RESEARCH ACTIVITIES

Center for Mesoscopic Sciences

In the past few decades, experimental and theoretical methods to analyze structures, dynamics, and properties of single-component (or single hierarchical) molecules and nanomaterials has been greatly progressed. Now we should also direct our attention to properties and functions of multi-hierarchical molecular systems. We develop innovative methods of measurements and analysis for molecular and materials systems to elucidate the processes that trigger the functions and reactions of the systems in the mesoscopic regime, that is the regime where micro and macroscopic properties influence each other.

Development of Advanced Nano-Optical Imaging and Application to Nanomaterials

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Keywords Nano Optics, Plasmons, Chirality

Studies of local optical properties of molecular assemblies and materials are key to understanding nanoscale physical and chemical phenomena, and for construction of nanoscale functional devices. Nano-optical methods, such as scanning near-field optical microscopy (SNOM), enable optical imaging with spatial resolution beyond the diffraction limit of light. Combination of nano-optical techniques with various advanced spectroscopic methods may provide a methodology to analyze directly nanoscale functionalities and dynamics. It may yield essential and basic knowledge to understand origins of characteristic features of the nanomaterials systems. We have constructed nano-optical (near-field and far-field) spectroscopic and microscopic measuring systems, for the studies on excited-state properties of nanomaterials, with the feasibilities of nonlinear and time-resolved measurements. The developed apparatuses enable nano-optical measurements of two-photon induced emission, femtosecond time-resolved signals, and chiro-optical properties (as typified by circular dichroism), in addition to conventional transmission, emission, and Raman-scattering. Based on these methods, we are investigating the characteristic spatial and temporal behavior of various metal-nanostructure systems and molecular assemblies. Typical examples are shown in Figure 1. We succeeded in visualizing wave functions of resonant plasmon modes in single noble metal nanoparticles, confined optical fields in noble metal nanoparticle assemblies. In recent few years, we have also succeeded in observing plasmon wave packet propagation dynamics with ultrafast time-resolved near-field imaging, local chiro-optical properties of chiral and achiral metal nanostructures, and so forth.

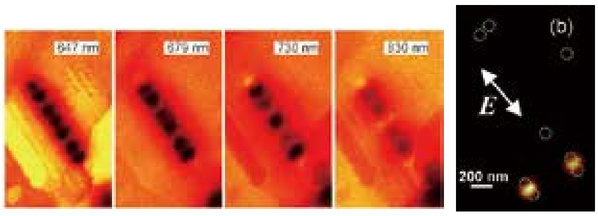

Figure 1. (Left four panels) Near-field transmission images of gold nanorod (20 nm^D × 510 nm^L). The wavelengths of observation were 647, 679, 730, and 830 nm from left to right. The spatial oscillating features were attributed to the square amplitudes of the resonant plasmonic wave functions. (Right) Near-field two-photon excitation image of dimers of spheric gold nanoparticles (diameter 100 nm) observed at 785 nm. The arrows indicates the incident light polarization. Dotted circles represent approximate positions of the particles.

Selected Publications

- H. Okamoto, T. Narushima, Y. Nishiyama and K. Imura, "Local Optical Responses of Plasmon Resonance Visualized by Near-Field Optical Imaging," *Phys. Chem. Chem. Phys.* **17**, 6192–6206 (2015).
- H. Okamoto and K. Imura, "Visualizing the Optical Field Structures in Metal Nanostructures," *J. Phys. Chem. Lett.* **4**, 2230–2241 (2013).
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1. Circular Dichroism Microscopy Free from Commingling Linear Dichroism via Discretely Modulated Circular Polarization[1)]

Circular dichroism (CD) is a representative method to detect chirality in materials spectroscopically. Microscopy based on CD signals provides unique and powerful technique for the studies of nanomaterials with chiro-optical and magneto-optical functions, biomolecular systems, *etc*. One of difficulty in CD microscopy is commingling of linear birefringence (LB) and linear dichroism (LD) signals, that arise from optical anisotropy of the samples. Because LB and LD signal levels are in general much larger than that of CD, slight imperfections of optical components and nonlinearity (in particular from polarization modulation devices) cause commingling LB and LD signals to CD.

