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 Ce^{3+}:$LiCaAlF_6$ Crystal for High-Gain or High-Peak-Power Amplification of Ultraviolet Femtosecond Pulses and New Potential Ultraviolet Gain Medium: Ce^{3+}:$LiSr_{0.8}Ca_{0.2}AlF_6$

LIU, Zhenlin[1]; KOZEKI, Toshimasa; SUZUKI, Yuji; SARUKURA, Nobuhiko; SHIMAMURA, Kiyoshi[2]; FUKUDA, Tsuguo[2]; HIRANO, Masahiro[1]; HOSONO, Hideo[1,3]
([1]ERATO; [2]Tohoku Univ.; [3]Tokyo Inst. Tech.)

[*IEEE J. Sel. Top. Quantum Electron.* **7**, 542 (2001)]

To develop high-peak-power ultrashort pulse laser systems in the ultraviolet region, a large Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) crystal, a tunable ultraviolet laser medium with large saturation fluence and broad gain spectrum width, was grown successfully with a diameter of more than 70 mm. To demonstrate high small signal gain, a four-pass confocal amplifier with 60 dB gain and 54 mJ output energy was constructed. Chirped pulse amplification (CPA) in the ultraviolet region was demonstrated using Ce:LiCAF for higher energy extraction. A modified bow-tie-style four-pass amplifier pumped by 100-mJ 266-nm 10-Hz pulses from a Q-switched Nd:YAG laser had 370-times gain and delivered 6-mJ 290-nm pulses. After dispersion compensation, the output pulses can be compressed down to 115 fs. This is the first ultraviolet, all-solid-state high-peak-power CPA laser system using ultraviolet gain media, and this demonstration shows further scalability of the Ce:LiCAF CPA system. Additionally, a new gain medium, Ce^{3+}:$LiSr_{0.8}Ca_{0.2}AlF_6$, with longer fluorescence lifetime and sufficient gain spectrum width over 18 nm was grown to upgrade this system as a candidate for a final power amplifier gain module.

VIII-A-2 Optical Fiber for Deep Ultraviolet Light

OTO, Masanori[1]; KIKUGAWA, Shinya[2]; SARUKURA, Nobuhiko; HIRANO, Masahiro[3]; HOSONO, Hideo[3,4]
([1]Showa Electric Wire Cable; [2]Asahi Glass Co. Ltd.; [3]ERATO; [4]Tokyo Inst. Tech.)

[*IEEE Photonics Technol. Lett.* **13**, 978 (2001)]

Deep ultraviolet optical fibers are fabricated using modified SiO_2 glasses containing 2000-ppm fluorine for the clad and 200 ppm for the core. The transmission at 193 nm is improved to more than 60%/m by optimizing the fiber drawing condition. The H-2-impregnation into the fiber suppresses the degradation of the transmission by irradiation of ArF excimer laser (50 mJ/cm^2/pulse). Further improvement may be expected by reducing oxygen-deficient center (I) defect generation in the drawing process.

VIII-A-3 Crystal Growth of Fluorides for Optical Applications

SHIMAMURA, Kiyoshi[1]; SATO, Hiroki[1]; BENSALAH, Amina[1]; SUDESH, Vikas[1]; MACHIDA, Hiroshi[2]; SARUKURA, Nobuhiko; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.; [2]Tokin Corp.)

[*Cryst. Res. Technol.* **36**, 801 (2001)]

Ce-doped and undoped $LiCaAlF_6$, $LiSrAlF_6$, $LiYF_4$, $LiLuF_4$ and $KMgF_3$ single crystals were grown by the Czochralski technique under CF_4 atmosphere. The effective distribution coefficients of Ce^{3+} in $LiCaAlF_6$, $LiSrAlF_6$, $LiYF_4$ and $LiLuF_4$ were determined to be 0.031, 0.028, 0.116 and 0.054, respectively. Laser output energy of 60 mJ and 27 mJ were obtained using the grown Ce:$LiCaAlF_6$ and Ce:$LiLuF_4$ single crystals, respectively. Undoped $LiCaAlF_6$ and $KMgF_3$ single crystals showed a transmission edge at 112 nm and 115 nm, respectively.

