Laser Research Center for Molecular Science

IX-R 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-R-1 Photonic-Crystal-Fiber Pigtail Device Integrated with Lens-Duct Optics for Terahertz Radiation Coupling

DIWA, Girbert; QUEMA, Alex; ESTACIO, Elmer; POBRE, Romeric[1]; MURAKAMI, Hidetoshi; ONO, Shingo; SARUKURA, Nobuhiko
([1]De La Salle Univ.)

[*Appl. Phys. Lett.* **87**, 151114 (2005)]

An integrated optics called terahertz (THz) pigtail, which is comprised of an emitter, an optically transparent launching media, and a waveguide, is devised and fabricated. The InAs emitter under a 1 T magnetic field is coupled to the launching media using silicone grease, an index matching liquid. The launching media, a lens duct made from a polymer based on poly 4-methyl pentene-1 (commonly known as TPX), is designed based on the concept of guiding THz radiation into Teflon photonic crystal fiber (PCF) waveguide by means of total internal reflection. It is found that the constructed THz lens duct is able to channel and couple the THz radiation into the PCF waveguide with a loss of < 1 dB. The results here show that the idea of using the THz pigtail can be a potential means of effectively directing THz radiation.

IX-R-2 Band-Structure Design of Fluoride Complex Materials for Deep-Ultraviolet Light-Emitting Diodes

ONO, Shingo; EL OUENZERFI, Riadh; QUEMA, Alex; MURAKAMI, Hidetoshi; SARUKURA, Nobuhiko; NISHIMATSU, Takeshi[1]; TERAKUBO, Noriaki[1]; MIZUSEKI, Hiroshi[1]; KAWAZOE, Yoshiyuki[1]; YOSHIKAWA, Akira[1]; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.)

[*Jpn. J. Appl. Phys.* **44**, 7285 (2005)]

The design principle for fluoride-containing optical devices for applications in the deep ultraviolet range is discussed. Variations in band gap energy, band structure and lattice constant of $LiBa_xCa_ySr_{(1-x-y)}F_3$ and $Li_{(1-x)}K_xBa_{(1-y)}Mg_yF_3$ have been studied. The band structure and transition type of these fluorides are predicted by ab initio band calculations based on the local density approximation. The lattice-matched double-hetero structure of direct-band-gap compounds $LiBa_xCa_ySr_{(1-x-y)}F_3$ on $LiSrF_3$ and $Li_{(1-x)}K_xBa_{(1-y)}Mg_yF_3$ on either $LiBaF_3$ or $KMgF_3$ is sufficiently feasible to fabricate.

IX-R-3 Ce^{3+}-Doped $LiCaAlF_6$ Crystals as a Solid-State Ultraviolet Saturable Absorber and Role of Excited State Absorption

HAI, Le Hoang[1]; HUNG, Nguyen Dai[1]; QUEMA, Alex; DIWA, Girbert; MURAKAMI, Hidetoshi; ONO, Shingo; SARUKURA, Nobuhiko
([1]Inst. Phys. Elect.)

[*Jpn. J. Appl. Phys.* **44**, 7984 (2005)]

Nonlinear absorption properties of Ce^{3+}-doped $LiCaAlF_6$ (Ce:LiCAF) crystals at wavelength of 266 nm are studied using open-aperture Z-scan method and a Q-switch Nd:YAG laser. Saturable absorption of solid-state materials. in ultraviolet region is demonstrated for the first time.

IX-R-4 Generation of Terahertz Radiation Using Zinc Oxide as Photoconductive Material Excited by Ultraviolet Pulses

ONO, Shingo; MURAKAMI, Hidetoshi; QUEMA, Alex; DIWA, Girbert; SARUKURA, Nobuhiko; NAGASAKA, Ryujiro[1]; ICHIKAWA, Yo[1]; OGINO, Hiraku[2]; OHSHIMA, Eriko[2]; YOSHIKAWA, Akira[2]; FUKUDA, Tsuguo[2]
([1]Nagoya Inst. Tech.; [2]Tohoku Univ.)

[*Appl. Phys. Lett.* **87**, 261112 (2005)]

Terahertz (THz) radiation generated from photoconductive antenna fabricated on a single crystal zinc oxide (ZnO) is presented. The THz-radiation power is saturated at bias voltages above 800 V/cm and the obtained spectrum extends up to 1 THz. Moreover, ZnO is found to be highly transparent in the visible, near-infrared, mid-infrared and THz frequency regions. The results depicted here will categorically unravel the prospects of using ZnO as a material for integrated active optics.

