RESEARCH ACTIVITIES

Center for Mesoscopic Sciences

In the past few decades, great progress in experimental and theoretical methods to analyze structures, dynamics, and properties of single-component (or single hierarchical) molecules and nanomaterials has been made. 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.

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. The information on nano-optical properties of the materials are also relevant to exploration of novel optical manipulation principles, which is another research topic of the research group.

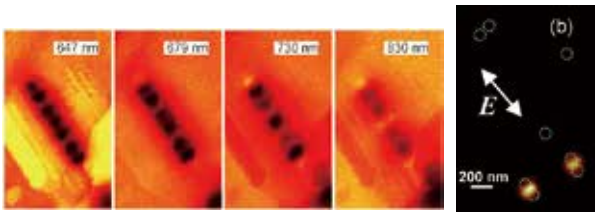

Figure 1. (Left four panels) Near-field transmission images of gold nanorod (20 nm^D × 510 nm^L). The wavelengths of observation were 647, 679, 730, and 830 nm from left to right. The spatial oscillating features were attributed to the square amplitudes of the resonant plasmonic wave functions. (Right) 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).
- H. Okamoto and K. Imura, "Near-Field Optical Imaging of Enhanced Electric Fields and Plasmon Waves in Metal Nanostructures," *Prog. Surf. Sci.* **84**, 199–229 (2009).

1. Imaging Chirality of Optical Fields near Achiral Metal Nanostructures Excited with Linearly Polarized Light[1)]

Chiral systems respond differently to left- and right-handed circularly polarized light macroscopically. As a consequence, only chiral materials show intrinsic macroscopic optical activity, and only chiral systems generate circularly polarized light from linearly polarized incident light. In the nanoscopic regime, in contrast to this general rule for macroscopic cases, it is theoretically expected that achiral (nonchiral) systems can locally generate circularly polarized fields. Here, we report experimental evidence for that situation in achiral systems consisting of gold nanostructures and linearly polarized incident light. The local circularly polarized fields were visualized by near-field polarimetry imaging. In this novel method, linearly polarized near-field radiation illuminates the sample, and the polarization state (ellipticity and rotational angle of polarization) of the far-field scattered light was measured at each position of excitation on the nanostructure, to construct the image. We found that highly circularly polarized fields were generated in the peripheries of the nanostructures, and the spatial features of the observed circularly polarized fields were qualitatively reproduced by a simple oscillating dipole model. The present results may provide a novel technique to produce controllable circularly polarized optical fields in nanospaces, the demonstration of which is now under way.

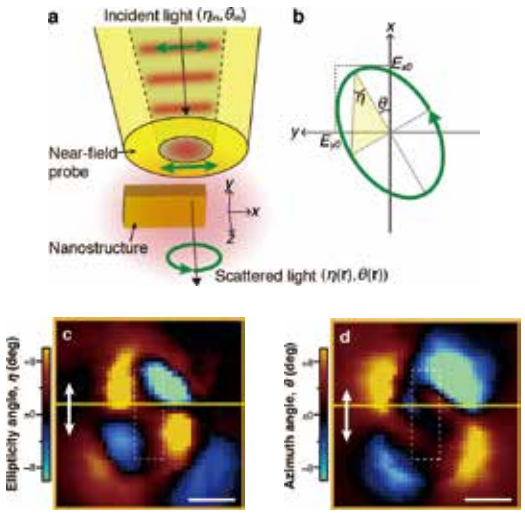

Figure 2. (a) Scheme of near-field polarimetry. (b) Definitions of ellipticity (η) and rotation (θ) angles. (c) Ellipticity and (d) optical rotation images (at 800 nm) of nano-rectangle. Scale bar: 100 nm.[1)]

2. Nanoscale Chiral Surface Relief of Azo-Polymers Induced by Near-Field Optical Angular Momentum Light[2)]

An optical vortex with orbital angular momentum (OAM) can be used to induce micrometer-scale chiral structures in various materials. Such chiral structures enable the generation of a near-field OAM light on a sub-wavelength scale, thereby leading to nanoscale mass-transport. We found formation of a nanoscale chiral surface relief (diameter ~400 nm) in azo-polymers due to near-field OAM light based on this principle. We analyzed near-field intensity and polarization state distributions in the periphery of the chiral structures using the near-field polarimetry imaging.