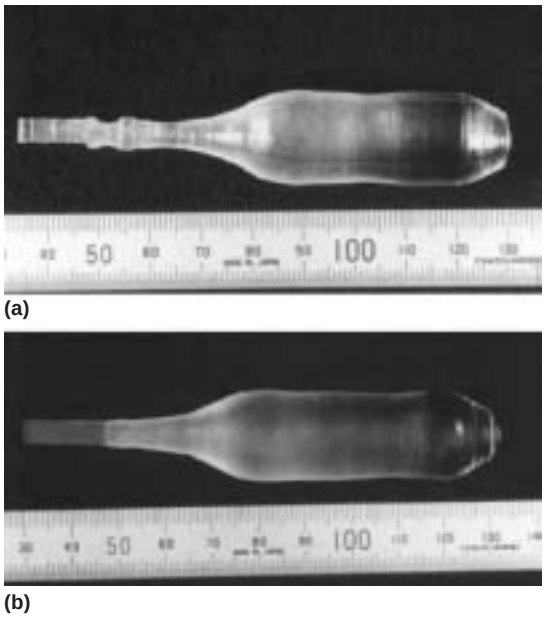

Figure 1. As-grown Ce-doped (a) $LiCaAlF_6$ and (b) $LiSrAlF_6$ single crystals 18 mm in diameter pulled along the *a*-axis.

VIII-A-4 Growth of Ce-Doped Colquiriite- and Scheelite-Type Single Crystals for UV Laser Applications

SHIMAMURA, Kiyoshi[1]; SATO, Hiroki[1]; BENSALAH, Amina[1]; MACHIDA, Hiroshi[2]; SARUKURA, Nobuhiko; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.; [2]Tokin Corp.)

[*Opt. Mater.* **19**, 109 (2002)]

Ce-doped Colquiriite- and Scheelite-type fluoride single crystals were grown by the Czochralski technique. The formation of inclusions and cracks accompanying the crystal growth was investigated. The effective distribution co-efficients of Ce^{3+} in $LiCaAlF_6$, $LiSrAlF_6$, $LiYF_4$ and $LiLuF_4$ were determined to be 0.031, 0.028, 0.116 and 0.054, respectively. Ultraviolet pulse generations with an output energy of 60 and 27 mJ were obtained from Ce:$LiCaAlF_6$ and Ce:$LiLuF_4$ lasers.

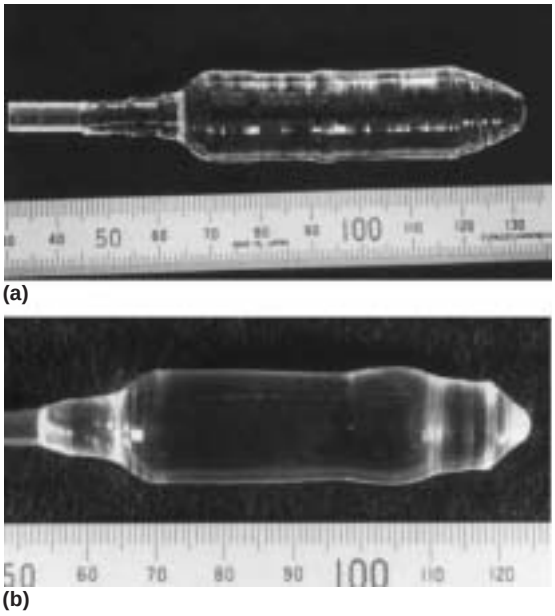

Figure 1. As-grown Ce-doped: (a) $LiYF_4$ and (b) $LiLuF_4$ single crystals of 18 mm in diameter pulled along the *a*-axis.

VIII-A-5 High-Energy Pulse Generation from Solid-State Ultraviolet Lasers Using Large Ce:Fluoride Crystals

LIU, Zhenlin[1]; SHIMAMURA, Kiyoshi[2]; FUKUDA, Tsuguo[2]; KOZEKI, Toshimasa; SUZUKI, Yuji; SARUKURA, Nobuhiko
([1]ERATO; [2]Tohoku Univ.)

[*Opt. Mater.* **19**, 123 (2002)]

A large Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) crystal with 15 mm diameter was grown successfully by the Czochralski method. Owing to its large size, 60 mJ, 289 nm pulses were generated directly from a quasi-coaxially pumped Ce:LiCAF laser. In addition, a new noncollinear Brewster-angle-pumping disk oscillator scheme was demonstrated for further output-energy scaling. An ultraviolet solid-state Ce^{3+}:$LiLuF_4$ (Ce:LLF) laser which was pumped transversely by a KrF excimer laser with the repetition rate of 1 Hz produced a 27 mJ, 309 nm pulse using a large Ce:LLFcrystal which was grown by the Czochralski method, and the slope efficiency was approximately 17%.

