

Final Report

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Project Title:	Towards Laboratory-Based Ultrafast Bright EUV and X-ray Sources: High-Power Fiber Laser Frequency combs and Cavity Enhanced Ultrafast Optics
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Helmholtz Centre:	DESY
Participating University:	University of Hamburg
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1) Summary (max. 1 DIN A4 page)

Please describe the main results and the progress achieved in comparison to the state of the art at the time of writing the application and give an outlook on possible future work and applications.

Ultrafast optics studies the generation, manipulation, transmission, and characterization of ultrashort pulses and their innumerable applications in physics, chemistry, biology, etc. The utmost component in this field is undoubtedly lasers that are able to generate ultrashort pulses. The emergence of new lasers (e.g. thin disk lasers, fiber lasers, ceramic lasers, integrated waveguide lasers, etc.) or novel laser technology (Q-switching, mode-locking, chirped-pulse amplification, beam combining, etc.) always opens up new application fields. In return, the rapid advances in these new fields soon begin to drive new innovations in lasers to meet more challenging demands. It is this continuous back-and-forth interaction between ultrafast lasers and scientific applications that keeps ultrafast optics rapidly growing without any sign of saturation.

The research in my group has focused on three sub-fields of ultrafast optics—ultrafast fiber lasers, ultrafast nonlinear optics, and femtosecond laser spectroscopy/microscopy; they correspond to the above-mentioned three key elements (i.e., laser sources, nonlinear manipulation of ultrashort pulses, and applications) in ultrafast optics, respectively. During the past five years, my group employed fiber-optic nonlinearities to develop novel ultrafast fiber laser sources and improved two essential properties of fiber lasers: energy scalability and wavelength tunability.

We demonstrated a novel laser amplification technique—pre-chirp managed amplification (PCMA), in which seeding pulses are nonlinearly amplified such that the amplified spectrum was substantially broadened. By properly pre-chirping seeding pulses, the amplified pulse can be compressed to a duration much shorter than the transform-limited duration allowed by the seeding spectrum. Using an Yb-doped rod-type large-pitch fiber (LPF) as the power amplifier, Wei obtained 75 MHz, ~60 fs, linearly-polarized pulses with >100-W average power. The high-quality μJ pulses are obtained by two key enabling components: (1) the large mode area of large-pitch Yb-fiber leads to intrinsically low fiber nonlinearity resulting in energy scaling; (2) additionally, introducing a small amount of group-delay dispersion (typically in the range from -0.05 and 0.05 ps^2) prior to amplification results in high-quality output pulses after final compression. The resulting laser parameters are suitable for extreme nonlinear optics application such as frequency conversion in femtosecond enhancement cavities.

We also proposed and demonstrated a new method of producing wavelength widely tunable femtosecond pulses. The method employs fiber-optic self-phase modulation (SPM) to broaden an input optical spectrum, followed by optical bandpass filters to select the leftmost or rightmost spectral lobes. Both simulation and experimental results show that the filtered spectral lobes correspond to nearly transform-limited pulses with ~100 fs pulse duration. Using 20-mm commercially photonic crystal fiber, Wei experimentally demonstrated a femtosecond (70-120 fs) source with wavelength tunable from 825 nm to 1210 nm with >1 nJ pulse energy. By optimizing fiber dispersion, shortening fiber length, and using large-mode-area fibers, We further increased the pulse energy to 20 nJ in the range of 1030-1215 nm. We also applied this SPM-enabled spectral selection (SESS) technique to Er-fiber lasers and demonstrated generation of ~100 fs pulses with >10-nJ pulse energy; the wavelength is tunable from 1300 nm to 1700 nm. These key parameters represent a milestone in fiber laser technology aiming for ultrafast applications.

2) Work and Results Report

a) Starting point (max. 1 DIN A4 page)

Ultrafast fiber lasers have found many important scientific applications. Fiber lasers exhibit superior power scalability thanks to the fiber's large surface-to-volume ratio that results in excellent heat dissipation. Other advantages of fiber lasers include high electrical-to-optical conversion efficiency, large single-pass gain (~30 dB), excellent beam quality (close to diffraction-limited) due to waveguide confinement, as well as robustness and compactness. High power (>50 W), high repetition-rate (1-10s MHz) ultrafast Yb-fiber laser sources are demanded for driving high harmonic generation (HHG) to significantly increase the photon flux of the resulting extreme EUV pulses. Mode-locked fiber oscillators normally produce femtosecond pulses with nano-joule pulse energy, an energy level too weak to meet most of the scientific applications. Amplification of these weak femtosecond pulses beyond micro-joule level is normally accomplished using fiber amplifiers incorporating chirped-pulse amplification (CPA) technique. To avoid nonlinearities during amplification, the pulse is strongly stretched to a duration of typically 0.2-2 ns. Limited by gain narrowing and residual dispersion mismatch, the compressed pulse duration is typically limited to values above >200 fs. Further reduction of the pulse duration well below 100 fs—as required for efficient HHG—necessitates a subsequent external nonlinear pulse compression stage, which increases the system complexity and has limited throughput efficiency.

Another drawback of mode-locked oscillators is their limited wavelength coverage due to the available gain materials. However, applications such as multi-photon microscopy (MPM) demands large penetration depth to achieve deep-tissue imaging, which is largely determined by the proper ultrafast laser source that drives the MPM. An ideal source should provide energetic (>10 nJ) femtosecond (~100 fs) pulses with their center wavelength located in the optical penetration window (e.g., 1100–1300 nm) of the tissue. Such a source benefits MPM twofold: (1) higher pulse-energy improves the generation efficiency of nonlinear signals and (2) the optimized center wavelength of these femtosecond pulses mitigates tissue attenuation (i.e., scattering and absorption) and allows much deeper penetration.

b) Description of the results (max. 4 DIN A4 pages)

(1) Pre-chirp managed amplification

We recently proposed and demonstrated a new type of fiber amplification technique, named pre-chirp managed amplification (PCMA), in which the seeding pulse was nonlinearly amplified

such that the amplified spectrum was substantially broadened. By properly pre-chirping the seeding pulse, the amplified pulse can be compressed with a duration much shorter than the transform-limited duration allowed by the seeding spectrum. The resulting high quality μJ -level pulses are obtained by two key enabling components: (1) large-pitch fibers with large mode area featuring intrinsically low fiber nonlinearity and (2) a pre-chirper that introduces a small amount of group-delay dispersion (typically between -0.05 and 0.05 ps^2) prior to direct amplification resulting in high output pulse quality after final compression. Figure 1 shows the schematic setup and a photo of the experimental setup. At 220-W pump power, the Yb-fiber PCMA system outputs 75-MHz spectrally broadened pulses with $>130\text{-W}$ average power with excellent optical beam profile.

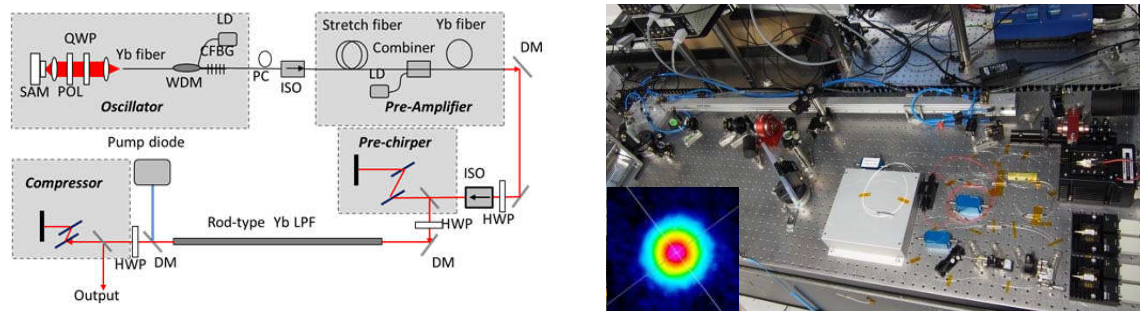


Fig. 1. (left) Schematic of the pre-chirp managed amplification system. SAM: saturable absorber mirror, QWP: quarter wave plate, POL: polarizer, LD: laser diode, CFBG: chirped fiber Bragg grating, PC: polarization controller, ISO: isolator, DM: dichroic mirror, LPF: large pitch. (right) Photo of the experimental setup. Inset: excellent beam profile at 130 W output power before compression.

