RESEARCH ACTIVITIES VIII
Laser Research Center for Molecular Science

VIII-A 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.

VIII-A-1 Intense THz Radiation from Femtosecond Laser Pulses Irradiated InAs in a Strong Magnetic Field

OHTAKE, Hideyuki; ONO, Shingo[1]; KAWAHATA, Eiji; LIU, Zhenlin; SARUKURA, Nobuhiko
([1]Sci. Univ. Tokyo)

Since the first observation of THz radiation from InAs surface irradiated with femtosecond laser pulses, considerable effort have been made to design an intense THz-radiation source and to understand the mechanism for generating THz radiation. However, the problem has not been solved. In this paper, we have investigated the intense THz radiation from InAs by applying a strong magnetic field up to 5 T. We compared several different geometries. Besides quadratic magnetic field dependence, we found saturation of the THz-radiation intensity around 3 T. Furthermore, the intensity decreased dramatically above 3 T. It represented that the most suitable magnetic field was 3 T to design an intense THz-radiation source. We also took spectra by a Polarizing Michelson interferometer. The spectral shapes for the different magnetic field directions were significantly different. The center frequency of these spectra shifted to lower frequency with increasing magnetic field. Through these experiments, we found the best configuration and the most suitable magnetic field to obtain an intense THz radiation for various applications such as imaging, sensing, and spectroscopy. This configuration dependence of the spectral shape and the center frequency is attributed to be the initial carrier acceleration processes modulated by a strong magnetic field.

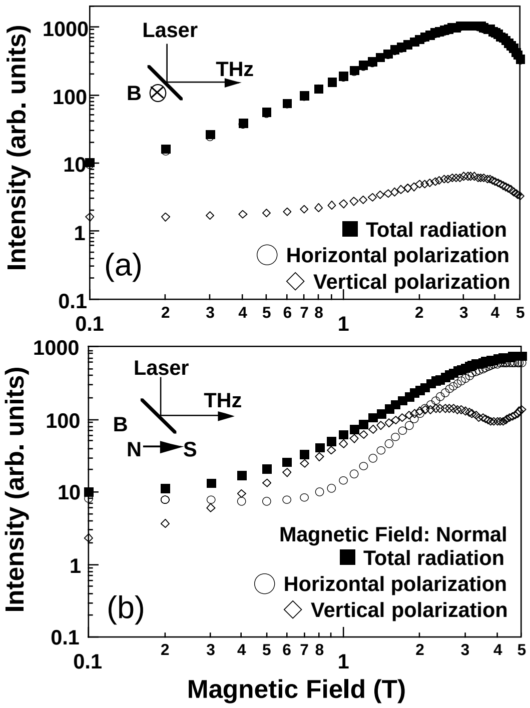

Figure 1. Magnetic field dependence of THz-radiation intensity. Inset indicates the experimental geometry. Closed squares, open circle and diamonds show total radiation, horizontal and vertical polarization, respectively. (a) The saturation of THz radiation intensity is clearly observed. (b) The saturation is not observed.

VIII-A-2 High-Repetition-Rate, High-Average-Power Mode-Locked Ti:Sapphire Laser with an Intracavity cw-Amplification Scheme

LIU, Zhenlin; ONO, Shingo[1]; KOZEKI, Toshimasa; OHTAKE, Hideyuki; SARUKURA, Nobuhiko
([1]Sci. Univ. Tokyo)

We have demonstrated a high-average-power, mode-locked Ti:sapphire laser with an intracavity cw-amplification scheme. The laser generated 150-fs pulses with 3.4-W average power at a repetition rate of 79 MHz. This simple amplification scheme can be applied for the power scaling of other lasers.

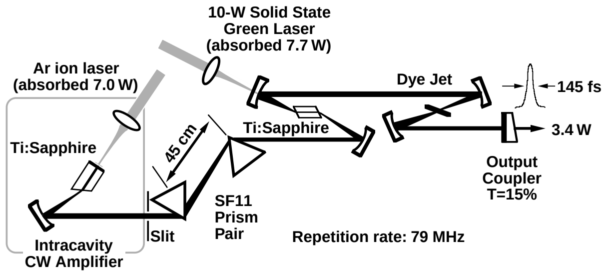

Figure 1. Configuration of high-repetition-rate high-average-power (3.4 W) femtosecond Ti:sapphire laser with an intracavity cw amplifier. The half-cut Brewster Ti:sapphire crystal composed the intracavity cw amplifier.