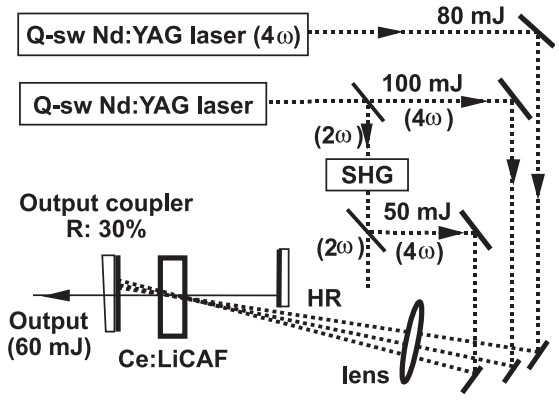

Figure 1. Experimental setup for high-power Ce:LiCAF laser pumped by the fourth harmonics of two Q-switched Nd:YAG lasers.

VIII-A-6 New Adjustment Technique for Time Coincidence of Femtosecond Laser Pulses Using Third Harmonic Generation in Air and its Application to Holograph Encoding System

KAWAMURA, Ken-ichi[1]; ITO, Naoko[2]; SARUKURA, Nobuhiko; HIRANO, Masahiro[1]; HOSONO, Hideo[1,3]
([1]ERATO; [2]Mater. Struct. Lab.; [3]Tokyo Inst. Tech.)

[*Rev. Sci. Instrum.* **73**, 1711 (2002)]

The third harmonic generation of light (266 nm) is enhanced, sensitively depending on the time delay between a pair of pulses split from a single 800 nm femtosecond laser pulse, when they are focused and collided in air. This finding offers a convenient and widely applicable technique to detect temporal and spatial overlapping of two femtosecond pulses. This technique has several advantages over the conventional sum frequency generation method using nonlinear optical crystals, since it obviates the need for expensive crystals, free from phase matching, and elimination of temporal walk off. By applying it to "a holographic encoding system using an interference femtosecond laser pulse," a periodic fringe spacing is minimized to ~ 430 nm by extending the colliding angle between two-pulse beams up to ~ 160 ºC.

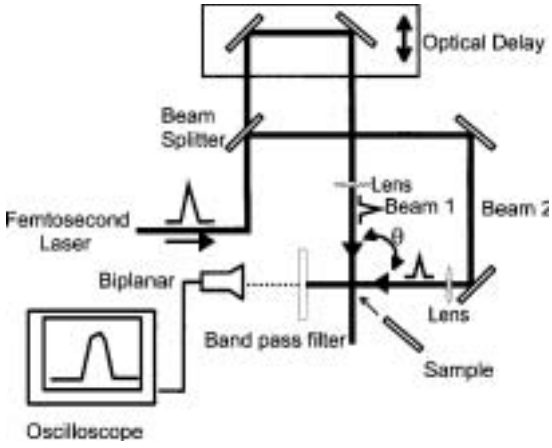

Figure 1. Experimental setup for holographic encoding system.

VIII-A-7 Hybrid Time-Resolved Spectroscopic System for Evaluating Laser Material Using a Table-Top-Sized, Low-Jitter, 3-MeV Picosecond Electron-Beam Source with a Photocathode

SUZUKI, Yuji; KOZEKI, Toshimasa; ONO, Shingo; MURAKAMI, Hidetoshi; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; NAKAJYO, Terunobu[1]; SAKAI, Fumio[1]; AOKI, Yasushi[1]
([1]Sumitomo Heavy Industries, Ltd.)

[*Appl. Phys. Lett.* **80**, 3280 (2002)]

Hybrid time-resolved spectroscopy of laser media comparing electron-beam excitation and optically excited cases is performed using a newly developed, table-top-sized, low-jitter, 3-MeV picosecond electron-beam source with a photocathode. The properties of an electron-beam-pumped Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) ultraviolet laser medium significant differ from those of an optically pumped medium.