In this work, we developed a far-field CD imaging microscope with a device to suppress the commingling of LB and LD signals. CD signals are, in principle, free from the commingling influence of LD and LB if the sample is illuminated with pure circularly polarized light, with no linear polarization contribution. Based on this idea, we here propose a novel circular polarization modulation method to suppress the contribution of linear polarization, which enables high-sensitivity CD detection (10^{-4} level in optical density unit or mdeg level in ellipticity) for microscopic imaging at a nearly diffraction limited spatial resolution (sub-μm level). The highly sensitive, diffraction-limited local CD detection will make direct analyses of chiral structures and spatial mappings of optical activity feasible for μm- to sub-μm-sized materials and may yield a number of applications as a unique optical imaging method.

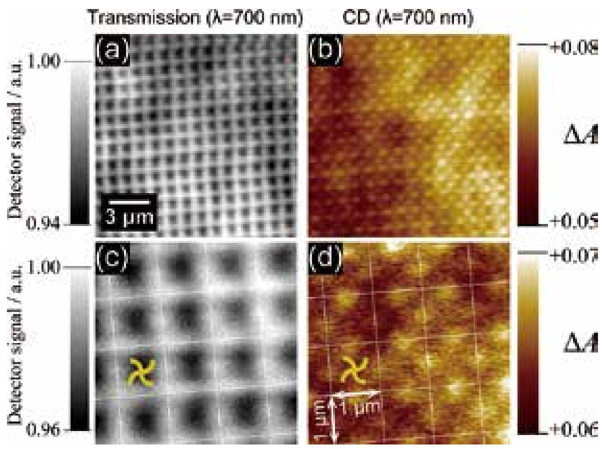

Figure 2. Transmission (a, c) and CD (b, d) images of the two-dimensional array of chiral (swirl-shaped) gold nanostructures. The wavelength of observation both for the transmission and CD images was 700 nm.[1)]

2. Near-Field Nonlinear CD Imaging of Single Gold Nanostructures[2)]

We demonstrated near-field nonlinear circular dichroism (CD) imaging of single rectangular (achiral) gold nanostructures using a two-photon excitation method. The gold rectangles were illuminated by pulses of circularly polarized light (CPL) to generate two-photon excitation images of the longitudinal plasmon modes. The observed images consisted of oval-shaped spatial features (corresponding to the antinodes of the plasmon modes) tilted from the long axis of the rectangles. The tilting direction depended on the handedness (left or right) of the CPL used for illumination, which led to the observation of a strong local dissymmetry of the two-photon excitation signals. The tilts of the oval features were not observed under linearly polarized pulse illumination with any polarization direction. The nonlinear CD images constructed from the differential two-photon excitation probability for left- and right-CPL pulses exhibited spatial features that were reasonably explained by the multipolar characters of the excited plasmon modes.

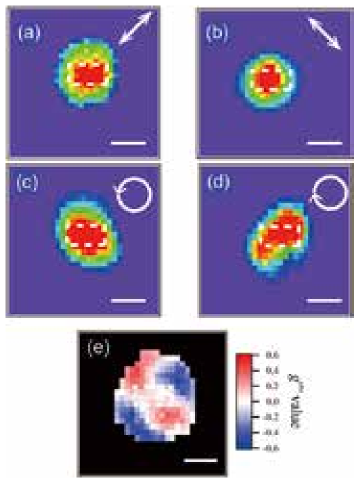

Figure 3. (a–d) Near-field two-photon excitation images of the rectangle at the excitation wavelength of 830 nm. Incident pulses are (a) linearly polarized, rotated by 45° from the long axis, (b) linearly polarized, rotated by −45° from the long axis, (c) left-circularly polarized, and (d) right-circularly polarized. (e) Near-field two-photon CD image evaluated from (c) and (d). The CD signals (g^{obs}) were not evaluated for the black areas outside the rectangular nanostructures because of low TPI-PL intensity. The dotted lines represent the approximate shape of the rectangle. Scale bars: 100 nm.[2)]

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2) Y. Nishiyama and H. Okamoto, *J. Phys. Chem. C* **120**, 28157–28162 (2016).

Awards

HASHIYADA, Shun; The Best Poster Presentation Award, NFO-14 (2016).
HASHIYADA, Shun; OSJ-OSA Joint Symposia Student Award (2016).

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Micro Solid-State Photonics

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Awards
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Keywords Solid-State Lasers, Nonlinear Optics, Micro Solid-State Photonics

"*Micro Solid-State Photonics*," based on the micro domain structure and boundary controlled materials, opens new horizon in the laser science. The engineered materials of micro and/or microchip solid-state, ceramic and single-crystal, lasers can provide excellent spatial mode quality and narrow linewidths with enough power. High-brightness nature of these lasers has allowed efficient wavelength extension by nonlinear frequency conversion, UV to THz wave generation. Moreover, the quasi phase matching (QPM) is an attractive technique for compensating phase velocity dispersion in frequency conversion. The future may herald new photonics.