VIII-A-3 Compact THz-radiation Source Consisting of a Bulk Semiconductor, a Mode-Locked Fiber Laser, and a 2-T Permanent Magnet

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([1]Sci. Univ. Tokyo; [2]JASCO Corporation; [3]Sumitomo Special Metals co., Ltd; [4]IMRA AMERICA, INC. JLO)

Various THz-radiation sources have been intensively studied including photo conductive switches irradiated with ultrashort optical pulses. An intense, compact, and simple light source is required for applications in sensing or imaging. We have demonstrated the strong enhancement of THz-radiation power with a magnetic field by using an InAs semiconductor. In this paper, we report on a compact THz-radiation source consisting of a fiber femtosecond laser and a newly designed 2-T permanent magnet shown in Figure 1. A mode-locked frequency doubled Er-doped fiber laser delivered 170-fsec pulses at 780 nm with a 48.5-MHz repetition rate (IMRA model FA7850/10SA) with 30-mW average power and 4.1-kW peak power. The mode-locked fiber laser is a completely turn-key system. It is much smaller than a mode-locked Ti:sapphire laser that requires daily alignment. The used semiconductor sample was undoped bulk InAs with a (100) surface. The 2-T permanent magnet unit consisted of 8 Nd-Fe-B magnet pieces. The remanence magnetic field of the Nd-Fe-B material itself was 1.3 T (NEOMAX-44H). Owing to the new magnetic circuit design, the magnetic field in the center exceeded the remanence magnetic field of the material. The permanent magnet only weighs about 5 kg. The 2-T permanent magnet unit is smaller and much lighter than an electromagnet. At present the average power is estimated to sub-micro watt level. The spectra of the THz radiation were obtained by a Polarizing Michelson interferometer. Many water vapor absorption lines were clearly observed. Therefore, the THz-radiation source is already usable for spectroscopy. Such a simple and compact source will open up new application for THz-radiation.

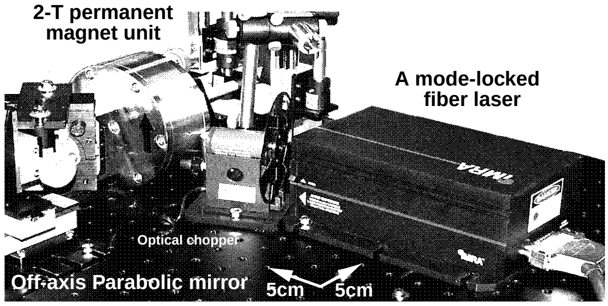

Figure 1. Photograph of a compact THz-radiation source with a bulk semiconductor, a fiber femtosecond laser, and a 2-T permanent magnet. Including the laser, the size is less than 40 × 30 × 15 cm.

VIII-A-4 Spectrum Control of THz Radiation from InAs in a Magnetic Field by Duration and Frequency Chirp of the Excitation Pulses

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The THz-radiation spectrum from InAs in a magnetic field irradiated with femtosecond pulses can be controlled by varying the excitation pulse width and chirp direction of the excitation pulse. A longer excitation pulse width produces lower frequency THz radiation. Also, positively chirped pulse excitation will generate higher power and higher frequency THz radiation, due to the corruption of the impulse response of the semiconductor in the longer pulse width region. The spectral shape of the radiation strongly depends on the chirp direction. This unexpected difference with the same excitation peak power and the same pulse duration with different chirp direction is rather surprising. This difference of THz-radiation for the chirping of the excitation pulses might be attributed to the difference of the photo-carrier relaxation process in the conduction band with oppositely chirped-pulse excitation.

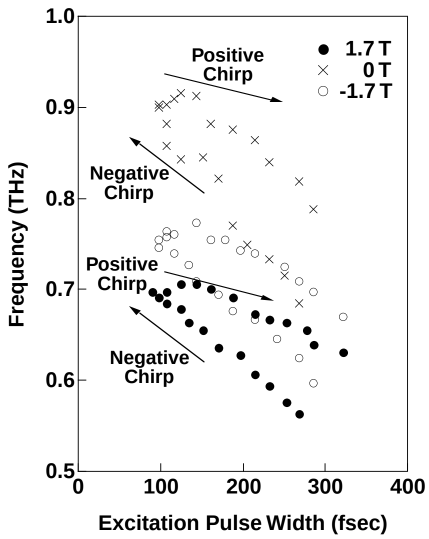

Figure 1. Center frequency spectrum dependence of THz radiation with different excitation chirp, pulse duration and magnetic field. Close circle, open circle and cross show 1.7 T, –1.7 T and 0 T, respectively.