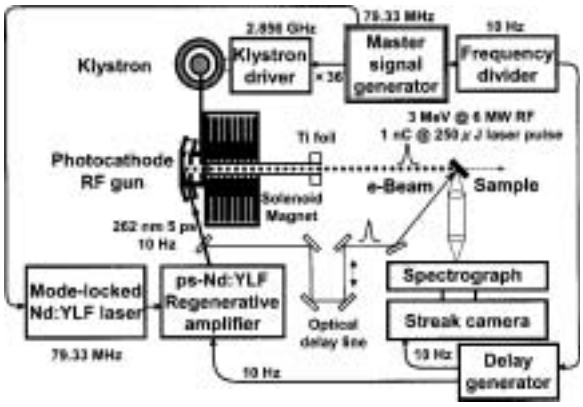

Figure 1. Experimental setup. The photocathode rf gun was irradiated with 262-nm optical pulses from a Nd:YLF regenerative system synchronously operated with a 2.856-GHz, 6-MW klystron to accelerate the extracted 1-nC photoelectron beam. The e-beam was irradiated onto the sample after being passed through titanium foil. A portion of the pumping optical pulse irradiated the same sample to compare the excitation scheme. A streak camera was equipped with a 30-cm spectrograph to measure the fluorescence spectrum and fluorescence lifetime.

VIII-A-8 Simultaneous Measurement of Thickness and Water Content of Thin Black Ink Films for the Printing Using THz Radiation

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([1]AISIN SEIKI Co., Ltd.; [2]Mitsubishi Heavy Industry Co., Ltd.)

[*Jpn. J. Appl. Phys., Part 2* **41**, L475 (2002)]

Using THz radiation, a simple, noncontact, simultaneous method is applied to measure thickness and water content of black ink films independently from frequency-dependent and frequency-independent absorption characteristics of black ink films.

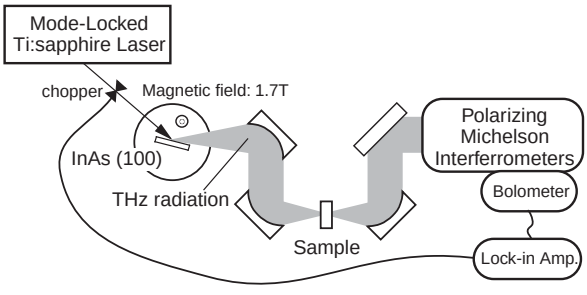

Figure 1. Experimental setup for simultaneously measuring thickness and water content of thin black ink films. The magnetic field direction is from the back to the surface.

VIII-A-9 Far-Infrared Absorption Measurements of Polypeptides and Cytochrome *c* by THz Radiation

YAMAMOTO, Kohji[1]; TOMINAGA, Keisuke[1]; SASAKAWA, Hiroaki[1]; TAMURA, Atsuo[1]; MURAKAMI, Hidetoshi; OHTAKE, Hideyuki; SARUKURA, Nobuhiko
([1]Kobe Univ.)

[*Bull. Chem. Soc. Jpn.* **75**, 1083 (2002)]

Pulsed terahertz (THz) radiation and black-body radiation are applied to measure far infrared (FIR) absorption spectra of polypeptides and cytochrome *c* in the wavenumber region from 7 cm^{-1} to 160 cm^{-1}. In the region from 7 cm^{-1} to 55 cm^{-1}, FIR absorption cross sections of polyglycine and poly-*L*-alanine in powder are greater than those of glycine and *L*-alanine in powder. On the other hand, FIR absorption spectra of cytochrome *c* in lyophilized powder show little dependence on protein structures. The structures of biopolymers are investigated by mid-IR absorption (polypeptides and cytochrome *c*) and by resonance Raman scattering (cytochrome *c*). FIR spectral features of biopolymers in the THz frequency region are qualitatively discussed in terms of density of states and homogeneous/inhomogeneous broadening.