IX-R-5 Ultraviolet Irradiation Effect of Ce^{3+}-Doped $BaMgF_4$ Crystals

HAYASHI, Eiji[1]; ITO, Kiyotaka[1]; YABASHI, Satoshi[1]; YAMAGA, Mitsuo[1]; KODAMA, Nobuhiro[2]; ONO, Shingo; SARUKURA, Nobuhiko

([1]Gifu Univ.; [2]Akita Univ.)

[*J. Alloys Compd.* **408**, 883 (2006)]

Ce^{3+}-doped $BaMgF_4$ (BMF) crystals have the absorption and luminescence spectra in the vacuum ultraviolet (VUV) and ultraviolet (UV) ranges. Strong excitation of the fourth harmonic (266 nm) of a pulsed Nd:YAG laser colours the BMF crystal brown and produces a new luminescence spectrum with double peaks at 445 and 500 nm and a lifetime of less than 10 ns. When the sample temperature is elevated up to 200 degrees C, the crystal colour is changed from brown to green. The colouration is due to localized electrons and holes created by the strong UV excitation, which are identified by the electron spin-resonance (ESR) technique. This new luminescence may be due to Ce^{3+} perturbed by the colour centres.

IX-R-6 Vacuum Ultraviolet and Ultraviolet Spectroscopy of $BaMgF_4$ Codoped with Ce^{3+} and Na^+

HAYASHI, Eiji[1]; ITO, Kiyotaka[1]; YABASHI, Satoshi[1]; YAMAGA, Mitsuo[1]; KODAMA, Nobuhiro[2]; ONO, Shingo; SARUKURA, Nobuhiko
([1]Gifu Univ.; [2]Akita Univ.)

[*J. Lumin.* **119**, 69 (2006)]

Ce^{3+}/Na^+-doped $BaMgF_4$ (BMF) crystals with a nonlinear property show strong absorption in the vacuum ultraviolet (VUV) and ultraviolet (UV) ranges. Three different fluorescence bands (A, B, C) at 300, 340, and 430 nm were observed when pumped at different wavelengths. Under excitation of the fourth harmonic (266 nm) from a pulsed Nd:YAG laser the BMF crystal changed its colour from transparent to brown due to formation of colour centres. The A, B and C bands are assigned to three different sites of Ce^{3+}: site A is Ce^{3+} substituting for perfect Ba^{2+} sites; site B is Ce^{3+} (Ba^{2+}) perturbed by Na^+ as a charge compensator; and site C is a complex composed of Ce^{3+} and F- vacancies, which trapped one or two electrons.

IX-R-7 Vacuum Ultraviolet Spectroscopy of Ce^{3+}-Doped $SrMgF_4$ with Superlattice Structure

YAMAGA, Mitsuo[1]; HAYASHI, Eiji[1]; KODAMA, Nobuhiro[2]; ITO, Kiyotaka[1]; YABASHI, Satoshi[1]; MASUI, Yuki[1]; ONO, Shingo; SARUKURA, Nobuhiko; HAN, T. P. J.[3]; GALLAGHER, H. G.[3]
([1]Gifu Univ.; [2]Akita Univ.; [3]Univ. Strathclyde)

[*J. Phys.: Condens. Matter* **18**, 6033 (2006)]

X-ray diffraction of Ce^{3+}-doped $SrMgF_4$ (SMF:Ce) crystals shows a superlattice structure, reflecting the distribution of Ce^{3+} polyhedra centres observed in optical experiments. Optical absorption bands and fluorescence bands from the Ce^{3+} polyhedra centres overlap in the vacuum ultraviolet (VUV) and ultraviolet (UV) regions, respectively, so that wide pumping and tuning ranges are expected for laser operation. The SMF: Ce crystals, as well as the isomorphous $BaMgF_4$, are candidates for a tunable laser gain material with nonlinear properties.