Recent investigations of the EUV power scaling show that shorter driving wavelengths yield higher conversion efficiencies. To show applicability of the PCMA system for shorter wavelength generation, we use a 0.5 mm long BBO crystal to perform a frequency doubling experiment. Figure 2 shows the second-harmonic generation (SHG) output power and conversion efficiency for different IR power at 60 fs pulse duration (compression with a grating pair). For IR power levels below 30 W, the conversion efficiency increases dramatically. At 71 W pump power the conversion efficiency reaches 32% and we obtain 21 W of SHG. To prevent BBO crystal damaging, we limit the maximum IR power at 71 W; at this power level, the conversion efficiency is far from saturation. The pulse duration of the SHG is estimated to be around 50 fs.

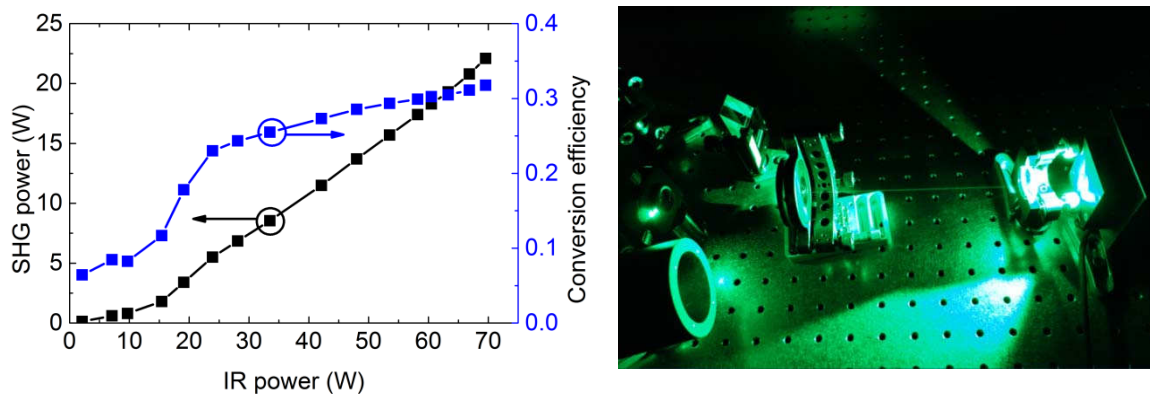


Fig. 2. (left) Second harmonic generation (SHG) output power and conversion efficiency for different IR power at 60 fs pulse duration. (right) photo of the SHG experimental setup

(2) Front-end source seeding cryogenic Yb:YLF amplifier for table-top X-ray source

Professor Franz Kärtner's group is developing a compact THz driven, attosecond X-ray source based on coherent inverse Compton scattering photons to a narrow forward cone by a free electron crystal. One of the key components composing this table-top X-ray source is a cryogenic Yb:YLF amplifier system that produces >100-mJ, sub-ps pulses as the driving source for THz generation and inverse Compton scattering. Cryogenic Yb:YLF amplifiers require for a seed laser operating at 1018-nm central wavelength and with a large stretching factor to permit efficient and damage-free energy extraction in subsequent 100 mJ-class amplifiers. My group developed the first high power fiber-based system specifically designed to optimally seed a high energy Yb:YLF amplifier chain. A fiber-format seeding source offers benefits including low-cost, reliability and available laser components.

As Fig. 3 shows, the laser system consists of three parts: the ultrafast oscillator, fiber amplifiers, and a rod-type fiber amplifier. Part 1 includes an Yb-fiber oscillator operating at 17.88-MHz repetition rate with the center wavelength at about 1016 nm. Part 2 includes two PM circulators connected to two PM chirped fiber Bragg gratings (CFBGs), which stretch the laser pulse to ~1 ns duration therefore alleviating nonlinear effects in subsequent amplifier stages. A pre-amplifier based on 2-m double cladding PM Yb-fiber is inserted between the two circulators to compensate the losses introduced by the circulators and CFBGs. Since the peak gain of this fiber amplifier is near 1030 nm, the center wavelength of the pulses shifts from 1016 nm to 1018 nm at this stage. Spliced directly to the port 3 of circulator 2, the second amplifier consists of a (2+1) beam combiner, a multi-mode pumping diode, and 1-m absorption-flattened Yb-doped double-clad fiber (FP fiber). This FP fiber provides higher gain at 1018 nm than 1030 nm, and thus efficiently amplifies the signal wavelength to an average power of 4 W.

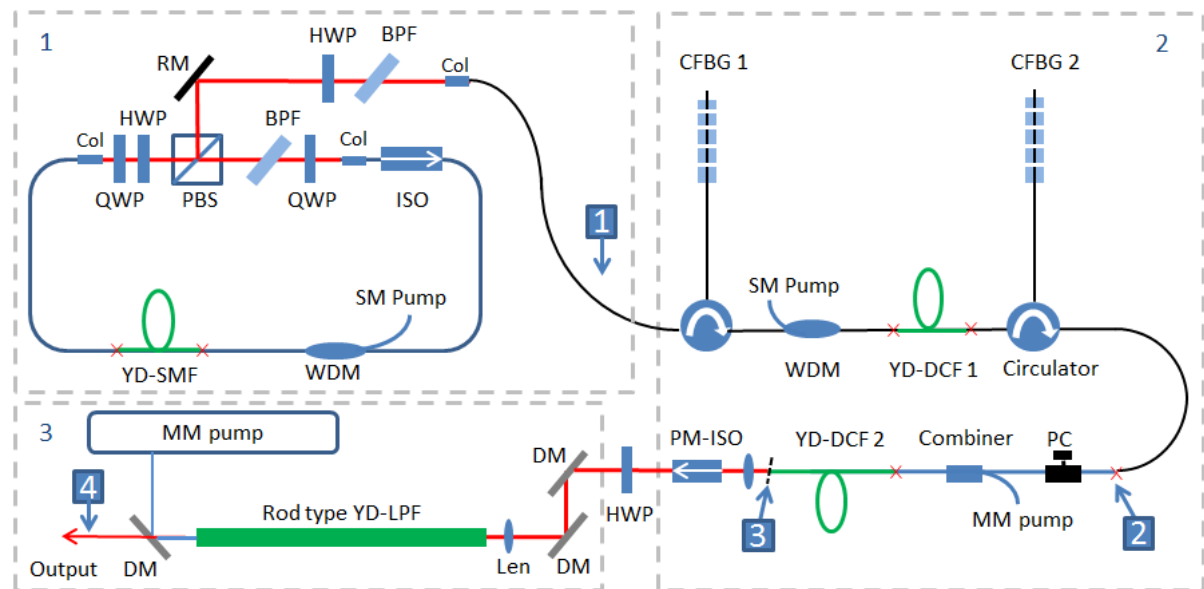


Fig. 3. Schematic setup of 1018 nm MOPA front-end for cryogenic Yb:YLF amplifiers. HWP: half wave plate, QWP: quarter wave plate, BPF: birefringent plate filter, PBS: polarization beam splitter, ISO: isolator, Col: collimator, SM: single mode, CFBG: chirped fiber Bragg grating, MM: multimode, PC: polarization controller, DM: dichroic mirror, LPF: large pitch fiber

Part 3 is the power amplifier consisting on a 1.2-m Yb-doped rod-type large-pitch fiber (LPF), pumped counter-propagating to the signal direction. The slope efficiency of the LPF amplifier is

limited to 49% (Fig. 4(a)) because the rod type fiber has its gain peaking at 1030 nm. Figure 4(b) shows the output spectrum versus average power of the amplified pulses. The signal spectrum remains unchanged, suggesting a linear amplification. Broadband ASE peaking at 1030 nm grows with the increased pump power, yet remains 26 dB below the signal spectral peak even at the highest average power of 87 W.