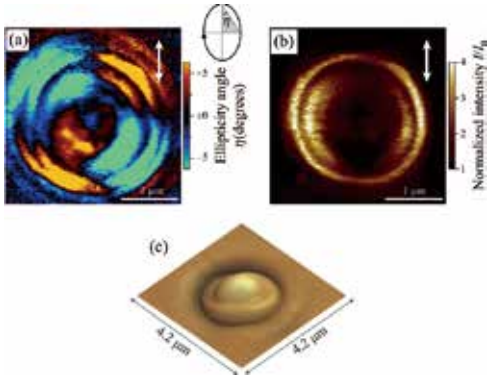

Figure 3. a) Near-field ellipticity, (b) near-field extinction, and (c) topographic images of left-handed spiral relief structure of azo-polymer fabricated with OAM light (532 nm).[2)]

3. Super-Resolution Optical Trapping: Theoretical Prediction of Nanoparticle Manipulation Using Nonlinear Optical Response[3)]

Optical manipulation of nanoparticles (NPs) with nanoscale precision is one of goals of nanomaterials science. A way to realize this is the usage of localized surface plasmon resonances. The electric fields near metallic structures are highly localized, which generate sufficient force to trap NPs, and the optical nonlinearity of NPs appears at the same time. In this study, we propose a scheme of spatially highly confined (beyond the diffraction limit) optical trap of the NP into a particular hotspot of the metallic nanostructure array. The scheme is based on the optical nonlinearity of NPs, and utilizes two kinds of structured light: Gaussian and Laguerre-Gaussian beams. The principle resembles that of stimulated emission depletion microscope, one of the major techniques of super-resolution microscope. The simulation results show the significant role of the optical nonlinearity in plasmon trappings. They are expected to open up new degrees of freedom to manipulate NPs.

References

1) S. Hashiyada, T. Narushima and H. Okamoto, *ACS Photonics* **5**, 1486–1492 (2018).
2) K. Masuda *et al.*, *Opt. Express* **26**, 22197–22207 (2018).
3) M. Hoshina, N. Yokoshi, H. Okamoto and H. Ishihara, *ACS Photonics* **5**, 318–323 (2018).

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Micro Solid-State Photonics

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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.

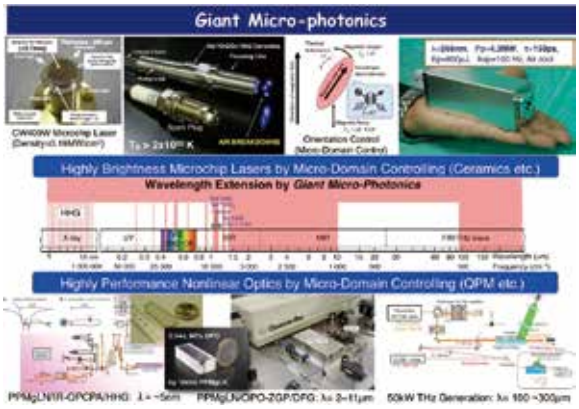

Figure 1. Giant micro-photonics.

