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 Supercritical-Fluid Cell with Device of Variable Optical Path Length Giving Fringe-Free Terahertz Spectra

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[*Rev. Sci. Instrum.* **71**, 4061 (2000)]

An optical cell suitable for supercritical fluids was constructed for measurements of far infrared absorption spectra with terahertz radiation as a light source. It was designed to withstand temperature up to 400 K and pressure up to 15 MPa. The cell has two characteristic devices; one is diamond windows set in the Brewster angle to the incident far infrared light and the other is a variable optical path length from 30 μm to 20 mm under high pressure conditions. Using the cell, fringe-free spectra of CHF_3 ranging from low-density gaseous states to high-density supercritical ones were measured in a low-energy region of 10–100 cm^{-1}.

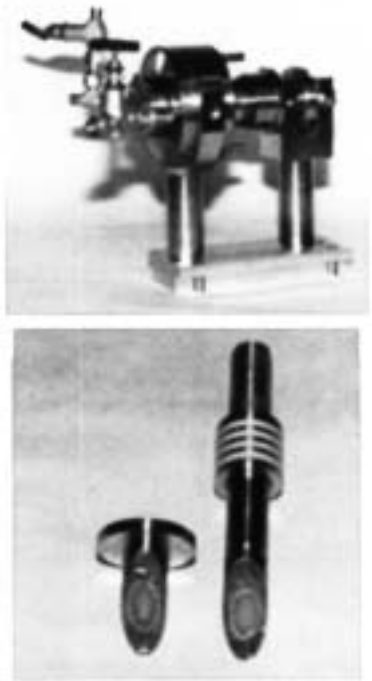

Figure 1. Photographs of an optical cell for far infrared absorption spectra measurements of supercritical fluids. The cell itself is shown in the upper part. The lower part represents flanges with diamond windows set in the Brewster angle, a cylinder, and a V packing.

VIII-A-2 Growth and Characterization of $KMgF_3$ Single Crystals by the Czochralski Technique under CF_4 Atmosphere

SHIMAMURA, Kiyoshi[1]; FUJITA, Tomoyo[1]; SATO, Hiroki[1]; BENSALAH, Amina[1]; SARUKURA, Nobuhiko; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.)

[*Jpn. J. Appl. Phys., Part 1* **39**, 6807 (2000)]

$KMgF_3$ (KMF) single crystals were grown by the Czochralski technique as a new candidate of vacuum-ultra-violet optical materials. The absorption edge of KMF single crystals was 115 nm. The distribution of birefringence in the radial direction was of the order of 10^{-7}. The thermal expansion coefficient of KMF single crystals along the <111> orientation was 1.98×10^{-5} K^{-1}. Together with its excellent mechanical properties, these characteristics show KMF to be superior to the current materials.

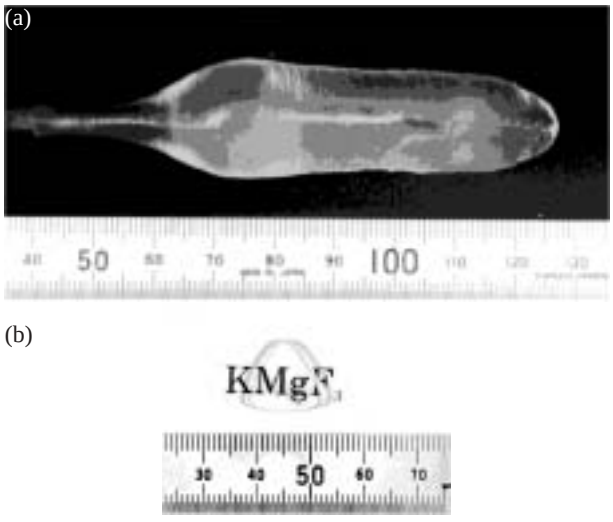

Figure 1. (a) As-grown $KMgF_3$ single crystal of 20 mm diameter pulled along the <111> orientation and (b) $KMgF_3$-wafer cut perpendicular to the growth axis with thickness of 2 mm.