Giant pulse > 10 MW was obtained in 1064nm microchip lasers using micro-domain controlled materials. The world first laser ignited gasoline engine vehicle, giant-pulse UV (355 nm, 266 nm) and efficient VUV (118 nm) pulse generations have been successfully demonstrated. Also, few cycle mid-IR pulses for atto-second pulses are demonstrated by LA-PPMgLN. We have developed new theoretical models for the micro-domain control of anisotropic laser ceramics. These functional micro-domain based highly brightness/brightness-temperature compact lasers and nonlinear optics, so to speak "Giant Micro-photonics," are promising. Moreover, the new generation of micro and/or microchip lasers by using orientation-controlled advanced ceramics can provide extreme high performances in photonics.

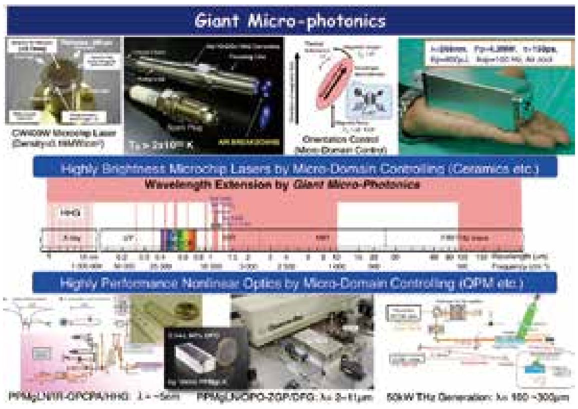

Figure 1. Giant micro-photonics.

Selected Publications

- H. Sakai, H. Kan and T. Taira, ">1 MW Peak Power Single-Mode High-Brightness Passively Q-Switched Nd^{3+}:YAG Microchip Laser," *Opt. Express* **16**, 19891–19899 (2008).
- M. Tsunekane, T. Inohara, A. Ando, N. Kido, K. Kanehara and T. Taira, "High Peak Power, Passively Q-Switched Microlaser for Ignition of Engines," *IEEE J. Quantum Electron.* **46**, 277–284 (2010).
- T. Taira, "Domain-Controlled Laser Ceramics toward Giant Micro-Photonics," *Opt. Mater. Express* **1**, 1040–1050 (2011).
- H. Ishizuki and T. Taira, "Half-Joule Output Optical-Parametric Oscillation by Using 10-mm-Thick Periodically Poled Mg-Doped Congruent $LiNbO_3$," *Opt. Express*, **20**, 20002–20010 (2012).
- R. Bhandari, N. Tsuji, T. Suzuki, M. Nishifuji and T. Taira, "Efficient Second to Ninth Harmonic Generation Using Megawatt Peak Power Microchip Laser," *Opt. Express* **21**, 28849–28855 (2013).

1. >MW Peak Power at 266 nm, Low Jitter kHz Repetition Rate from Intense Pumped Microlaser

Intense pulse pumped microlaser is proposed for high peak power and low timing jitter at high repetition rate. It is based on Intense and Fast Pulse Pump (IFPP) technique, in which fast pulse pumps up the upper-level population and then dumps it rapidly by Q-switching. That could come close to complete pumping efficiency to reduce thermal problems and contribute to suppress the timing jitter of passively Q-switched laser. In this work, linearly polarized 1064 nm beam from [100]-cut YAG/Nd^{3+}:YAG and [110]-cut Cr^{4+}:YAG passively Q-switched microlaser is directly guided into nonlinear crystals to obtain 532 nm and 266 nm output. By implementing IFPP concept, over 1 MW peak power, 215 ps pulse duration, 1 kHz pulses at 266 nm with reduced standard deviation timing jitter of 37 ns were obtained.