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. Thermal Reduction through Distributed Face Cooling (DFC) in a High Power Giant-Pulse Tiny Laser

Sapphire/Nd^{3+}:YAG based DFC chip was obtained with thermal reduction as compared with those from conventional Nd^{3+}:YAG chip. The CW diode laser pumped round-trip cavity loss was 0.51% from a 9-disk DFC chip, which was close to theoretically calculated total Fresnel reflection loss of 0.2% from 8 Sapphire/Nd^{3+}:YAG interfaces. The depolarization ratio from 8-disk DFC chip was 40 times lower than that from YAG/Nd^{3+}:YAG chip. The DFC chip underwent no crack at pump power of 86 W while Nd^{3+}:YAG single chip suffered crystal crack under pump power around 54 W, as shown in Figure 2.

Over megawatt peak power from DFC tiny integrated laser was demonstrated at 1 kHz with 3-pulse burst modes. It is concluded that DFC structure could relieve thermal effects as expected.

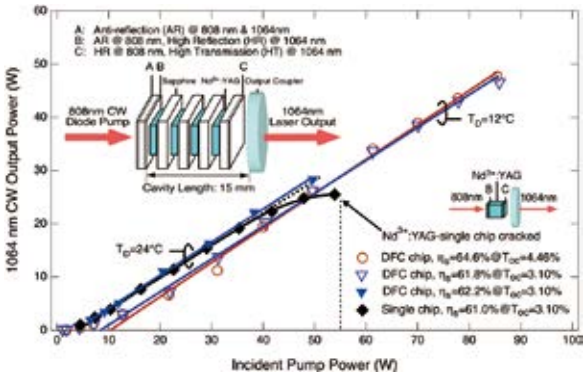

Figure 2. Output power from DFC chip under continuous wave laser pump. T_D is the temperature of diode laser.

2. Q-switching Laser Oscillation of Microdomain-Controlled Yb:FAP Laser Ceramics

The process control of microdomains with quantum mechanical calculations is expected to increase the optical power extracted per unit volume in gain media. Design of extensive variables allows us to evaluate the crystalline magnetic anisotropy in microdomains. Using this process control, we generate over 2 kW laser output from orientation-controlled microdomains made of Yb:Fluoroapatite (FAP).

In Figure 3, we compared the repetition rate and extraction energy density as the figure of merit for Giant-microphotonics, where our microdomain-controlled Yb:FAP laser ceramics showed excellent future possibility of power scaling.

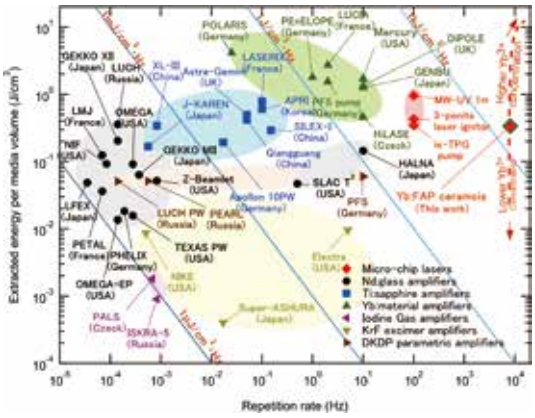

Figure 3. The figure of merit for Giant-microphotonics.

3. Crystal Quartz for High-Intensity, Sub-Nanosecond Wavelength Conversion

Crystal quartz for high-intensity wavelength conversion was evaluated. Pure durability of crystal quartz for sub-ns pulse region at 1.064 μm irradiation was measured as 602 GW/cm^2, which was 2-times higher than undope YAG crystal. QPM-structured quartz constructed by multi-plate stacking was evaluated by a sub-ns high-energy MCL-MOPA pumping. Maximum SH energy of 250 μJ could be obtained at E_p = 52 mJ with conversion efficiency of 0.48% as shown in Figure 4(a). Increasing characteristics of maximum E_{SH} on plate-stacking number N at E_p = 50~55 mJ is shown in Figure 4(b). Our experimental results well fitted the N^2-characteristics of the QPM characteristics.

As a result, availability of crystal quartz for high-intensity wavelength conversion could be demonstrated. QPM quartz is expected for both high-intensity operation and short-wavelength conversion.