VIII-A-3 Chirped-Pulse Amplification of Ultraviolet Femtosecond Pulses by Use of Ce^{3+}:$LiCaAlF_6$ as a Broadband, Solid-State Gain Medium

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([1]ERATO; [2]Tohoku Univ.; [3]Tokyo Inst. Tech.)

[*Opt. Lett.* **26**, 301 (2001)]

Chirped-pulse amplification in the ultraviolet region is demonstrated by use of a broadband Ce^{3+}:$LiCaAlF_6$ laser medium. A modified bow-tie-style four-pass amplifier pumped by 100-mJ, 266-nm pulses from a *Q*-switched Nd:YAG laser has a gain factor of 370 and delivers 6-mJ, 290-nm pulses. After dispersion compensation, the output pulses can be compressed to 115 fs.

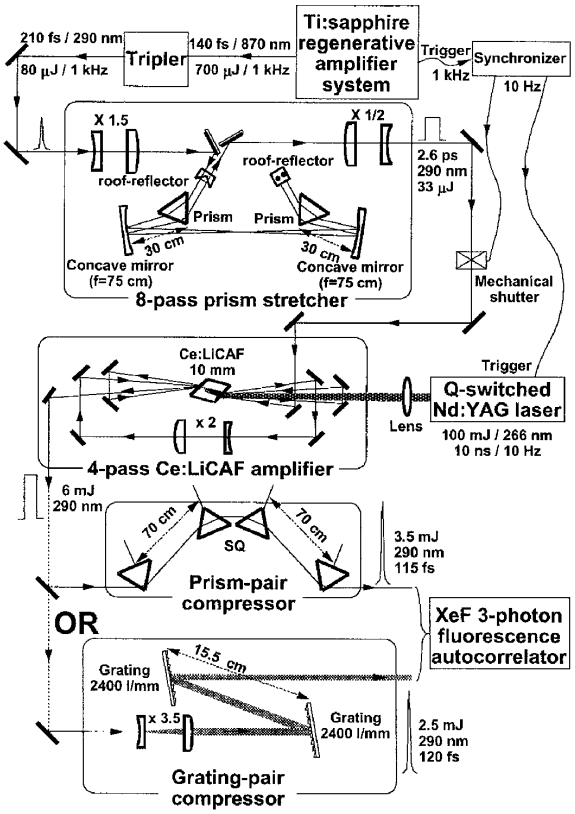

Figure 1. Experimental setup of the Ce:LiCAF CPA laser system. SQ, synthetic quartz.

VIII-A-4 Terahertz Radiation from a Shallow Incidence-Angle InAs Emitter in a Magnetic Field Irradiated with Femtosecond Laser Pulses

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([1]Sci. Univ. Tokyo)

[*Appl. Opt.* **40**, 1369 (2001)]

The optimized incidence angle and magnetic field direction geometry of an InAs terahertz radiation emitter irradiated with femtosecond laser pulses in a magnetic field is reported. The optimum geometric layout is the magnetic field direction parallel to the semiconductor surface and at an incidence angle that is slightly larger than the Brewster angle. Additionally, we also observed a center frequency shift of terahertz radiation spectrum by changing the incidence angle of the excitation laser.

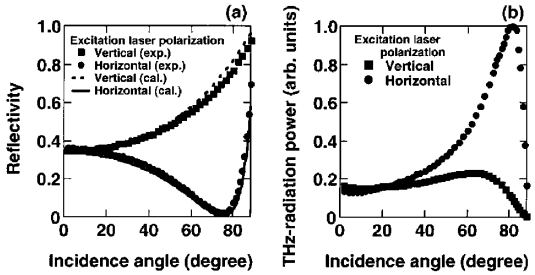

Figure 1. Angular dependence of (a) 800-nm excitation laser reflectivity and (b) THz radiation intensity from InAs. The solid and dotted curves represent the results of our theoretical calculations for $n = 3.729$ and $k = 0.448$.