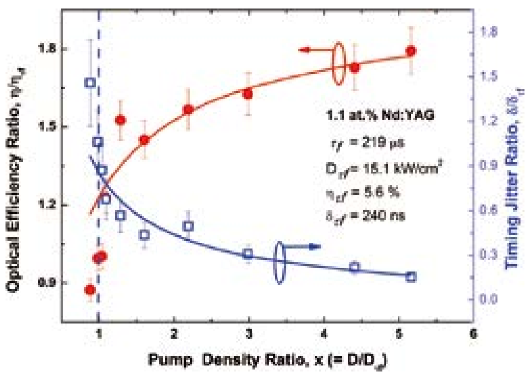

Figure 2. Optical efficiency ratio $\eta/\eta_{\tau f}$ and timing jitter ratio $\delta/\delta_{\tau f}$ as a function of pump power density ratio $D/D_{\tau f}$ at 1 kHz.

2. Sub-Nanosecond Laser Induced Air-Breakdown with Giant-Pulse Duration Tuned Nd:YAG Ceramic Micro-Laser by Cavity-Length Control

Laser induced breakdown (LIB) in air has been explained by different ways along pulse duration τ. Cascade ionization (CI) is considered as the mechanism for long pulse durations of over 1 ns. On the other hand, laser filamentation is alternative mechanism for fs pulses. However, for intermediate ps pulse, the mechanism is unclear due to a lack of knowledge about air-breakdown in sub-ns region and a contribution of multi-photon ionization. In addition, the knowledge about pulse duration dependence of breakdown energy in gas is practically useful for laser ignition. We first demonstrated a continuously giant-pulse duration tunable (0.5–9 ns) laser based on a short monolithic Nd:YAG/Cr:YAG ceramic by cavity-length control. LIB in laboratory air was investigated as a function of τ in sub-ns region using the developed laser. Air-breakdown threshold intensity I_{th} was measured using three different focusing conditions (Figure 3). We confirmed that (1) the measured I_{th} was almost constant at the longer τ than τ_{CI} named as limit-pulse-duration of CI, (2) I_{th} had ~τ^{-2} scaling for $\tau < \tau_{CI}$, (3) the increase of I_{th} is not connected to a specific intensity level, and (4) τ_{CI} was not constant and depended on focusing conditions.

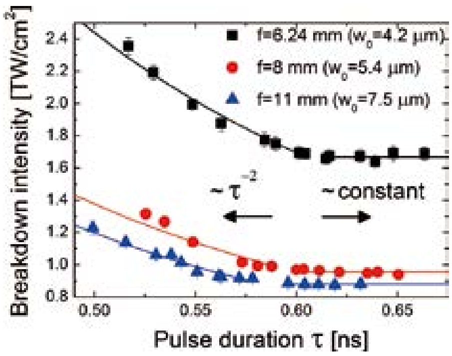

Figure 3. Measured breakdown threshold intensities as a function of pulse duration in laboratory air for three focusing conditions.

3. Damage Threshold Evaluation by Bulk-Shaped Nonlinear and Laser Materials

Laser-induced damage threshold (LIDT) of various optical materials, such as crystal quartz, YAG, and glass at sub-nanosecond pulse duration are evaluated. Bulk-shaped materials are used in the LIDT evaluation as shown in Figure 4, which can eliminate surface effects, such as adhesive dirt and polishing quality.

Evaluation were demonstrated using a laser source with 0.7-ns pulse-duration of 1.064 μm wavelength. LIDT of crystal quartz for piezoelectric and optical purpose were measured around 700 GW/cm^2 and 900 GW/cm^2, respectively. Also, LIDT of Nd:YAG and borosilicate glass were measured around 400 GW/cm^2 and 610 GW/cm^2, respectively.

Although conventional plate-shape measurement should be affected by its plate-surface condition, measurement using the bulk-shaped material can evaluate pure material characteristics.

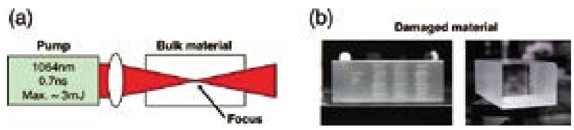

Figure 4. Laser-induced damage threshold evaluation using bulk-shaped material. (a) Set up of evaluation, (b) Example of damaged material.

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2) H. H. Lim and T. Taira, *Opt. Express* **25**, 6302–6310 (2017).
3) H. Ishizuki and T. Taira, *The 24th Congress of the International Commission for Optics (ICO-24)*, Tu1D-06 (2017).