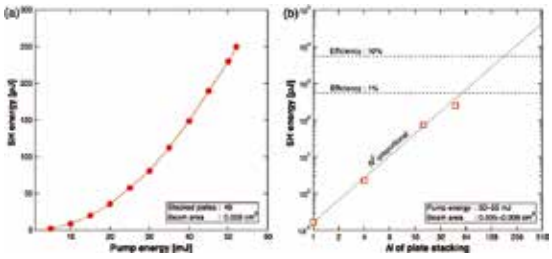

Figure 4. (a) SH energy on pump energy at N = 48, and (b) SH energy on stack number N.

References

1) L. H. Zheng, A. Kausas and T. Taira, *Opt. Mater. Express* **7**, 3214–3221 (2017).
2) Y. Sato, J. Akiyama and T. Taira, *Sci. Rep.* **7**, 10732 (2017).
3) H. Ishizuki, V. Yahia and T. Taira, *Opt. Mater. Express* **8**, 1259–1264 (2018).

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TAIRA, Takunori; The Commendation for Lazer Advancement of Taizan Prize (2017).

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Ultrafast Laser Science

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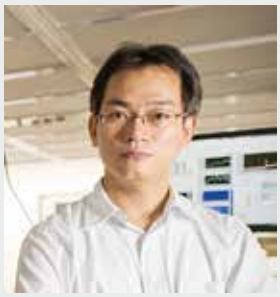

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Keywords Ultrafast Science, Laser Physics, Nonlinear Optics

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.

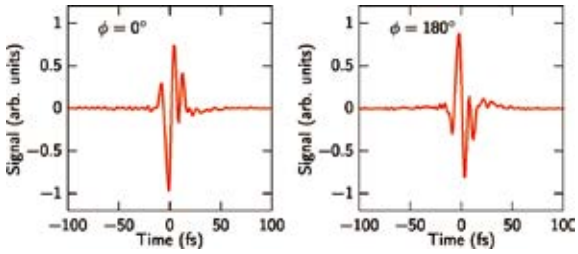

Figure 1. Infrared light waveforms measured with FROG-CEP. The phase difference between the two infrared pulses was clearly measured.

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.

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. High Harmonic Generation in Solids Driven by Sub-Cycle Mid-Infrared Pulses from Two-Color Filamentation

High-harmonic generation (HHG) is one of the most important nonlinear processes for the generation of attosecond pulses. In the last few years, HHG in solid materials is attracting a lot of attention in the fields of ultrafast science and solid-state physics. Since the atomic density is much higher in solids than in gases, solid-state HHG would be much more efficient than the HHG in atomic gases. The solid-state HHG would be a key technology to realize a compact solid-state attosecond pulse generator or petahertz electronics.

Naturally, experimental study with well-characterized single-cycle or sub-cycle pulses is one of the most straightforward approaches to investigate highly nonlinear process. Here, we report the demonstration of the HHG in a Si membrane driven by carrier-envelope phase (CEP) controlled sub-cycle mid-infrared (MIR) pulses generated through two-color filamentation.

The light source was based on a Ti:Sapphire multipass amplifier system. The generation scheme of the sub-cycle MIR pulses is the same as that published before.[1,2] In brief, the fundamental (800 nm, ω_1) and second-harmonic (400 nm, ω_2) pulses were gently focused into nitrogen, in which the sub-cycle MIR pulse (ω_0) was generated by using four-wave mixing ($\omega_1 + \omega_1 - \omega_2 \rightarrow \omega_0$) through filamentation. As is the case with the difference frequency generation, the CEP of the MIR pulse is passively stabilized.