VIII-A-5 Crystal Growth of Ce-Doped and Undoped $LiCaAlF_6$ by the Czochralski Technique under CF_4 Atmosphere

SHIMAMURA, Kiyoshi[1]; BALDOCHI, Sonia L.[1]; RANIERI, Izilda M.[1]; SATO, Hiroki[1]; FUJITA, Tomoyo[1]; MAZZOCCHI, Vera L.[2]; PARENTE, Carlos B. R.[2]; PAIVA-SANTOS, Carlos O.[3]; SANTILLI, Celso V.[3]; SARUKURA, Nobuhiko; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.; [2]Nucleares-IPEN/CNEN-SP; [3]UNESP)

[*J. Cryst. Growth* **223**, 382 (2001)]

Ce-doped and undoped $LiCaAlF_6$ (LiCAF) single crystals 50 mm in diameter were grown by the Czochralski technique. The formation of inclusions and cracks accompanying the crystal growth was investigated. The dependence of lattice parameters on the temperature was measured for LiCAF and $LiSrAlF_6$ single crystals. Linear thermal expansion coefficients for both these crystals were evaluated. Higher transmission properties for LiCAF single crystals were achieved in the UV and VUV wavelength regions.

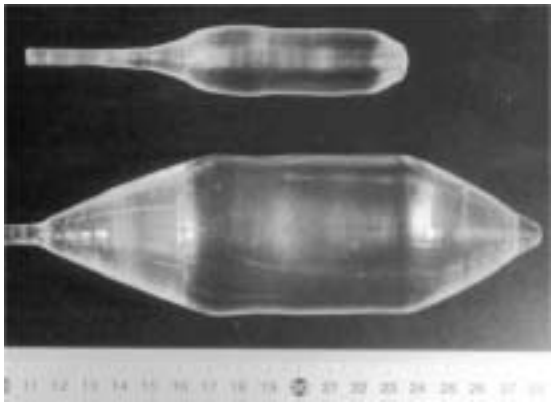

Figure 1. As-grown 2" diameter Ce-doped $LiCaAlF_6$ single crystal.

VIII-A-6 Ultraviolet Femtosecond Pulse Amplification with High Gain Using Solid-State, Broad-Band Gain Medium Ce^{3+}:$LiCaAlF_6$

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.)

[*Jpn. J. Appl. Phys., Part 1* **40**, 2308 (2001)]

Femtosecond pulse amplification with high gain in the ultraviolet region has been demonstrated using the solid-state, broad-band crystal Ce^{3+}:$LiCaAlF_6$. With the seed pulses coming from the third harmonic generation of a cw mode-locked Ti:sapphire laser, the confocal, four-pass amplifier pumped by 15-mJ, 266-nm, 10-ns pulses from a Q-switched Nd:YAG laser demonstrates 60-dB gain and delivers 54-μJ, 289-nm, 200-fs, 10-Hz pulses. There is almost no satellite pulse even without any special single-pulse selection.

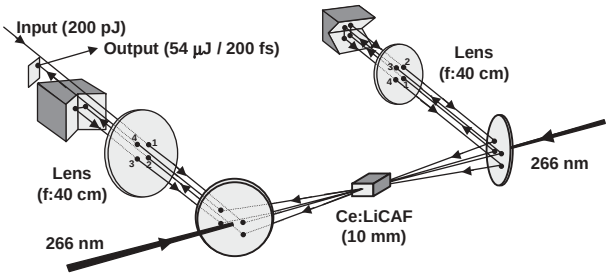

Figure 1. Experimental setup of the confocal, four-pass Ce:LiCAF amplifier. The input is the frequency-tripled output of a cw mode-locked femtosecond Ti:sapphire laser.