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Ultrafast Laser Science

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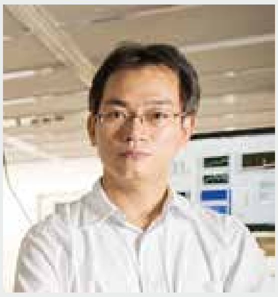

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Keywords Ultrafast Science, Laser Physics, Nonlinear Optics

Light is very common in daily life, on the other hand, light has many interesting physical properties, for example, constancy of velocity, wave-particle duality, *etc*. The study of light itself is still important in modern physics.

Light is electro-magnetic field, same as radio wave, however, the measurement of the waveform of light is not easy task even in the 21st century. The difficulty comes from the extremely fast oscillation of the light wave. The oscillation frequency of light wave is the order of hundred terahertz (THz = 10^{12} Hz), in other words, the oscillation period of light wave is the order of femtosecond (fs = 10^{-15} s).

In 2013, we have developed a new method for the measurement of light wave. It is called FROG-CEP, frequency-resolved optical gating capable of carrier-envelope determination. Our method does not need attosecond pulses, even self-referencing is possible. The electric field oscillations of infrared light with the period of several femtoseconds were clearly measured with the method as is shown in Figure 1.

Currently, amplitude modulation and phase modulation are common encoding techniques in optical communication. If we can encode information in the shape of the light wave itself, the communication speed becomes 3 orders of magnitude faster. We believe that our method, FROG-CEP, becomes very important to realize such communication technology.

Other than FROG-CEP, ultrabroadband mid-infrared continuum generation through filamentation, single-shot detection of ultrabroadband mid-infrared spectra, and development of 2 μm ultrafast lasers have been realized in our laboratory. We are developing such cutting edge technologies for ultrafast laser science.

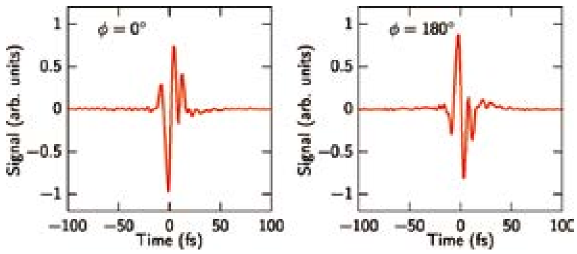

Figure 1. Infrared light waveforms measured with FROG-CEP. The phase difference between the two infrared pulses was clearly measured.

Selected Publications

- T. Fuji and Y. Nomura, "Generation of Phase-Stable Sub-Cycle Mid-Infrared Pulses from Filamentation in Nitrogen," *Appl. Sci.* **3**, 122–138 (2013).
- Y. Nomura, H. Shirai and T. Fuji, "Frequency-Resolved Optical Gating Capable of Carrier-Envelope Phase Determination," *Nat. Commun.* **4**, 2820 (11 pages) (2013).
- Y. Nomura M. Nishio, S. Kawato and T. Fuji, "Development of Ultrafast Laser Oscillators Based on Thulium-Doped ZBLAN Fibers," *IEEE J. Sel. Top. Quantum Electron.* **21**, 0900107 (7 pages) (2015).
- T. Fuji, Y. Nomura and H. Shirai, "Generation and Characterization of Phase-Stable Sub-Single-Cycle Pulses at 3000 cm^{-1}," *IEEE J. Sel. Top. Quantum Electron.* **21**, 8700612 (12 pages) (2015).
- T. Fuji, H. Shirai and Y. Nomura, "Ultrabroadband Mid-Infrared Spectroscopy with Four-Wave Difference Frequency Generation," *J. Opt.* **17**, 094004 (9 pages) (2015).
- H. Shirai, Y. Nomura and T. Fuji, "Self-Referenced Measurement of Light Waves," *Laser Photonics Rev.* **11**, 1600244 (6 pages) (2017).

1. Self-Referenced Waveform Measurement of Ultrashort Pulses

As is written in the previous page, it is still very difficult to directly sample an optical field transient on a time scale below the oscillation period. It has been naturally believed that the field oscillation can be detected only by using a reference pulse that has a shorter duration than the period of the oscillation.

In 2013, we proposed a new scheme of waveform characterization, frequency-resolved optical gating capable of carrier-envelope phase determination (FROG-CEP),[1)] which is based on a combination of frequency-resolved optical gating (FROG) with a carrier-envelope phase (CEP) sensitive time-domain nonlinear interferometer. The intensity and relative spectral phase of the target pulse are obtained from the FROG measurement, and the CEP is obtained from the nonlinear interferometer. Combining these data sets, we are able to retrieve a complete waveform of the target pulse. In this method, it is possible to measure a waveform with a reference pulse that has a longer duration than the period of the target pulse. The fact suggests that self-referenced waveform characterization is possible by using FROG-CEP.