The optical absorption, excitation, and fluorescence bands observed in the SMF: Ce crystals at low temperatures are ascribed to five distinct fluorescent centres. Three centres have well-known Ce^{3+} optical characters, for example, fluorescence with double peaks separated by 2000 cm^{-1} and five resolved absorption/excitation bands. These centres are assigned to Ce^{3+}-polyhedra classified by weak and strong crystal fields as a consequence of the superlattice structure. The other two fluorescence bands observed in the visible region have 1.5–2 times larger linewidths than those of the former three bands. These bands are interpreted as optical transitions from complexes consisting of Ce^{3+} and one or two electrons trapped at a vacancy of the nearest neighbour F-ligand ions.

IX-S 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-sate lasers and advanced nonlinear frequency conversion technique.

IX-S-1 300W Continuous-Wave Operation of Diode Edge-Pumped, Hybrid Composite Yb:YAG Microchip Laser

TSUNEKANE, Masaki; TAIRA, Takunori

[*Opt. Lett*. **31**, 2003–2005 (2006)]

300 W continuous-wave operation of a diode edge-pumped, hybrid (single-crystal/ceramic) composite, Yb^{3+} :YAG microchip laser with a 5-mm-diameter and 300-µm-thickness, single-crystal core uniformly bonded to a water-cooled heat-sink by the new Au–Sn soldering system has been demonstrated. The beam quality factor, M^2 follows the mode-mismatch between the core and the fundamental mode, and was improved to 17 with the maximum output power of 230 W. The thermally induced convex mirror with a spherical radius of curvature ranging from –2.5 to –1.5 m, which decreases as the pump power increases by thermal deformation of a microchip, was observed.

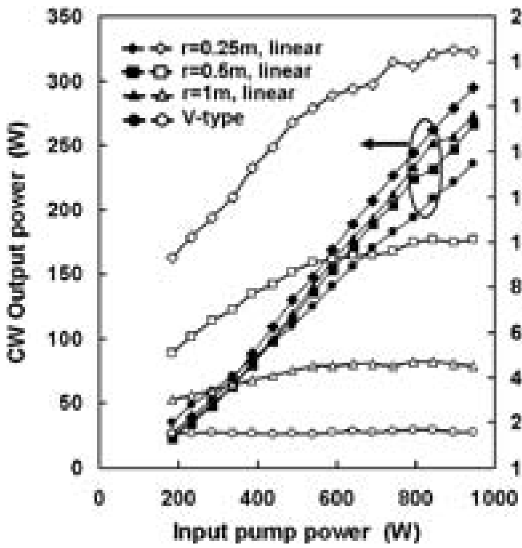

Figure 1. CW output power and the beam quality characteristics (M^2 factor) with four different cavity configurations as a function of input pump power.

IX-S-2 Spectroscopic Properties and Laser Emission in Layer-by-Layer Type Nd:$Y_3ScAl_4O_{12}$/Nd:$Y_3Al_5O_{12}$ Composite Ceramics

SATO, Yoichi; TAIRA, Takunori; IKESUE, Akio[1]
([1]JFCC)

[*OSA Topical Meeting of Advanced Solid State Photonics* TuB3 (2006)]

We have fabricated the all-ceramic layered composite device with Nd:YAG and Nd:YSAG, which can perform efficient laser oscillation. From its spectroscopic properties, this layer-by-layer composite device will offer new function of laser oscillation by pump wavelength tuning. For example when pumped from YSAG side at 810.5 nm, it can oscillate at 1064 nm. On the other hand, it will oscillate at 1061 nm when pumped at 808.5 nm.

Due to the difference in the dependence on the wavelength of, the portion of the pumped power absorbed in Nd:YAG-layer and in Nd:YSAG-layer depends on the pumping wavelength. This resulted in the tuning of the component ratio of the Nd:YAG and Nd:YSAG in the fluorescence. The dependence of fluorescence profiles in this composite on the pump wavelength is shown in Figure 1.

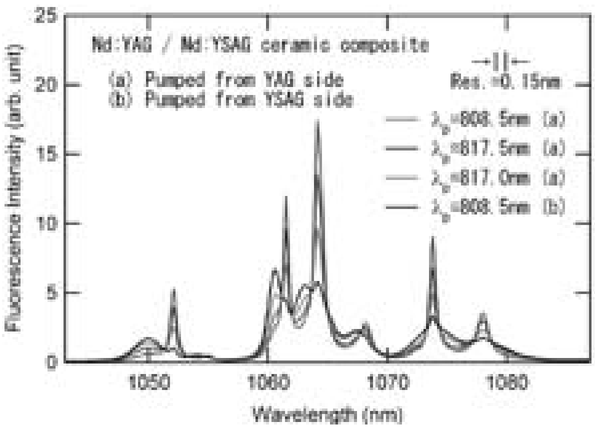

Figure 1. Measured fluorescent spectral profiles by changing pumping wavelength.