VIII-A-7 Terahertz Absorption Spectra of Supercritical CHF_3 to Investigate Local Structure Through Rotational and Hindered Rotational Motions

SAITOW, Ken-ichi[1]; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; NISHIKAWA, Keiko[1]
([1]Chiba Univ.)

[*Chem. Phys. Lett.* **341**, 86 (2001)]

Far infrared absorption spectra of neat supercritical fluoroform (CHF_3) are measured with terahertz (THz) radiation. The spectra covering from 10 to 100 cm^{-1} are obtained at reduced temperature $T/T_c = 1.02$ on densities varied by a factor of 200. As density increases, dominant component of spectra changes from rotational to hindered-rotational motion. However, the change is nonlinear to the density. Such specificity arises from difference between bulk and local densities, and the most enhanced local density is observed near the thermodynamic state of maximum density fluctuation. In the pure fluid system, the relationship between density fluctuation and local density enhancement is experimentally presented.

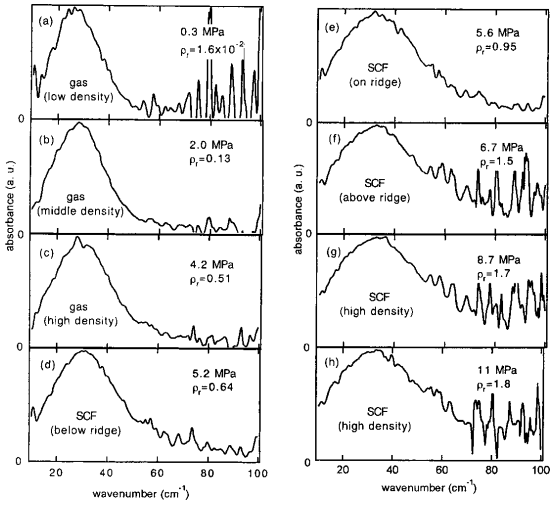

Figure 1. Far infrared absorption spectra of supercritical CHF_3 measured at reduced temperature by $T_r = T/T_c = 1.02$. The (a)–(c) are data below critical pressure (P_c) and (d)–(h) those of above P_c. The (d), (e), and (f) are ones below, on, and above the ridge, respectively. The (g) and (h) are ones at dense supercritical states above the ridge.

VIII-A-8 Observation of New Excitation Channel of Ceriumion through Highly Vacuumultraviolet Transparent LiCAF Host Crystal

KOZEKI, Toshimasa; SUZUKI, Yuji; SAKAI, Masahiro; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; LIU, Zhenlin; SHIMAMURA, Kiyoshi[1]; NAKANO, Kenji; FUKUDA, Tsuguo[1]
([1]Tohoku Univ.)

[*J. Cryst. Growth* **229**, 501 (2001)]

The transmission spectra of $LiCaAlF_6$ (LiCAF) and $LiSrAlF_6$ (LiSAF) are investigated in the ultraviolet (UV) and the vacuum ultraviolet (VUV) region. The transmission edge of LiCAF (112 nm) shows almost the same value as that of LiF. Taking into account difficulties of material processing and polishing due to the cleavage or the hydroscopic nature of LiF, LiCAF is regarded as a more suitable optical material in the UV and the VUV region. Moreover, the new excitation channel around 112 nm is discovered for Ce:LiCAF crystal. This excitation is originated not from absorption of Ceriumions but fromabsorption around the bandgap of the host crystal.

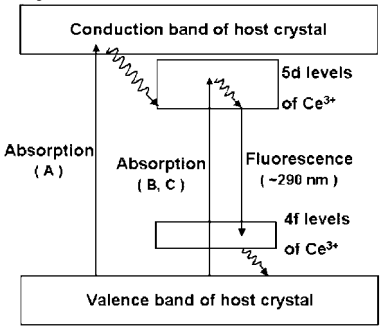

Figure 1. Schematic of energy levels and possible decay channel in Ce:LiCAF crystal.