VIII-A-5 LiCAF Crystal as a New Vacuum Ultraviolet Optical Material with Transmission Down to 112 nm

KOZEKI, Toshimasa; SAKAI, Masahiro; LIU, Zhenlin; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; SEGAWA, Yusaburo[1]; OBA, Tomoru[2]; SHIMAMURA, Kiyoshi[3]; BALDOCHI, Sonia L.[3]; NAKANO, Kenji[3]; MUJILATU, Na[3]; FUKUDA, Tsuguo[3]
([1]RIKEN; [2]Optron Inc.; [3]Tohoku Univ.)

$LiCaAlF_6$ (LiCAF) was found to be an ideal optical material for the vacuum ultraviolet region due to its superior transmission characteristic of down to 112 nm, its non hydroscopic nature, and its better mechanical properties compared with LiF.

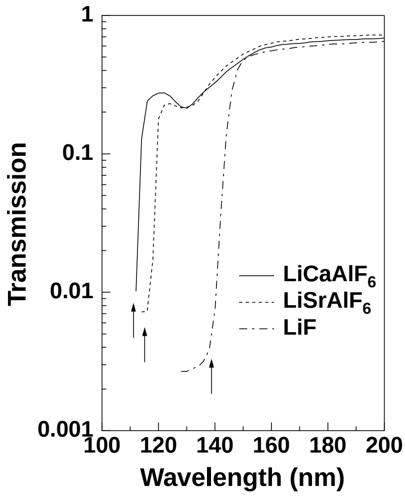

Figure 1. Transmission characteristics of LiCAF, LiSAF, LiF.

VIII-A-6 High Average Power Mode Locked Ti:Sapphire Laser with Intracavity Continuous-Wave Amplifier and Strained Saturable Bragg Reflector

LIU, Tze-An; HUANG, Kai-Fung; PAN, Ci-Ling; LIU, Zhenlin; OHTAKE, Hideyuki; SARUKURA, Nobuhiko

We demonstrate a new scheme for the generation of high average power femtosecond pulses by incorporating an intracavity amplifier and a strained saturable Bragg reflector with low saturation fluence for self-starting mode locking. When the Ti:sapphire oscillator and intracavity amplifier are pumped at 10 W and 15 W, respectively, the average output power is as high as 1.62 W. The pulse duration is about 145 fs, and the peak power reaches 160 kW at a 68 MHz repetition rate. The pulse-formation time of 400 μs is sufficiently short to sustain stable mode locking.

VIII-A-7 THz Radiation from Intracavity Saturable Bragg Reflector in Magnetic Field with Self-Started Mode-Locking by Strained Saturable Bragg Reflector

LIU, Tze-An; HUANG, Kai-Fung; PAN, Ci-Ling; LIU, Zhenlin; ONO, Shingo; OHTAKE, Hideyuki; SARUKURA, Nobuhiko

We demonstrate a new configuration for intracavity generation of THz radiation. A magnetic-field-biased saturable Bragg reflector (SBR) located inside the femtosecond laser cavity is the emitter, while a strained saturable Bragg reflector (SSBR) achieves self-started mode-locking without focusing. The calibrated power of the emitted THz radiation is estimated to be approximately 45 nW with a peak frequency at 0.72 THz and width of approximately 0.7 THz under a 0.88 T magnetic field. The quadratic dependence of THz-radiation power by the SBR on the magnetic field is also observed for the first time.

VIII-A-8 High-Gain, Reflection-Double Pass, Ti:Sapphire Continuous-Wave Amplifier Delivering 5.77 W Average Power, 82 MHz Repetition Rate, Femtosecond Pulses

LIU, Zhenlin; MURAKAMI, Hidetoshi; KOZEKI, Toshimasa; OHTAKE, Hideyuki; SARUKURA, Nobuhiko

A confocal, reflection-double-pass, Ti:sapphire continuous-wave ~cw! amplifier with a small signal gain of 4.2 has been invented. Femtosecond pulses with an 82 MHz repetition rate from a mode-locked Ti:sapphire laser are amplified to 5.77 W average power with a slightly saturated gain of 3.7 through an amplifier pumped by three cw green lasers, and the extraction efficiency reaches 10.6%.