IX-S-3 Thermal Properties of $Y_3Al_5O_{12}$, $GdVO_4$, and YVO_4

SATO, Yoichi; TAIRA, Takunori

[*Conf. Lasers Electro-Optics* JThC30 (2006)]

We have measured thermal conductivity of $Y_3Al_5O_{12}$, $GdVO_4$, and YVO_4. In order to avoid the miss leading from three-dimensional (3D) thermal diffusion, we developed the quasi-one-dimensional (q1D) flash method. By taking in account the heat radiation effect in transparent materials for this measurement, YVO_4 was found to have larger thermal conductivity than $GdVO_4$. The measured thermal conductivities were 12.1, 10.5, 10.1, 8.9, and 8.5 W/mK for *c*-cut YVO_4, *c*-cut $GdVO_4$, YAG, *a*-cut YVO_4, and *a*-cut $GdVO_4$, respectively. The measured value in the range from room temperature to 200 °C is shown in Figure 1. The dependence of Nd-conductivity coefficient ($d\kappa/dC_{Nd}$) for convenient evaluation of the doping effect in thermal conductivity is also discussed.

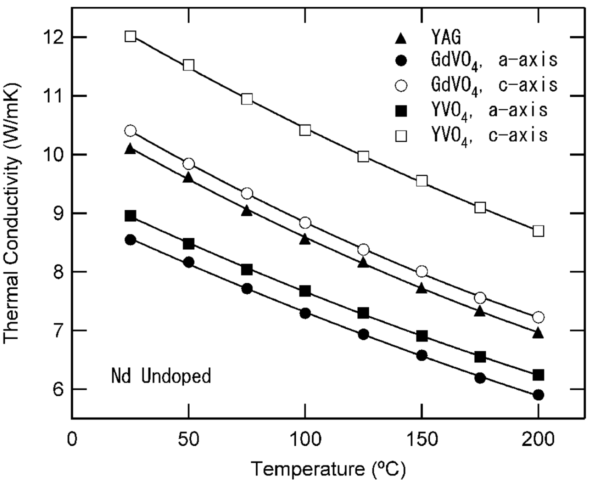

Figure 1. Thermal conductivities of undoped YAG, $GdVO_4$, and YVO_4.

IX-S-4 Spectroscopic Properties and Laser Operation of RE^{3+}-Ion Doped Garnet Materials

TAIRA, Takunori; SATO, Yoichi; SAIKAWA, Jiro; IKESUE, Akio[1]
([1]JFCC)

[*Proc. SPIE* **6216**, 62160J (2006)]

Lately developed RE^{3+}-ion-doped disordered laser ceramic materials, $Y_3Sc_xAl_{5-x}O_{12}$, which are a solid solution of YAG and $Y_3Sc_2Al_3O_{12}$ (YSAG), have been interested in because of its compositional tuning of parameter *x*. The disordered $Y_3ScAl_4O_{12}$ (YAG/YSAG) ceramics exhibit relatively low minimum pump intensity (I_{min}) and broad emission bandwidth. The value of I_{min} in the Yb:$Y_3ScAl_4O_{12}$ ceramics was found to be 2/3 compared with the Yb:YAG single crystal under 970nm zero-line pumping. Efficient laser oscillation of 72% slope efficiency was obtained for input power. Next, we have demonstrated passively mode-locked Yb:Y_3Sc Al_4O_{12} disordered ceramic laser by using a semiconductor saturable-absorber mirror. Pulses as short as 280 fs having an average power of 62 mW at 1035.8 nm was obtained as shown in Figure 1. As a conclusion, the possibility of tailored fluorescence spectral profile in layer-by-layer type ceramic composite is also discussed.