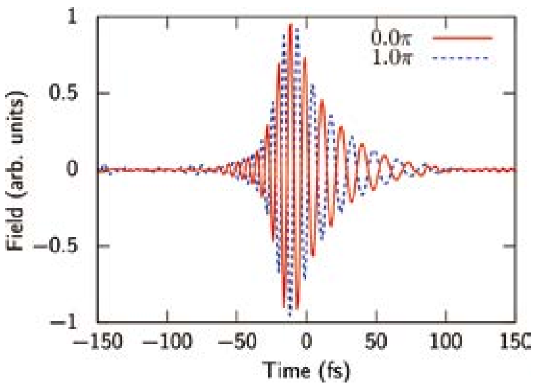

Figure 2. Retrieved electric-field in time domain. The solid line is the electric-field reconstructed with the method described in the text. The dotted line is the electric field where the CEP of the pulse is experimentally changed by π.

Here, we show the experimental demonstration of self-referenced FROG-CEP for few-cycle MIR pulses.[2)] We simultaneously measured a trace of second harmonic generation FROG (SHG-FROG) and interferogram between second harmonic (SH) and self-diffraction (SD) signals. The intensity and relative spectral phase of the target pulse are obtained from SHG-FROG, and the CEP is obtained from the interferogram. The retrieved waveform is shown in Figure 1.

2. Direct Amplification of 2 μm Femtosecond Pulses in a Tm:ZBLAN Fiber

Ultrafast lasers working in the 2 μm region have been attracting a lot of attention owing to a number of possible applications in various scientific and industrial fields. Thulium (Tm)-doped fiber lasers are one of the most promising candidates to generate ultrashort pulses in this wavelength region because of their broad emission spectra. Here we report the development of a fiber amplifier.

To develop high power, femtosecond light sources at the 2 μm region, the most straightforward method is the chirped-pulse amplification (CPA) technique. Another approach to build an ultrafast fiber laser amplifier is to make use of the nonlinearity within the fiber to broaden the spectrum rather than suppressing the nonlinearity. This approach was successful especially for developing ultrafast ytterbium-doped fiber laser amplifiers. However, the situation is quite different for Tm-doped fiber lasers because the fibers have anomalous dispersion around 2 μm region and thus the nonlinearity during amplification leads to wave breaking, which is usually considered detrimental.

We demonstrate direct generation of sub-50 fs pulses from a Tm-based fiber amplifier by utilizing nonlinearities within the amplifier fiber itself. Pulses with a duration of 48 fs are obtained at an average power of 2.5 W. The core-pumping scheme helped to obtain broad spectra extending into relatively short wavelength region around 1.7 μm. The setup uses no stretcher or compressor, resulting in an extremely simple system.[3)]

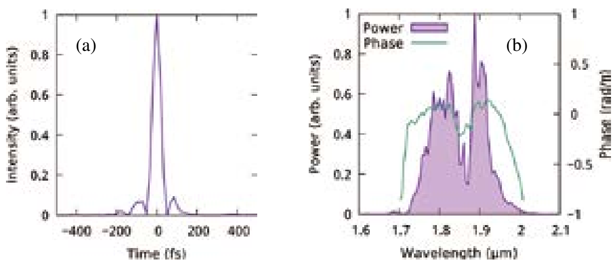

Figure 3. (a) Pulse shape retrieved from the FROG measurement. (b) Spectral intensity (blue solid curve) and phase (green dashed curve) profiles retrieved from the FROG measurement.

Figure 3 summarizes the results of FROG measurements of the amplified pulse. The pulse shape retrieved from the FROG measurement is shown in Figure 3(a). The duration of the pulse is as short as 48 fs, which is only 7% above the transform limit duration of 45 fs. The spectral intensity and phase profiles retrieved from the same FROG trace are shown in Figure 3(b). It can be seen that the phase is more or less flat for the main parts of the spectrum, whereas distorted in the region where the spectral intensity is relatively low.

References

1) Y. Nomura, H. Shirai and T. Fuji, *Nat. Commun.* **4**, 2820 (2013).
2) H. Shirai, Y. Nomura and T. Fuji, *Laser Photonics Rev.* **11**, 1600244 (2017).
3) Y. Nomura and T. Fuji, *Opt. Express* **25**, 13691 (2017).