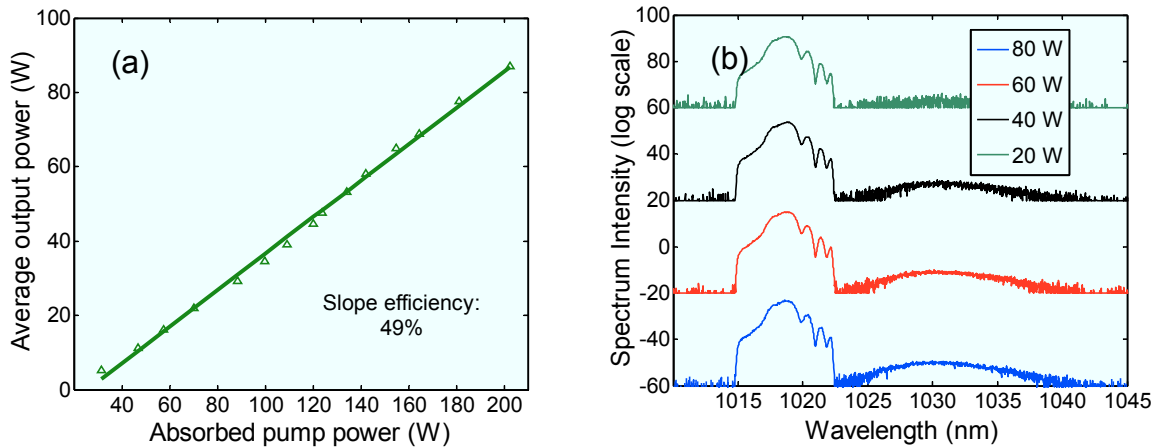


Fig. 4. (a) Measured output spectrum of rod-type LPF amplifier at 1018 nm for increasing pump powers; (b) measured characteristics output power versus absorbed pump power of rod-type LPF amplifier.

(3) Wavelength tunable femtosecond source based on SPM

My group demonstrated a new method of producing widely wavelength tunable femtosecond pulses. The method employs Kerr fiber-optic nonlinearities (dominated by SPM) to broaden an input optical spectrum, followed by optical bandpass filters to select the leftmost or rightmost spectral lobes. Without external compression, the filtered spectral lobes correspond to ~100-fs (nearly transform-limited) pulses with >1 nJ pulse energy.

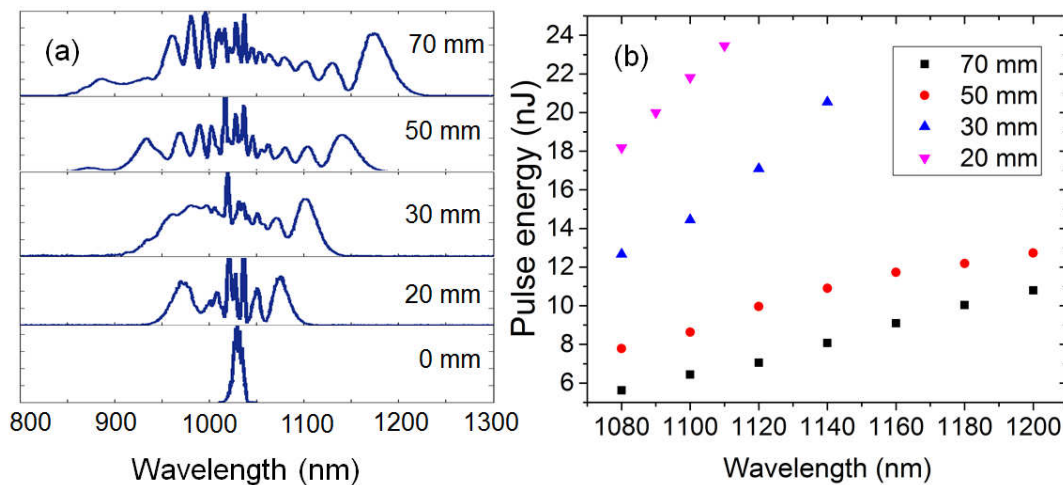


Fig. 5. (a) SPM-enabled spectral broadening in fiber LMA 8 of different length: 20 mm, 30 mm, 50 mm, and 70 mm. The coupled average power is 3 W for all fibers. (b) Pulse energy of the filtered rightmost spectral lobes at different central wavelength for different fiber length

To achieve more pulse energy, we employ large-mode-area (LMA) fiber (LMA-8) for spectral broadening. We coupled 3-W, 190-fs pulses (55-MHz repetition rate) into fiber LMA-8 of four different lengths: 20 mm, 30 mm, 50 mm, and 70 mm. As Fig. 5(a) shows, the rightmost spectral lobe red-shifts with the increased fiber length and peaks at 1080 nm (1170 nm) for 20-mm (70-mm) LMA-8. We then use suitable optical bandpass filters to select these rightmost spectral lobes, resulting nearly transform-limited pulses with ~ 100 -fs duration. We varied the coupled power into these fibers to generate the filtered spectra at different wavelengths. Figure 3(b) summarizes the pulse energies corresponding to the filtered spectra peaking at different wavelengths for the different fiber lengths. As expected, the filtered spectra from shorter fibers exhibit higher pulse energies for the same wavelength shift. For example, the filtered spectrum at 1100 nm has pulse energy of 6.5 nJ, 8.5 nJ, 14.5 nJ, and 22 nJ for fiber length of 70 mm, 50 mm, 30 mm, and 20 mm, respectively. This SPM-enabled source can provide wavelength tunable (in 1.15-1.22 μm) femtosecond pulses with 100 times more pulse energy compared with Raman soliton based tunable source. Numerical simulation shows that for 11-W average power (200-nJ pulse energy) coupled into 20-mm LMA-8, the rightmost spectral lobe is shifted to 1200 nm and the filtered spectrum exhibits >40 nJ pulse energy with >2.2 -W average output power.