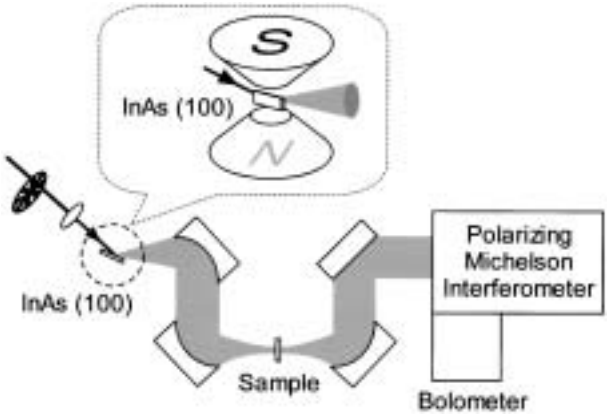

Figure 1. FIR absorption spectrometer using the THz radiation. Pulses at 800 nm with 70-fs width at 80-MHz repetition rate irradiate an InAs wafer, which is placed in a magnetic flux density of 1.7 T generated by an electromagnet. The (100)-InAs surface is parallel to a direction of the magnetic flux density. The THz radiation from the (100)-InAs is collimated by an off-axes parabolic mirror. The THz radiation is focused at a sample position and collimated again by an off-axes mirror. Intensity of the THz radiation is detected by a germanium bolometer which is cooled by helium liquid. Chopping frequency of the pump pulse is 200 Hz. A polarizing Michelson interferometer is vacuumsealed. All the FIR absorption system other than the polarizing Michelson interferometer is open to air. When using the black-body radiation for FIR absorption measurements, the generation part of the THz-radiation is replaced by a black-body.

VIII-A-10 0.43 J, 10 Hz Fourth Harmonic Generation of Nd:YAG Laser Using Large $Li_2B_4O_7$ Crystals

SUZUKI, Yuji; ONO, Shingo; MURAKAMI, Hidetoshi; KOZEKI, Toshimasa; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; MASADA, Genta[1]; SHIRAISHI, Hiroyuki[1]; SEKINE, Ichiro[1]
([1]Mitsubishi Materials Corp.)

[*Jpn. J. Appl. Phys., Part 2* **41**, L823 (2002)]

Using large-sized $Li_2B_4O_7$ crystals, 0.43 J, 266 nm pulses are obtained from a 10 Hz Nd:YAG laser with a total conversion efficiency of 30.5%. Moreover, 4 W operation for over 15 h is demonstrated.

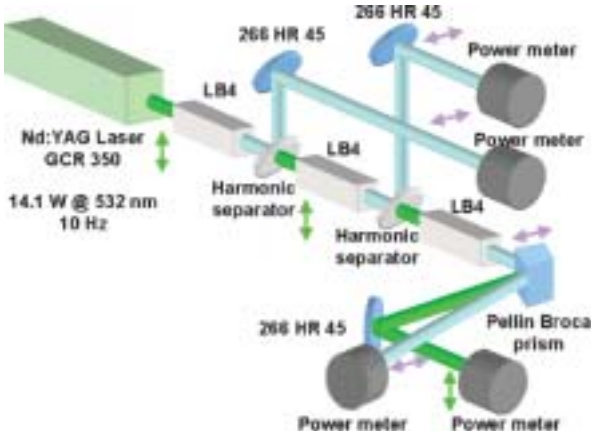

Figure 1. Experimental setup for three-cascade, fourth harmonic generation using LB_4 crystals.

VIII-A-11 Electron-Beam Excitation of a Ce^{3+}:$LiCaAlF_6$ Crystal for Future High-Peak-Power UV Lasers

KOZEKI, Toshimasa; SUZUKI, Yuji; SAKAI, Masahiro; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; SHIMAMURA, Kiyoshi[1]; FUKUDA, Tsuguo[1]; NAKAJYO, Terunobu[2]; SAKAI, Fumio[2]; AOKI, Yasushi[2]
([1]Tohoku Univ.; [2]Sumitomo Heavy Industries Ltd.)

[*Appl. Phys. B* **74**, S185 (2002)]

In this experiment, we performed ultrafast spectroscopy on an electron-beam-excited Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) crystal. The time-resolved fluorescence spectrum and lifetime with e-beam pumping differ significantly from those in the optically pumped case. These results suggest a new pumping scheme for an ultrashort pulse amplifier.

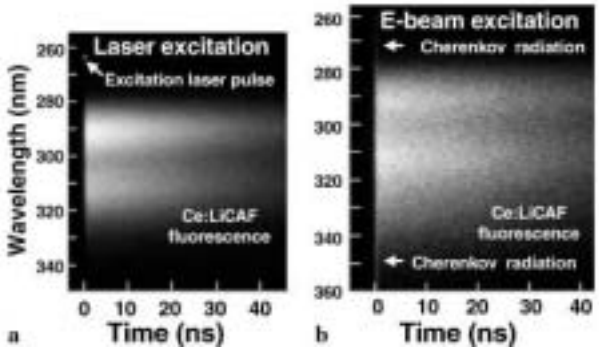

Figure 1. The streak camera images of the fluorescence from Ce:LiCAF, excited **a** by a 262-nm-pulse and **b** by an electron pulse. In the case of electron pulse excitation, a broadband, short-duration Cherenkov radiation is clearly seen.