VIII-A-9 THz-Radiation Emitter and Receiver System Based on a 2 T Permanent Magnet, 1040 nm Compact Fiber Laser and Pyroelectric Thermal Receiver

OHTAKE, Hideyuki; SUZUKI, Yuji; SARUKURA, Nobuhiko; ONO, Shingo[1]; TSUKAMOTO, Takeyo[1]; NAKANISHI, Akio[2]; NISHIZAWA, Seiji[3]; STOCK, Michelle L.[4]; YOSHIDA, Makoto[4]; ENDERT, Heirich[4]

([1]Sci. Univ. Tokyo; [2]Sumitomo Special Metal Co., Ltd.; [3]Shinshu Univ.; [4]Imra America, Inc.)

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

Thermal receiver detectable terahertz (THz) radiation is generated from InAs irradiated with a 1040 nm, 80 fs, 180 mW, 48-MHz-repetition-rate mode-locked fiber laser in a 2 T field by a compact permanent magnet. THz radiation is monitored by means of a deuterated triglycine sulfate (DTGS) pyroelectric thermal receiver. DTGS operates at room temperature and does not require time-gating adjustment or cryogen cooling with liquid helium. The THz-radiation emitter system, including the excitation laser head, is almost the same size as a conventional notebook computer.

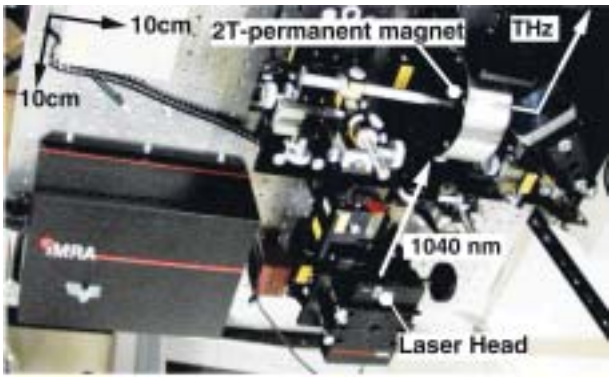

Figure 1. Photograph of THz-radiation emitter system. Laser beam is focused onto the sample with a 2 T permanent magnet.

VIII-A-10 Nanocluster Crystals of Lacunary Polyoxometalates as Structure-Design-Flexible, Inorganic Nonlinear Materials

MURAKAMI, Hidetoshi; KOZEKI, Toshimasa; SUZUKI, Yuji; ONO, Shingo[1]; OHTAKE, Hideyuki; SARUKURA, Nobuhiko; ISHIKAWA, Eri[2]; YAMASE, Toshihiro[2]

([1]Sci. Univ. Tokyo; [2]Tokyo Inst. Tech.)

[*Appl. Phys. Lett.* in press]

Lacunary polyoxometalates, large inorganic, structure-design-flexible, nanocluster crystals are found to have higher optical nonlinearity than KH_2PO_4 (KDP) by the powder second-harmonic-generation (SHG) method. Moreover, the capability of generating ultra-violet radiation down to around 300 nm is found. The basic criteria to design the high nonlinearity are also discovered by the reduction of the molecular symmetry.

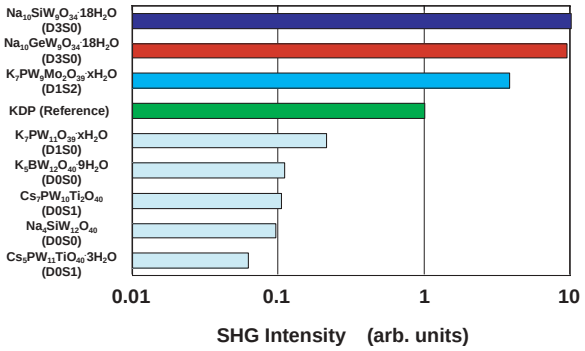

Figure 1. Powder second-harmonic-generation method results using a 1064-nm optical pulse from a Q-switched Nd:YAG laser as the fundamental radiation. Label S denotes the substitution number of metal atom.