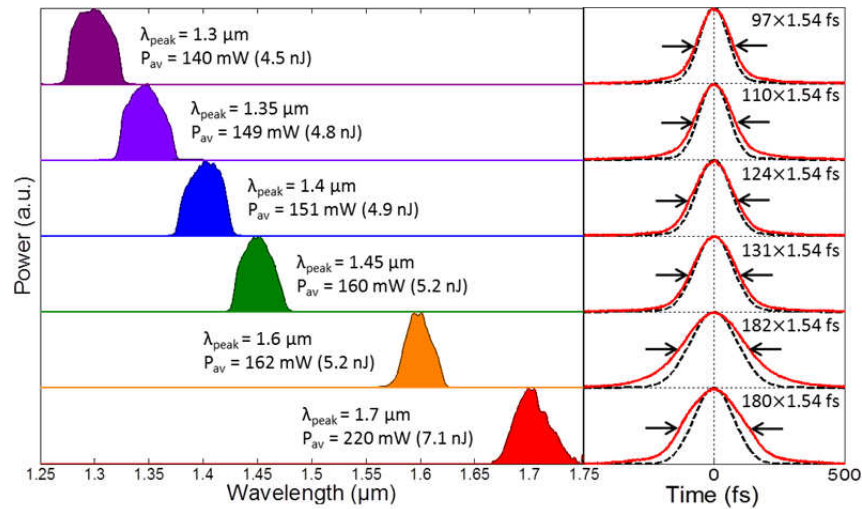


Fig. 6. (Left column) Filtered optical spectra from 2-cm highly nonlinear fiber; their peak wavelength, average power, and pulse energy are labeled in the figure. (Right column) Measured autocorrelation traces (red solid curves) and autocorrelation traces calculated from the transform-limited pulses allowed by the filtered spectra (black dotted curves).

We further applied our method to Er-fiber laser technology and achieved wavelength-tunable femtosecond sources in 1300-1700 nm (Fig. 6). By applying this SPM-enabled spectral broadening method to both an Yb-fiber laser system and an Er-fiber laser system, we can generate ~ 100 -fs pulses tunable in 825-1700 nm, exceeding one octave wavelength coverage. With further optimization, pulse energy may be scaled up to ~ 100 -nJ level. Since all the free-space components in our source can be replaced by fiber or fiber-pigtailed devices, sub-100-nJ-level femtosecond sources can be constructed in an all-fiber format. We believe that such SPM-enabled femtosecond fiber-laser sources provide an energetic, compact, and robust solution for many applications.

c) Outlook on future work, sustainability (max. 2 DIN A4 pages)

The advanced laser technologies pursued by my group have set new impulses for the collaboration research with other groups. Besides further collaboration with my DESY host Prof. Franz Kärtner, my group works together with the research group led by Professor Christian Betzel from University of Hamburg to apply our SPM-enabled tunable ultrafast sources to drive a laser-scanning microscope to identify and score protein nano-crystals. This technology allows controlled production of nano-crystals and therefore enables the study of previously inaccessible protein structures using X-ray radiation. For example, interest in the structure and function of integral membrane proteins is driven largely by their role in human disease. High-resolution protein structures can provide information on ligand binding locations and protein function. The most common strategy for obtaining such structures is currently X-ray diffraction, often performed at synchrotron sources. Complications commonly plaguing soluble protein crystal detection in high-throughput systems are exacerbated in studies of membrane proteins for several reasons. Consequently, crystallizing membrane proteins is notoriously difficult, and often requires screening a large number of trials. Second harmonic generation (SHG) microscopy is a promising candidate for the preliminary observation of chiral crystals. Via the SHG imaging contrast, the presence of protein crystals can be visualized with good sensitivity and background suppression: coherent SHG signals only arise from non-centrosymmetric structures, and they can be enhanced with certain classes of ordered systems. To develop such harmonic-generation microscopy method for protein nanocrystal imaging and sorting, we submitted a joint proposal with Prof. Betzel's group to Federal Ministry of Education and Research, Germany in 2016 and received 75,000 Euros to develop a fiber-laser driven harmonic-generation microscope. Such a microscope will be installed at European XFEL in the near future.

The mid-IR wavelength range between 6 and 20 μm , in particular, has been known as the fingerprint region. Spectroscopic information of these vibrational bands reveals the molecular structure and, in turn, identifies the ingredients of the sample under test. In this scenario, a high power, low noise, tunable mid-IR femtosecond source is highly desired from the viewpoint of rapid high-resolution sensing and spectroscopy.

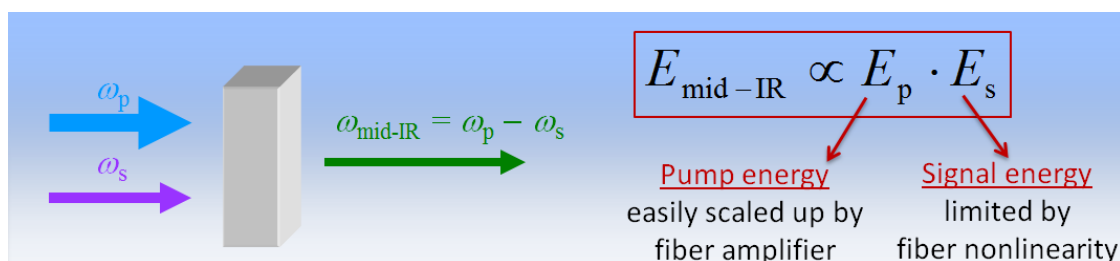


Fig. 7. Mid-IR generation based on difference frequency generation between a pump beam and a signal beam. The mid-IR pulse energy is proportional to the pulse energy product between the pump and signal.

Figure 7 illustrates the difference-frequency generation (DFG) process inside an optical crystal, which involves a pump beam (i.e., 1.03 μm pulse produced by our Yb-fiber laser system), a signal beam (i.e., the wavelength tunable pulse in 1.15-1.22 μm), and the generated mid-IR beam. In the conventional design, the pump pulse and the signal pulse differs in energy (or power) by orders of magnitude. For example, the pump pulse in our experiment is derived from the Yb-fiber laser system with a pulse energy of >400 nJ while the signal pulse has an energy

of ~100 pJ limited by Raman soliton pulse energy. Our numerical simulation indicates that the mid-IR pulse energy is proportional to the energy product between the pump pulse and the signal pulse, implying energy doubling either the pump pulse or the signal pulse will result in the same mid-IR pulse energy. The simulation shows that DFG between 90-nJ pump pulse and 22-nJ signal pulse can generate 1-nJ mid-IR pulse at 10 μm , representing >10 times energy increase compared with current demonstrated DFG-based mid-IR source. This immediately suggests that energy scaling a DFG-based mid-IR source by increasing the signal pulse energy rather than increasing the pump pulse energy constitutes a more efficient avenue. In fact, the pump pulse energy has to be kept below a certain value to prevent crystal damage. Therefore increasing the signal pulse energy becomes a powerful and practical solution to achieve high-power mid-IR pulses. The tunable ultrafast sources—especially the high-power mid-IR femtosecond source—play an important role in ultrafast spectroscopy of condensed matters. In the investigation of topological insulators, the known bandgap is below 100 meV, which falls within the mid-IR (5-10 μm) range. However current available optical parametric oscillators at this wavelength range have very limited output power.

Our DFG-based mid-IR source holds the promise to produce widely tunable femtosecond pulses with the average power at least one order of magnitude higher than current mid-IR laser technology. Such a powerful mid-IR source forms the core for the collaboration with the research group led by Prof. Jimin Zhao from the Institute of Physics, Chinese Academy of Science (CAS). With a powerful mid-IR laser source, we will investigate the resonant ultrafast dynamics of the topological insulators (e.g. the most typical 3D topological insulator Bi_2Se_3). In the previous ultrafast optical investigation, excitation was achieved with 800 nm photons, and the electrons will pass a long way in the momentum space (through electron-phonon interaction) to reach the Dirac cone right above the bandgap. Thus the direct relaxation and carrier lifetime is unclear. Herein we use the developed mid-IR laser pulses to directly excite and probe electrons that have been promoted to the Dirac cone conduction band. Only in such a way does the carrier relaxation lifetime directly reflect the topological order such that we can detect the Z_2 property of the surface state. If conditions permit we will apply magnetic field to test its inversion symmetry as well. Furthermore we will develop magnetic ion doped Bi_2Se_3 sample to verify the anomalous effects that break the time-reversal symmetry; such an optical investigation is absent due to the unavailability of a high-power ultrafast mid-IR laser source. This study may be extended to topological insulators considering interaction between electrons in different materials, *i.e.* in the non-Fermi-gas picture. Current investigations of topological insulators, for example transport measurements, focus on states near the Fermi surface. Mid-IR laser spectroscopy allows us to study topological properties at excited state—a field rarely explored.