VIII-B 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.

VIII-B-1 Performance of Widely Tunable Yb:YAG Microchip Lasers

SAIKAWA, Jiro; KURIMURA, Sunao; PAVEL, Nicolaie; SHOJI, Ichiro; TAIRA, Takunori

[*OSA TOPS on ASSL 2000* **34**, 106 (2000)]

The properties of the trivalent ytterbium ion doped YAG, such as a smaller quantum defect, longer upper state lifetime, simple energy structure (no excited state absorption) and so on, would promise to achieve a high power and high stability laser operation in microchip configurations. In addition, the wide emission bandwidth of the material around 1 µm allows tunable and/or mode-locked operation. In this work, we have developed a tunable intracavity frequency-doubled Yb:YAG microchip laser that outputs a maximum green power of 520mW with single frequency around the Ar^{3+} laser wavelength of 515 nm by using a 400-µm thick Yb:YAG. By using LBO crystal a wide tuning range from 515.3 to 537.7 nm ($\Delta\lambda$ = 22.4 nm, $\Delta\nu$ = 24.4 THz) was obtained (Figure 1). Then, we applied this tunable green laser to a wavelength-multiplexing holographic memory. We recorded 3 discrete images at 3 different wavelengths in the same position of a 600 ppm Fe-doped $LiNbO_3$ crystal and each image was reconstructed at each wavelength. The laser was proven to have a narrow linewidth and a wide tunability, in order to satisfy wavelength-multiplexing in the holographic storage system as shown in Figure 2. Next, in order to evaluate the potential of Yb:YAG tunability, the wide-bandwidth reflectivity dielectric mirror was deposited directly onto the Yb:YAG microchip. The output coupling mirror had a radius of curvature of 30 mm, and the cavity length was 25 mm. Experimental result of the tunability of the Yb:YAG microchip laser for different output couplers. With a reflectivity of 99.9% around 1010~1100 nm, the widest tunability of 84.5 nm, from 1024.1 to 1108.6 nm was obtained. The output beam was coupled as partially reflected beam at the birefringent filter. The oscillation range that extends beyond the Yb:YAG gain bandwidth, 9.5 nm, was realized since it has a simple energy- level manifolds. If it possible to keep wide band ($\Delta\nu$ ~ 22.4 THz) laser oscillation under mode-locking operation, transform-limited pulsewidth of approximately 50 fs should be feasible. The tuning bandwidth increased by using the high-reflectivity output coupler and peak wavelength shifted to longer wavelength. The shorter band-edge was limited by increase of reabsorption loss in the Yb:YAG, and longer band-edge by coating bandwidth in our experiment. The bandwidth of 22.4 THz indicates the potential of mode-locked operation of the Yb:YAG laser.

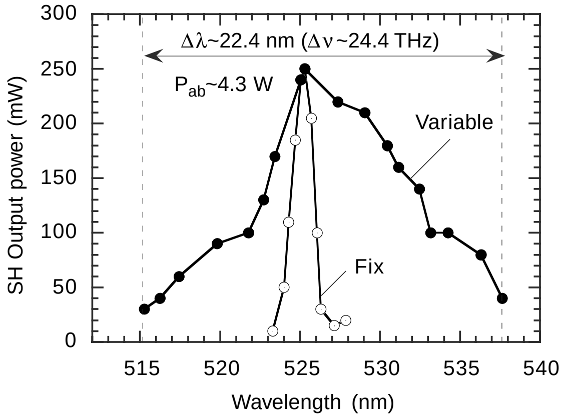

Figure 1. Tuning curve of the intracavity frequency-doubled Yb:YAG microchip laser.