Figure 2(a) shows a typical power spectrum and (absolute) spectral phases of the MIR pulses obtained with FROG-CEP measurements.[3] The spectrum covers the entire MIR region, corresponding to more than three octaves, and the spectral phase has some nonlinear term; namely, the pulse is slightly chirped. The pulse duration of the MIR pulse is estimated as ~8.5 fs at FWHM, corresponding to 0.64 optical cycles at 4 μm center wavelength. The waveforms of MIR pulses for the phases of -0.10π and -0.77π at 4 μm are shown in Figure 2(b). We can control the CEP of the MIR pulse very precisely by tilting the delay plate on a kinematic mount with a piezoelectric inertia actuator.

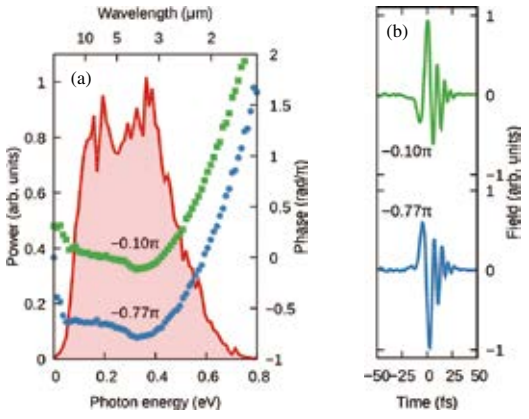

Figure 2. a) Power spectrum (shaded curve) and (absolute) spectral phases (closed circles and squares) of the MIR pulses obtained with the FROG-CEP technique. (b) Retrieved waveforms of the MIR pulses.

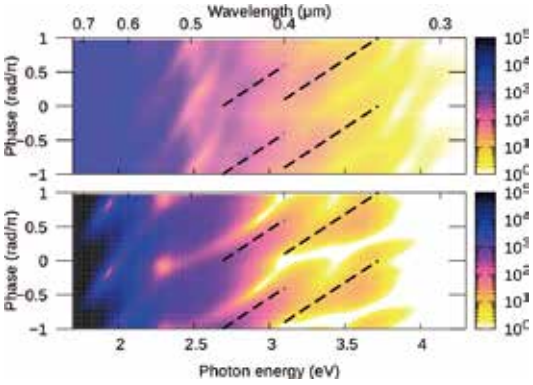

Figure 3. CEP dependence of the HH spectrum. (a) Experimental result. (b) Numerical simulation result obtained from optical Bloch equations.

Figure 3(a) shows the CEP dependence of the high-harmonic (HH) spectrum. We continuously recorded the HH spectra while scanning the CEP from $-\pi$ to π. The HH spectra reach <300 nm, the ultraviolet region. The spectrum shifts to the higher photon energy region by increasing the CEP (indicated by dotted lines), and the same spectrum appears every π phase shift.

To investigate the complex structure and CEP dependence of the HH spectrum, we numerically simulated the CEP dependence of the HH spectrum based on the optical Bloch equations generalized to the case of a two-band semiconductor. In this numerical simulation, we used the waveform of the sub-cycle MIR pulse measured with FROG-CEP as a driving field.

Figure 3(b) shows the simulation result of the CEP dependence of the HH spectrum. As is the case with the experimental results in Figure 3(a), the spectral shape shifts to a higher photon energy region by increasing the CEP of the MIR pulse, and the same spectrum appears at every π phase shift. In addition, discontinuous change of the CEP dependence of the HH spectrum at around 3.1 eV, which corresponds to the direct band gap energy, is also reproduced [see dashed lines in Figures 3(a) and 3(b)]. We believe that the CEP dependence change is due to the increase in the imaginary part of the refractive index at around the direct band gap. In this simulation, the main features of the experimental result are qualitatively reproduced.

References

1) T. Fuji and Y. Nomura, *Appl. Sci.* **3**, 122 (2013).
2) T. Fuji, Y. Nomura and H. Shirai, *IEEE J. Sel. Top. Quantum Electron.* **21**, 8700612 (2015).
3) Y. Nomura, H. Shirai and T. Fuji, *Nat. Commun.* **4**, 2820 (2013).

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