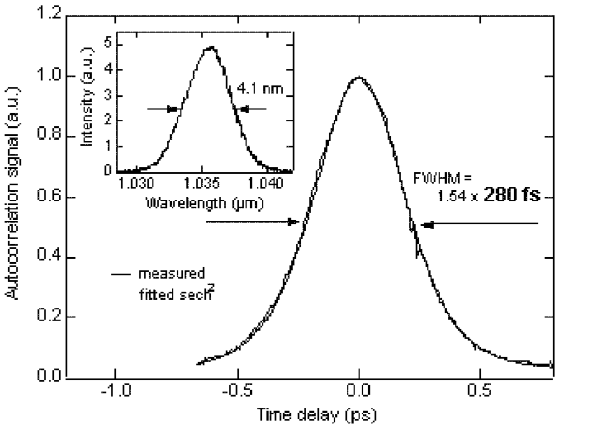

Figure 1. Second-harmonic autocorrelation trace and spectrum (inset) for the shortest-pulse generation with a 0.5% output coupler. The solid-line indicates ideal $sech^2$ shape.

IX-S-5 Comparative Study on the Spectroscopic Properties of Nd:$GdVO_4$ and Nd:YVO_4 with Hybrid Process

SATO, Yoichi; TAIRA, Takunori

[*IEEE J. Sel. Top. Quantum Electron.* **11**, 613 (2005)]

We have proposed the hybrid procedure of determining spectroscopic parameters for uniaxial solid-state laser crystals. Figure 1 shows the procedure of this process. By using our procedure the spectroscopic properties of Nd:$GdVO_4$ were evaluated and compared to those of Nd:YVO_4. The product of stimulated emission cross section and fluorescence lifetime ($\sigma\tau$ product) of Nd:$GdVO_4$ was smaller than that of Nd:YVO_4 under 1.0-at.% of Nd^{3+}-doping concentration. Because of the low value of radiative quantum efficiency of Nd:$GdVO_4$ (50%), careful cavity design is required for creating a well performing solid-state laser with Nd:$GdVO_4$, based on the larger $\sigma_{em}\tau_f$ product than $\sigma_{em}\tau_f$ product of Nd:YAG.

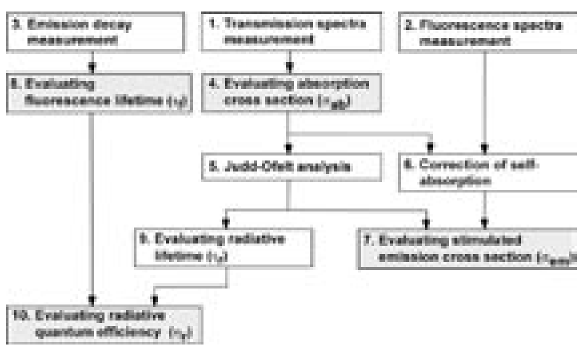

Figure 1. Flow-chart of the Hybrid process in this work.

IX-S-6 High-Energy, Narrow-Bandwidth 2-µm Optical Parametric Oscillator/Power Amplifier Based on Periodically Poled MgO:$LiNbO_3$

SAIKAWA, Jiro; FUJII, Masaaki[1]; ISHIZUKI, Hideki; TAIRA, Takunori

(*[1]Tokyo Tech*)

[*Conf. Lasers Electro-Optics* CThG3 (2006)]

We demonstrated a high-energy, high-efficiency quasi-phase-matched optical parametric oscillator/power amplifier system based on 5 × 5 mm^2 large aperture periodically poled $MgO:LiNbO_3$. Maximum pulse energy of 52 mJ (optical conversion efficiency of 60%) with resolution limit spectral bandwidth of < 2 nm at 2.128 µm degeneracy point was obtained. These experimental result shows that large aperture PPMgLN device is useful for the development of the high-energy, efficient, narrowband 2µm light source.

IX-S-7 High-Energy Quasi-Phase Matched Optical-Parametric Oscillation in Periodically Poled $MgO:LiNbO_3$ Device with 5 mm x 5 mm Aperture

ISHIZUKI, Hideki; TAIRA, Takunori

[*Opt. Lett.* **30**, 2918–2920 (2005)]

Fabrication of 5mm-thick periodically poled MgO-doped $LiNbO_3$ device with 32.1µm period for mid-infrared generation was demonstrated. The periodic structure was evaluated by measurement of second-harmonic generation with d_{31}-coefficient. Optical-parametric oscillation using the device with uncoated 5 mm × 5 mm aperture and 36mm effective length realized a high-energy output of 77 mJ for both signal (wavelength: 1.83 µm) and idler (2.54 µm) waves with 72% slope efficiency at 110mJ pumping of Q-switched Nd:YAG laser with 12ns pulse duration.