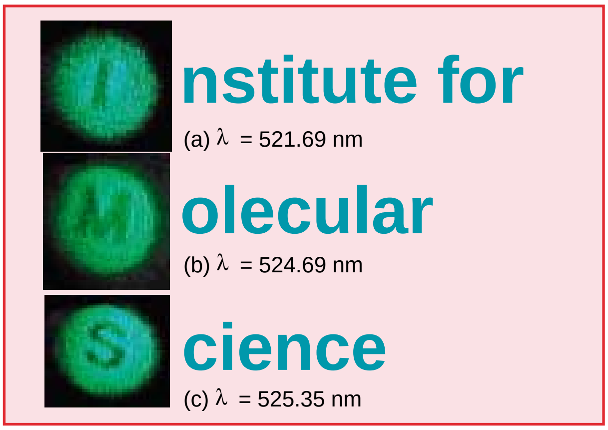

Figure 2. Reconstructed images from wavelength-multiplexed holograms by the tunable intracavity frequency-doubled Yb:YAG laser.

VIII-B-2 High Average Power Diode-Pumped Composite Nd:YAG Laser with Cr^{4+}:YAG Saturable Absorber for Passive Q-Switching

PAVEL, Nicolaie; KURIMURA, Sunao; SHOJI, Ichiro; SAIKAWA, Jiro; TAIRA, Takunori

[*CLEO/Europe* CWB 0006 (2000)]

Passive Q-switching technique is attractive for scientific and industrial applications that do not require accurate repetition rates. This technique can significantly simplify the operation, improve the efficiency, the reliability and the compactness, and reduce the laser costs. In a composite rod both the peak temperature rise and the thermal stress induced by optical pumping are reduced compared to a nonbonded crystal. In this work we show that these improvements, which reduce the thermal lensing effect, made a composite medium a good solution for improving output performances of high average power passively Q-switched lasers.

In experiments we used a composite Nd:YAG rod fabricated by diffusion bonding of a Nd:YAG (5-mm length, 1.1-at.% Nd doping) to an undoped YAG piece (1-mm length). The medium was end-pumped by a 1.55-mm diameter, 0.11-NA OPC fiber-bundles diode. With a plane-plane resonator of 80-mm length and an output mirror of 95% reflectivity at 1064nm, this configuration delivers a cw maximum output power of 7.7 W with an optical efficiency of 36.9%. The slope efficiency is 39.4%, and the laser beam M^2 factor varies among 1.1 and 2.3 on the pump power range. With a Nd:YAG medium (10-mm length, 1.3-at.% Nd doping), the maximum cw power was 8.2-W at 21.8-W absorbed power, the slope efficiency was 41.6%, and the laser beam M^2 factor varies between 1.2 and 3.3 on the pump power range. Cr^{4+}:YAG crystals with varying low-signal transmission T_0 have been used, as well as resonators of various length and output couplings. As an example, Figure 1 shows the average output power for the composite medium and Cr:YAG absorbers of T_0 = 89%, 85%, and 80%. A plane-plane resonator of 80-mm length with an R = 90% output coupler was considered. A maximum average power of 4.21-W in a laser beam of M^2 = 1.3 resulted for the Cr:YAG absorber of T_0 = 89%. The laser generated pulses of 48-ns duration at 24-kHz (~3.65 kW peak power). With the Nd:YAG medium a maximum average power of 3.9-W in a beam of M^2 = 1.9 resulted. When the resonator length was of 40-mm and a Cr:YAG crystal of T_0 = 80% was used, the composite Nd:YAG laser outputs a maximum average power of 2.6-W in a beam of M^2 = 1.45. The pulse width was 17.5-ns, the pulse energy is 0.285-mJ, and the peak power is 16.3-kW. Using the Nd:YAG medium, the maximum average power and beam M^2 factor decreases to 1.8-W and 1.9, respectively. Shorter resonators in spite of reducing the average output power will increases the Q-switched pulse peak power. This way a simple and compact-pumping source for parametric conversion into mid-IR region could be obtained.

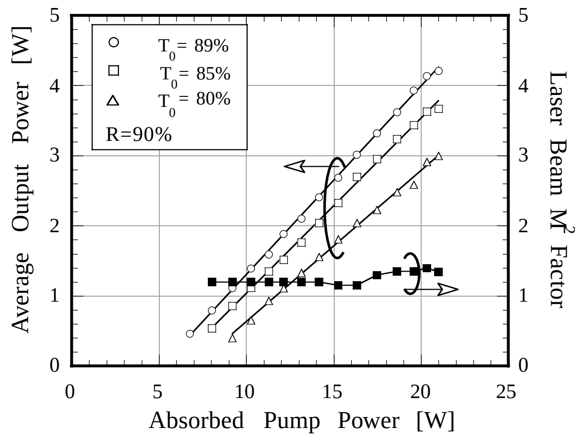

Figure 1. Average output power and beam M^2 factor as a function of absorbed pump power for the composite Nd:YAG, Cr:YAG absorbers.