We have submitted a proposal to CAS to further advance the mid-IR laser technology aiming for understanding the fundamental mechanism of light-matter interaction and time-resolved quantum phase transition in mid-IR wavelength regime. We plan to submit a joint DFG-NSFC proposal in 2018 to further support our next-level collaboration.

d) Potential for application/exploitation (max. 2 DIN A4 pages)

The wavelength tunable ultrafast sources developed in this program promise many important potential applications. We plan to apply them driving multi-photon microscope (MPM) to perform biomedical imaging. The success of MPM is largely driven by the rapid advance of femtosecond laser sources in the near infrared wavelength range. To date, no single laser can provide >1 nJ femtosecond pulses tunable in the entire 800-1300 nm wavelength range. Conventional solution relies on multiple laser systems each covering a portion of 800-1300 nm and the stitched spectrum spans the whole wavelength range. A typical combination is a

Ti:sapphire laser tunable from 700 to 1040 nm plus a solid-state optical parametric oscillator (OPO) that covers the wavelength tuning range of 1040-1300 nm. However, high complexity (e.g., water cooling required), high cost, and large size of such a solid-state laser solution limits the use of MPM to specialized laboratories. All these drawbacks associated with solid-state lasers can be eliminated in our SPM-enabled ultrafast sources that employ fiber-optic technologies. Figure 8 compares the solid-state laser solution and our fiber laser solution. Together with our collaborators, we are pursuing two applications: (1) biomedical imaging of cancer cells with submicron optical resolution and (2) imaging and sorting protein nanocrystals.

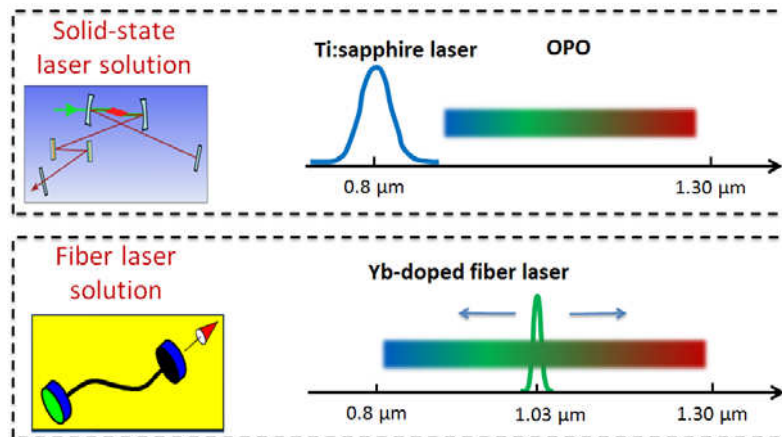


Fig. 8. Ultrafast laser driving source for MPM: solid-state laser solution versus fiber laser solution.

Biomedical imaging of cancer cells with submicron optical resolution: To show that such a widely tunable source is suitable for driving MPM imaging using different contrast modalities, we first tune the source to output <100-fs pulses at ~920 nm to match the two-photon excitation wavelength of enhanced green fluorescence protein (EGFP)—the most commonly used labeling protein. Using this source to drive our MPM and collecting the fluorescence signal, we obtained the two-photon excitation fluorescence image of breast cancer cells labeled by EGFP; see Fig. 9(a).

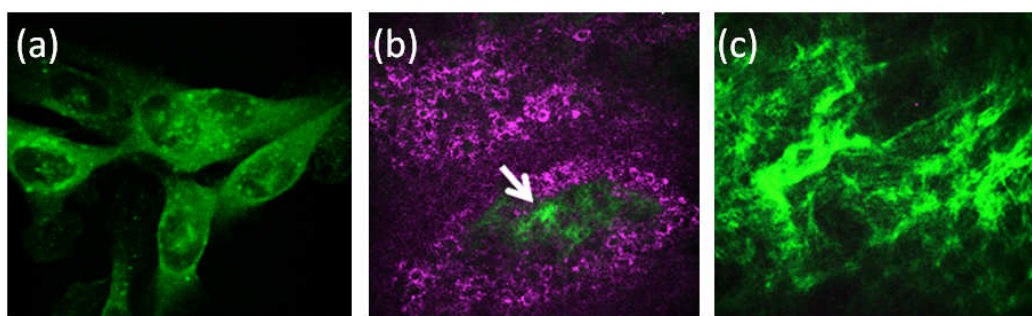


Fig. 9. (a) two-photon excitation fluorescence imaging of breast cancer cells labeled by EGFP. (b,c) Horizontal-sectioned epi-SHG/THG images of ex vivo human skin driven by the filtered source at 1100 nm. Imaging depth: 65 μm for (b) and 110 μm for (c). Cell morphology from the *stratum corneum* to the *stratum basale* in epidermis from THG contrast. Combined with the epi-SHG modality, the collagen fibers in the dermal papilla [arrowhead in (b)] are revealed. Magenta: THG, Green: SHG.

Collaborating with Dr. Rüdiger Greinert's group at Skin cancer center Buxtehude, we applied our SPM-enabled source to harmonic generation tomography of human skin. Unlike fluorescence microscopy, optical harmonics provide naturally endogenous contrasts; their

coherent nature offers additional insights to visualize the tissue morphology as bio-photonic crystals. Figure 9(b,c) show the combined SHG/third-harmonic generation (THG) images (SHG in green and THG in magenta) of ex vivo human skin from *stratum corneum* to the reticular dermis. Our SPM-enabled source is tuned to 1100 nm to drive the SHG/THG microscope. The cell morphology in epidermis (Figure 9(b)) and the collagen distribution in dermis (Figure 9(c)) can be clearly distinguished in the 10-frame-averaged video-rate (30Hz) images. >220 μm of imaging penetration depth from the top of the stratum corneum can be obtained. To our best knowledge, this work represents the first demonstration that a femtosecond wavelength-tunable fiber-source can enable THG imaging in the penetration window around 1030 nm – 1215 nm. Ongoing work is to further optimize the tunable ultrafast source and carry out a systematic study aiming to push the penetration depth beyond 1 mm that is necessary for skin cancer investigation. We are also collaborating with Prof. Hartmut Schlueter (from the University Medical Center Hamburg-Eppendorf) and Prof. Dwayne Miller (from Max Planck Institute for the Structure and Dynamics of Matter) to apply our MPM imaging technique to assist laser surgery.

Imaging and sorting protein nanocrystals: The relationship between structure and function of biological macromolecules is essential to many areas in the life sciences, pharmacy, medicine and biotechnology, because it provides an understanding about life processes, or their faults at the atomic level. To date, diffraction data for the structural analysis of biomolecules can be measured only at relatively large and regular crystals using synchrotron sources. Using high energy X-ray pulses from a free-electron laser (FEL), however, Prof. Henry Chapman's group at DESY were the first to determine the structure of protein nanocrystals. The capability of controlling production of protein nanocrystals becomes critical for subsequent analysis of these nanocrystals using FELs.

