILPRP, Romania)

[*European Conf. Lasers Electro-Optics CA-19-MON* (2005)]

This work reports on 1.34- μm emission in Nd:YAG under direct pumping into the emitting level, at 885 nm, the first demonstration of such a system to the best of our knowledge. CW output power of 3.8 W at 1.34 μm with overall optical-to-optical efficiency of 0.26 is obtained from a 2.5-at.% Nd:YAG single crystal pumped into the $^4F_{3/2}$ level with a high-brightness 885-nm diode laser. The influence of the lasing wavelength and medium concentration on the thermal effects induced by optical pumping into the active component is discussed. It is shown that laser emission at 1.3 μm increases the thermal effects in medium only under a certain value of C_{Nd} . This behavior was checked by mapping the temperature of the output surface of the Nd:YAG crystals. These results demonstrate that diode laser pumping into the emitting level of concentrated Nd-based laser materials is a solution for construction of efficient micro-lasers and for scaling to high power.

IX-X-12 High-Power Multi-Pass Pumped Microchip Nd:GdVO₄ Laser

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(¹IAP-NILPRP, Romania)

[*Pacific Rim Conf. Lasers Electro-Optics* CTuI3-5
(2005)]

We describe a continuous-wave thin-disk Nd:GdVO₄ laser in a multi-pass pumping scheme. Employing a 250- μm thick, 0.5-at.% Nd:GdVO₄ crystal, a maximum output power of 13.9 W was obtained for 22 W absorbed power; the slope efficiency in absorbed power was 0.65. Pumping at 879 nm, directly into the $^4F_{3/2}$ emitting level, resulted in laser operation with slope efficiency in absorbed power of 0.69; the laser output power was 3.6 W at an absorbed pump power of 6.2 W. We appreciate that with an improved heat transfer between the Nd:GdVO₄ and the cooling system, a Nd:GdVO₄ medium of lower doping and an increased number of passes of the pump radiation, this geometry could be also a good solution for efficient laser emission at 912 nm.

IX-X-13 Highly Efficient New Pumping Configuration for Microchip Solid State Laser

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(¹IAP-NILPRP, Romania)

[*European Conf. Lasers Electro-Optics* CThu4-2
(2005)]

We propose a new pumping scheme which allows 94% absorption efficiency in Yb:YAG 15 at.% with 200 μm thickness with uniform pump distribution. This configuration can be scaled up and it is very attractive especially for high power quasi-4-levels lasers where the reabsorption losses are important.

IX-X-14 High Energy Quasi-Phase-Matched Optical Parametric Oscillation in a 3-mm-Thick Periodically Poled MgO:LiNbO₃ Device

ISHIZUKI, Hideki; SHOJI, Ichiro; TAIRA, Takunori

[*Opt. Lett.* **29**, 2527 (2004)]

We presented high energy optical parametric oscillation (OPO) using 3-mm-thick periodically poled MgO:LN (PPMgLN) device. The maximum total output energy of 22 mJ at the pump energy of 46 mJ was obtained using a Q-switched Nd:YAG laser of 30 Hz repetition rate with 15 ns pulse duration. The large-aperture PPMgLN devices would enable us to realize high-energy wavelength conversion, short pulse amplification, and pulse compression.