VIII-B-3 Optical Properties and Laser Characteristics of Highly Nd^{3+}-Doped $Y_3Al_5O_{12}$ Ceramics

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([1]JFCC; [2]Osaka Inst. Tech.)

[*Appl. Phys. Lett.* **77**, 939 (2000)]

Diode-pumped microchip solid-state lasers have such advantages as compactness, high efficiency, high power, and low numbers of longitudinal and transverse modes. A wide variety of materials have been investigated to develop more efficient and higher power microchip lasers. Although $Nd:YVO_4$ is a suitable material for highly efficient microchip laser owing to its large absorption cross section, high power operation is difficult because its thermo-mechanical properties are poor. On the other hand, while Nd:YAG has good thermal properties, highly efficient microchip laser has never been reported since high doping (> 1 at. %) of neodymium into the YAG crystal is impossible, limiting pump absorption. Recently developed transparent Nd:YAG ceramics are attractive materials because high doping of neodymium is possible without degrading the thermal conductivity. In this work we report measurements of the absorption spectra and the fluorescence lifetime of ceramic Nd:YAG in order to show that it is a promising material as a highly efficient and high power microchip laser. Moreover, we characterized its laser performance. Figure 1 shows the absorption spectra of 2.2 and 4.8 at. % Nd:YAG ceramics and 1.1 at. % single crystal. The 4.8 at. % Nd:YAG ceramic has an absorption coefficient (30.4 cm^{-1}) as large as that of $Nd:YVO_4$.

The input-output power characteristics of 2.4 and 4.8 at. % ceramics and a 0.9 at. % single crystal with microchip structures (the thickness of laser medium < 1mm) are shown in Figure 2. For the 4.8 at. % ceramic, 2.3 times higher output was achieved than that for the single crystal, which indicates the advantage of Nd:YAG ceramics as highly efficient miniature or microchip lasers. We estimated the round-trip cavity losses by obtaining the slope efficiencies with different output-couplers. From this, we found that the loss of the 2.4 at. % ceramic is as low as that of the single crystal. It is concluded that highly Nd^{3+}-doped YAG ceramics are promising as a highly efficient, high-power microchip laser material.

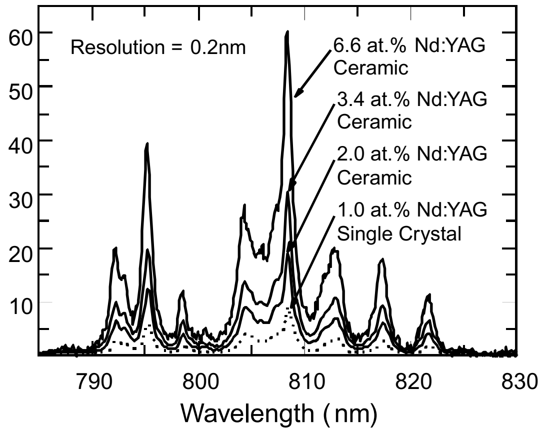

Figure 1. Absorption spectra of Nd:YAG ceramics (solid curves) and Nd:YAG single crystal (dashed curve).

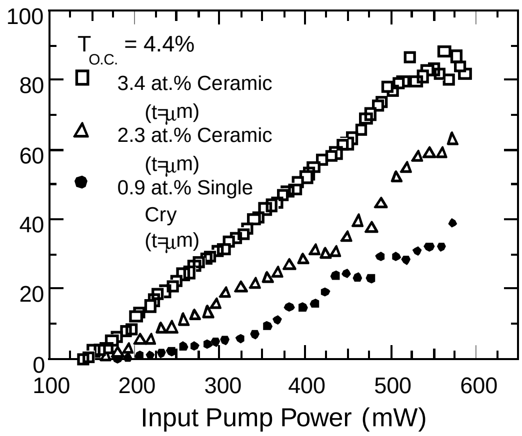

Figure 2. Dependence of the output power on the input pump power for the Nd:YAG ceramics and the single crystal.