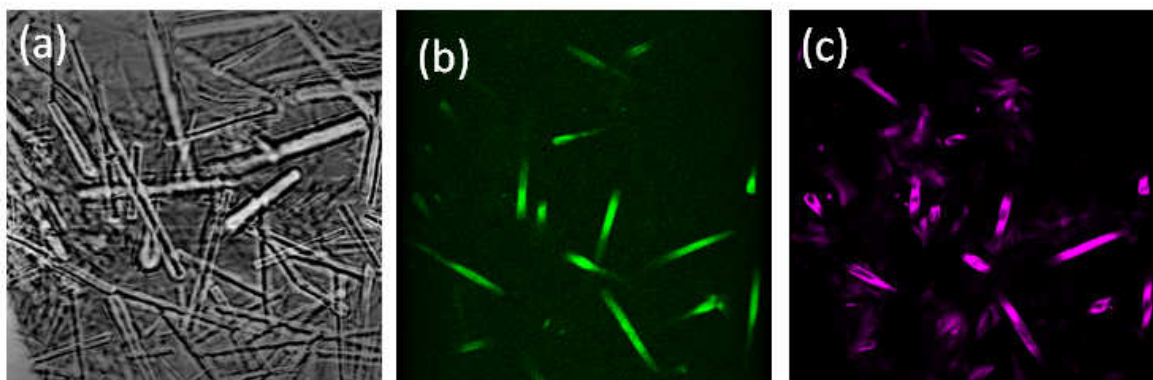


Fig. 10. Imaging protein nano-crystals using 1300-nm femtosecond pulses at different imaging contrast: (a) forward scattering, (b) second-harmonic generation, and (c) third-harmonic generation

To distinguish nanocrystals from amorphous particles, we are developing a harmonic generation microscope to image protein nanocrystals; the microscopy is driven by our SPM-enabled, Er-fiber laser based femtosecond source at 1300 nm. Figure 10 presents experimental results of imaging protein nano-crystals based on different imaging contrasts, i.e., forward scattering (Fig. 10(a)), SHG (Fig. 10(b)), and THG (Fig. 10(c)).

The development of such harmonic-generation microscopy method for protein nano-crystal imaging and sorting has enabled us to submit a joint proposal with Prof. Betzel's group (from University of Hamburg) to Federal Ministry of Education and Research, Germany in 2016, and we received 75,000 Euros to develop a fiber-laser driven harmonic-generation microscope. We also continuously received funding from the Hamburg Centre for Ultrafast Imaging (CUI) with the German excellence initiative to further improve the technology. In the future we will

integrate the microscope with the method of depolarized dynamic light scattering (which measures the size of nanocrystals) developed by Prof. Christian Betzel's group. The whole system permits a high quality and fast characterization of nanocrystals for the structural analysis with FELs. Professor Chapman is a further project partner involved to carry out the final measurements to verify the quality of the protein nano-crystals produced at FELs.

We believe that our tunable ultrafast laser source represents a cost-effective substitute of the conventional MPM driving source, i.e., a combination of Ti:sapphire laser plus a solid-state OPO. With the possibility of implementing our ultrafast source in an all-fiber format (fiber laser plus fiber-optic spectral broadening), this energy scalable source paves an avenue to operate MPM in rugged environments outside research labs. Several renowned laser companies (Such as Topica and Coherent) have expressed their interest in our laser technology, showing that our laser sources hold a great promise to be commercialized.

3) Qualification of Junior Researchers (max. 2 DIN A4 pages)

Please describe the structure of the Young Investigators Group in the course of the funding period and the main achievements regarding personal qualifications (including your own): Bachelor, Master; Diploma degrees, conferring of doctorates, "Habilitations", appointments/junior professorships, tenure track, awards, etc. Please also describe any particularities as well as your work-related plans after the end of the funding period.

Funded by Helmholtz Association, I started to set up the Helmholtz Young Investigator group—Ultrafast Laser Optics and Coherent Microscopy group—on August 1st, 2012 at the Center for Free-Electron Laser (CFEL), DESY. The research group aims to pushing the limits of ultrafast laser technologies with the applications for laboratory-based EUV/X-ray source, high power mid-IR ultrafast sources for spectroscopy, and multi-photon microscopy. The education of research personnel and students is an essential and deep-rooted component of this YIG group, which provides an excellent teaching vehicle for graduate students, postdoctoral associates, and visiting scientists. Participants learned aspects of photonics, quantum electronics, nonlinear dynamics, ultrashort pulse lasers, noise, and precision optical measurement techniques. In addition, the investigators teach courses in Nonlinear Optics and Ultrafast Optics.

Current group include the group leader (myself) and 5 PhD students. Wei Liu, the first PhD student joining the group, has graduated with the PhD degree from University of Hamburg. Now Wei is working as a postdoctoral research associate at SLAC, USA. Gengji Zhou, the second PhD student will defend his thesis on October 26th 2017; he will be a postdoctoral research fellow at the University of Michigan, USA. Table 1 lists the group members and their projects.

Table 1: current group members and their ongoing projects

Name	Starting time	Project
Wei Liu	01.09.2012	Advanced Yb-fiber lasers employing fiber nonlinearities
Gengji Zhou	01.09.2013	High power mid-IR laser source
Qian Cao	01.09.2014	GHz laser frequency comb
Hsiang-Yu Chung	01.10.2014	Multiphoton microscopy
Yi Hua	01.09.2015	Yb-fiber front-end seeding cryo Yb:YLF amplifier
Yizhou Liu	15.09.2015	High power Yb-fiber laser for table-top HHG source

My group hosted many visiting scholars, including 4 Master students, 2 PhD students, and 1 Associate Professor; see Table 2 for a full list. Some of the visiting students eventually returned to DESY to assume a higher position. For example, Rudrakant and Chen joined Dr. Ingmar Hartl's laser group as a PhD student and as a postdoc fellow, respectively; Hsiang-Yu is currently pursuing PhD degree in my group.

Table 2: visiting members in the group and their projects

Name	Affiliation	Visiting period	Project
Hsiang-Yu Chung (Master student)	National Taiwan University	01.03.2013—30.04.2013	All normal dispersion fiber lasers
Rudrakant Sollapur (Master student)	Imperial college (UK)	01.04.2013—31.07.2013	Bi-doped fiber amplifiers. (<u>This work became part of his Master thesis at imperial college, UK.</u>)
Jiawei Mei (Master student)	Tsinghua University (China)	01.07.2013—01.08.2013	Saturable absorber mode-locked fiber oscillators
Tao Chen (PhD student)	Zhejiang University (China)	01.09.2013—31.12.2013	High-power Yb-fiber laser
Chen Li (PhD student)	Beijing University (China)	01.09.2014—31.08.2015	Low-noise Yb-fiber lasers. (<u>He also contributed to the experimental results we published on an Optics Express paper and he is the second author.</u>)
Xiang Gao (Master student)	Karlsruhe Institute of Technology (Germany)	01.08.2015—31.10.2015	1 GHz Yb-fiber laser based frequency comb
Shanhui Xu (Associate professor)	South China University of Technology	15.07.2014—15.08.2014	High-repetition rate Yb-fiber laser

In December 2016, I was granted tenure by DESY after external evaluation. In September 2017, I was offered a Professor position at the Institute of Physics, Chinese Academy of Science.

4) Public relations

By which means did you gain publicity (e.g. reporting in media, own website)?

We mainly gain publicity by presenting our research results at renowned international conferences, such as Advanced Solid State Lasers Congress (ASSL), Conference on Lasers and Electro-Optics (CLEO), the international Ultrafast Optics conference (UFO), Focus on Microscopy (FOM), Europhoton etc. The research results achieved were presented >30 times to the attendees of these conferences, including several invited talks. These conference presentations have drawn attention of many other research groups and also several companies. As the group leader, I frequently traveled to different countries to give invited talks at Universities, national labs, and research institutes.

5) Networking

What co-operation and communication structures (centre/university if applicable) have been

developed during the course of the funding? How satisfied are you with the co-operation with the Helmholtz-Centre / university?

My group work in close collaboration with research groups at DESY, the University of Hamburg, and other institutes in Germany. Table 3 lists these collaborators, their affiliation, and our collaborating projects. For example, my group collaborate with Professor Franz Kärtner (my host at DESY) and Prof. Andreas Tünnermann and Prof. Jens Limpert (both from Helmholtz Institute at Jena) on developing high-power fiber laser sources to implement a table-top, high photon-flux EUV source. Collaborating with Professor Henry Chapman (from DESY) and Professor Christian Betzel (from University of Hamburg), my group developed a harmonic-generation microscope to analyze protein nanocrystals. Currently we are integrating the harmonic-generation microscope with an apparatus that measures the size of nanocrystals based on depolarized dynamic light scattering. The instrument will be installed at European XFEL in 2017. We further modified our harmonic-generation microscope and make it suitable for deep-tissue biomedical imaging applications. We are working with Dr. Rüdiger Greinert (Skin cancer center Buxtehude) to study skin cancers.

Table 3: Collaborators in Germany

Name	Affiliation	Collaborating Project
Prof. Franz Kärtner	DESY	Yb-fiber laser seeding source for cryo Yb:YLF amplifier; low noise Yb-fiber laser frequency combs for low-noise microwave extraction and THz optical parametric amplifier
Prof. Henry Chapman Prof. Christian Betzel	DESY University of Hamburg	Multiphoton microscopy imaging to analyze and sort protein nanocrystals. We are developing an instrument that incorporating both multiphoton microscopy imaging and depolarized dynamic light scattering. The instrument will be installed at European XFEL in 2017.
Dr. Markus Perbandt Dr. Alke Meents	DESY	On-chip room-temperature time-resolved protein micro-crystallography based on UV-induced structural changes
Prof. Dwayne Miller	DESY	Low repetition rate, high energy Yb-fiber laser for electron emission
Dr. Rob Meijers	DESY	Multiphoton microscopy imaging to analyze protein nanocrystals
Prof. Andreas Tünnermann Prof. Jens Limpert	Helmholtz Institute Jena	High power Yb-fiber lasers based on rod-type large pitch fiber. This group pioneer in designing and fabricating Yb-fibers with the largest mode area for single transverse mode operation.
Dr. Rüdiger Greinert	Skin cancer center Buxtehude	Second-harmonic generation and third-harmonic generation microscopy deep-tissue imaging for skin cancer diagnosis

My group also developed strong collaboration with many outside-Germany collaborators; see Table 4 for detailed information. For example, we are collaborating with Prof. Zhigang Zhang (from Beijing University, China) to develop low-noise Yb-fiber laser frequency combs for microwave extraction. Together with Prof. Jimin Zhao (from Institute of Physics, Chinese Academy of Science), we successfully set up a Joint Helmholtz-CAS research group, aiming at developing high power ultrafast mid-IR laser source and applying it to quantum material spectroscopy. Since the ultrafast sources developed in my group are based on fiber laser technology, optical fibers constitute the most critical device. We therefore collaborate with several prestigious international groups, which excel in fabricating specialty fibers. These fibers

include highly Yb-doped fibers (Prof. Zhongmin Yang and Prof. Shanhui Xu from South China University of Technology), chirally-coupled-core fiber with large mode area and low birefringence (Prof. Almantas Galvanauskas from University of Michigan, USA), Nd-doped fiber (Prof. Daniel Milanese from Politecnico di Torino, Italy), Bi-doped fiber (Prof. Eugeny Dianov from Russian Academy of Sciences, Russia).

Table 4: Collaborators outside Germany

Name	Affiliation	Collaboration Project
Prof. Zhigang Zhang	Beijing University (China)	1 GHz Yb-fiber laser frequency comb
Prof. Jimin Zhao	Institute of Physics, Chinese Academy of Science (China)	High power ultrafast mid-IR laser source for quantum material spectroscopy ("Joint Helmholtz-CAS research group")
Prof. Zhongmin Yang Prof. Shanhui Xu	South China University of Technology (China)	High repetition rate Yb-fiber lasers. <u>Their group manufactured the Yb-fibers with the highest possible doping concentration, a necessary requirement for constructing high repetition rate (> 1GHz) lasers.</u>
Prof. Yonghang Shen	Zhejiang University (China)	Generation of high power mid-IR pulses at 3-5 μ m using difference frequency generation. <u>His group fabricated PPLN with the largest aperture.</u>
Prof. Chi-Kuang Sun	National Taiwan University (Taiwan)	Compact harmonics microscopy imaging system
Prof. Almantas Galvanauskas	University of Michigan (USA)	High energy fiber-optic ultrafast source. His group invented chirally-coupled-core fiber, featuring large mode area and low birefringence.
Prof. Daniel Milanese	Politecnico di Torino (Italy)	Nd-fiber laser
Prof. Eugeny Dianov	Russian Academy of Sciences (Russia)	Bi-doped fiber amplifiers
Dr. Sebastien Ermeneux	ALPHA NOV optics & laser technology center (France)	Fiber laser control system

6) List of Publications

Articles in scientific journals, written contributions to scientific meetings, contributions to books, other publications.

Journal papers:

- [10] H. -Y. Chung, W. Liu, Q. Cao, F. X. Kaertner, and **G. Q. Chang**, "Er-fiber laser based, energy scalable ultrafast sources tunable in 1300-1700 nm," Opt. Express 25, 15760 (2017)
- [9] W. Liu, S. -H. Chia, H. -Y. Chung, F. X. Kaertner, and **G. Q. Chang**, "Energetic ultrafast fiber laser sources tunable in 1030-1215 nm for deep tissue multiphoton microscopy," Opt. Express 25, 6822 (2017)
- [8] W. Liu, C. Li, Z. Zhang, F. X. Kaertner, and **G. Q. Chang**, "Self-phase modulation enabled, wavelength-tunable fiber laser sources: an energy scalable approach," Opt. Express 24, 15319 (2016)
- [7] F. X. Kaertner, F. Ahr, A. -L. Calendron, H. Cankaya, S. Carbajo, **G. Q. Chang**, G.

Cirmi, K. Doerner, U. Dorda, A. Fallahi, T. Hartin, M. Hemmer, R. Hobbs, Y. Hua, R. Huang, R. Letrun, N. Matlis, V. Mazalova, O. Muecke, E. Nanni, W. Putnam, K. Ravi, R. Reichert, I. Sarrou, X. Wu, H. Ye, L. Zapata, D. Zhang, C. Zhou, R. J. D. Miller, K. Berggren, H. Graafasma, A. Meents, R. W. Assmann, H. N. Chapman, and P. M. –L. Fromme “AXSIS: exploring the frontiers in attosecond X-ray science, imaging and spectroscopy,” Nuclear Instruments and Methods in Physics Research A S0168900216002564 (2016)