VIII-B-4 Development of Multifunction Nonlinear Optical Wavelength Converter

KURIMURA, Sunao; PAVEL, Nicolaie; SHOJI, Ichiro; TAIRA, Takunori

[*Autumn Meeting of Japan Applied Physics Society* 3a-Q-22 (2000)]

Wavelength conversion based on nonlinear optics yields high efficiency without any sacrifice in coherency. Recently developed *Quasi-Phase Matching (QPM)* technique has produced designability of phase matching wavelength and efficiency, together with artificial characteristics of converters in space, frequency, and time domains by using digital patterns defined by photolithography. QPM giving new degree of freedom thus brought about stronger impact than growth of new crystals in the nonlinear optics. We here proposed efficient wide-band infrared generation with cascaded QPM crystals for optical parametric oscillation (OPO) and difference frequency generation (DFG) around 6 µm region, where double bond structures of molecules have characteristic absorption lines. Figure 1 illustrates the geometry, measured emission wavelengths in OPO and required QPM period in DFG in efficient nonlinear crystal, lithium niobate (LN). Since LN has moderate absorption at 6 µm, direct access to 6 µm induces thermal fluctuation enhanced in OPO cavity. Our approach is to use OPO in the transparent region of LN and access to 6 µm by single-pass DFG, which is less sensitive to thermal disturbance. We devised first OPO stage and obtained IR emission plotted in Figure 1(b) with closed circles. The 0.5mm-thick QPM device with a period of 30.9 µm was fabricated by electric field poling in liquid electrodes as shown in Figure 1(d).

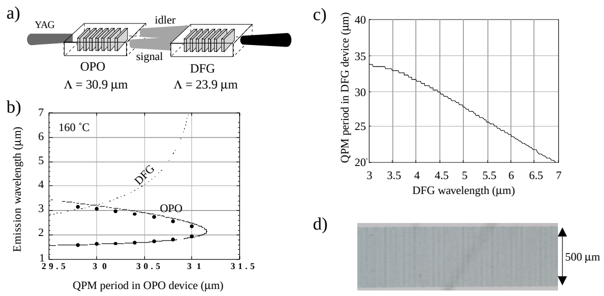

Figure 1. QPM OPO-DFG light source for widely tunable IR light: a) geometry, b) wavelength tunability depending on QPM period, c) calculated DFG period, d) periodical domains in a QPM LN device.

VIII-B-5 Periodical Twinning in Crystal Quartz for Ultraviolet Nonlinear Optics

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([1]Stanford Univ., U. S. A.; [2]Waseda Univ.)

[*Oyobuturi* **69**, 548 (2000)]

Crystal quartz is attractive for operation in ultraviolet nonlinear optics, which has low absorption from 150 nm, high chemical stability, and low thermooptic coefficients compared with conventional ultraviolet nonlinear crystals. Growth techniques are well established because of widespread in surface-acoustic-wave and timing applications, but unfortunately, it doesn't meet the birefringent phase matching condition due to small birefringence, and electric field poling condition due to lack of ferroelectricity. We devised a new poling technique in crystal quartz using mechanical twinning and demonstrated periodical polarity reversal by using thermal stress. Figure 1 shows an observed twin structure with a period of 80 µm, obtained by thermally induced stress between patterned Cr films and a quartz. The Cr patterned substrate was heated to just below Curie temperature in order to attain reasonable film stress and reduce coercive stress. Twins tend to generate from the edge of Cr pattern and the required duty ratio of Cr to the period was more than 0.5. The depth of twins, however, were several microns, indicating not suitable for bulk nonlinear optics. New technique is under development to improve the depth profile of the twins for a practical UV generator.

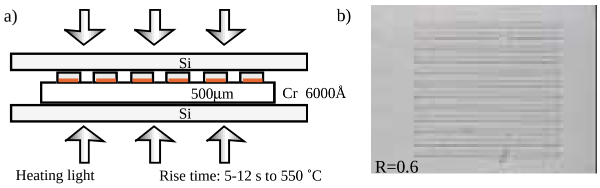

Figure 1. Twin patterning in crystal quartz: a) patterning method by the thermally induced in-plane stress, b) observed periodical twins with a period of 80 µm period, *R*: duty ratio of the Cr film to the period.