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- [5] A. G. Glenday, C. –H. Li, N. Langellier, **G. Q. Chang**, L.-J. Chen, G. Furesz, A. Zibrov, F. X. Kärtner, D. F. Phillips, D. Sassellov, A. Szentgyorgyi, and R. L. Walsworth, “Operation of a broadband visible-wavelength astro-comb with a high-resolution astrophysical spectrograph,” Optica 2, 250 (2015)
- [4] W. Liu, D. N. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “Pre-chirp managed nonlinear amplification in fibers delivering 100 W, 60 fs pulse,” Opt. Lett. 40, 151 (2015).
- [3] J. K. Lim, H. –W. Chen, S. H. Xu, Z. M. Yang, **G. Q. Chang**, and F. X. Kaertner, “3 GHz, Watt-level femtosecond Raman soliton source,” Opt. Lett. 39, 2060 (2014)
- [2] S. –H Chia, L. –J Chen, Q. Zhang, O. D. Muecke, **G. Q. Chang**, and F. X. Kaertner, “Broadband continuum generation in mode-locked lasers with phase-matched output couplers,” Opt. Lett. 39, 1445 (2014)
- [1] H. –W. Chen, H. Zia, J. K. Lim, S. H. Xu, Z. M. Yang, F. X. Kaertner, and **G. Q. Chang**, “3 GHz, Yb-fiber laser based, few-cycle ultrafast source at the Ti:sapphire laser wavelength,” Opt. Lett. 38, 4927 (2013)

Conference papers:

- [42] **G. Q. Chang**, “Advanced ultrafast laser sources harnessing fiber nonlinearities,” Paper STh3k.1, CLEO/QELS, San Jose (2017) (Invited)
- [41] Q. Cao, F. X. Kaertner, and **G. Q. Chang**, “Towards high power and low noise mid-infrared DFG ultrafast source,” Paper JTU3L.5, CLEO/QELS, San Jose (2017)
- [40] H. –Y. Chung, W. Liu, and **G. Q. Chang**, “Er-fiber laser enabled femtosecond source tunable from 1.3-1.7 μm for nonlinear optical microscopy,” Paper SM3L.2, CLEO/QELS, San Jose (2017)
- [39] H. –Y. Chung, W. Liu, F. X. Kaertner, and **G. Q. Chang**, “Femtosecond source widely tunable from 1.3-1.7 μm for three-photon microscopy,” Paper P1-A/4, Focus on Microscopy (2017)
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- [37] G. J. Zhou, M. Xin, Y. Z. Liu, F. X. Kaertner, and **G. Q. Chang**, “SPM-enabled fiber laser source beyond 1.2 μm ,” Advanced Solid-State Lasers, Boston (2016)
- [36] W. Liu, S. –H. Chia, H. –Y. Chung, F. X. Kaertner, and **G. Q. Chang**, “Energy

scalable ultrafast fiber laser sources tunable in 1030-1200 nm for multiphoton microscopy,” Advanced Solid-State Lasers, Boston (2016)

- [35] Y. Z. Liu, W. Liu, D. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “100-W few-cycle Yb-fiber laser source based on pre-chirp managed amplification employing circular polarization,” Advanced Solid-State Lasers, Boston (2016)
- [34] W. Liu, Y. Z. Liu, D. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “Pre-chirp managed nonlinear amplification for >100 W ultrafast sources,” paper PA115-22, SPIE Photonics Asia, Beijing (2016) (invited)
- [33] W. Liu, Y. Z. Liu, D. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “Pre-chirp managed nonlinear amplification for >100 W ultrafast sources,” paper 2586791, the 8th International Symposium on Ultrafast Phenomena and Terahertz Waves, Chongqing, China (2016) (invited)
- [32] W. Liu, C. Li, H. –Y. Chung, S. –H. Chia, Z. G. Zhang, F. X. Kaertner, and **G. Q. Chang**, “Ultrafast fiber laser source tunable in 825-1210 nm for multi-photon microscopy,” paper FWG-3, Europhoton, Vienna (2016)
- [31] Y. Z. Liu, W. Liu, D. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “Energy scaling of pre-chirp managed nonlinear amplification using circular polarization,” paper 18.2, Europhoton, Vienna (2016)
- [30] J. Ruppe, S. Y. Chen, T. Zhou, M. Sheikhsola, Z. G. Zhang, **G. Q. Chang**, F. X. Kaertner, J. Nees, and A. Galvanauskas, “Coherent pulse stacking extension of CPA to 9 ns effectively-long stretched pulse duration,” paper SM4I.2, CLEO/QELS, San Jose (2016)
- [29] Q. Cao, C. Li, Y. Z. Liu, X. Gao, Z. G. Zhang, F. X. Kaertner, and **G. Q. Chang**, “Passively offset-free Yb: fiber laser source at 1 GHz repetition rate,” Paper JTh2A.141, CLEO/QELS, San Jose (2016)
- [28] Y. Hua, W. Liu, L. E. Zapata, M. Hemmer, G. J. Zhou, N. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “87-W, 1018-nm Yb-fiber ultrafast seeding source for cryogenic Yb:YLF amplifier,” Paper SM4Q.5, CLEO/QELS, San Jose (2016)
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- [26] H. –Y. Chung, W. Liu, S. –H. Chia, F. X. Kaertner, and **G. Q. Chang**, “Fiber-nonlinearity enabled femtosecond laser sources for nonlinear light microscopy,” Paper P1-C/32, Focus on Microscopy (2016)
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- [24] Q. Cao, F. X. Kaertner, and **G. Q. Chang**, “Timing jitter optimization of Raman soliton and dispersive wave,” Paper UFO0092, Ultrafast Optics X (2015)
- [23] W. Liu, C. Li, Z. G. Zhang, F. X. Kaertner, and **G. Q. Chang**, “Yb-fiber laser based

- ultrafast source emitting 50-fs pulses at 920 nm,” Paper UFO0123, Ultrafast Optics X (2015)
- [22] G. J. Zhou, M. Xin, F. X. Kaertner, and **G. Q. Chang**, “Timing jitter optimization of Raman solitons,” Paper UFO0055, Ultrafast Optics X (2015)
 - [21] J. K. Lim, H. –W. Chen, S. H. Xu, Z. M. Yang, F. X. Kaertner, and **G. Q. Chang**, “3 GHz frequency comb via difference frequency generation,” Ultrafast Optics X (2015)
 - [20] D. F. Phillips, A. G. Glenday, C.-H Li, N. Langellier, **G. Q. Chang**, G. Furesz, F. X. Kaertner, D. Sassellov, A. Szentgyorgy, and R. Walsworth, “Solar oscillations and the search for Venus enabled by a laser frequency comb,” paper M8.00005, 46th Annual meeting of the APS (DAMOP), Columbus (2015)
 - [19] C.-H Li, A. G. Glenday, D. F. Phillips, N. Langellier, **G. Q. Chang**, G. Furesz, F. X. Kaertner, D. Sassellov, A. Szentgyorgy, and R. Walsworth, “Progress with a green astro-comb for exoplanet searches,” paper D1.00060, 46th Annual meeting of the APS (DAMOP), Columbus (2015)
 - [18] D. Schimpf, W. Liu, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “CPA-free ultrafast fiber laser source based on pre-chirp managed nonlinear amplification,” paper STu10.4, CLEO/QELS, San Jose (2015)
 - [17] G. J. Zhou, M. Xin, F. X. Kaertner, and **G. Q. Chang**, “Relative timing jitter and its effect on nonlinear wavelength conversion,” paper SF2D.4, CLEO/QELS, San Jose (2015)
 - [16] **G. Q. Chang** and F. X. Kaertner, “Ultrafast fiber laser technologies for multiphoton microscopy,” paper TU-AF2-PAR-E-1, Focus on Microscopy, Gottingen, Germany (2015)
 - [15] **G. Q. Chang**, C.-H Li, A. G. Glenday, G. Furesz, N. Langellier, J. K. Lim, H. –W. Chen, D. F. Phillips, D. Sassellov, A. Szentgyorgy, R. Walsworth, and F. X. Kaertner, “Femtosecond laser frequency comb for precision calibration of HARPS-N,” paper ATh3A.2, Advanced Solid-State Lasers, Shanghai (2014)
 - [14] G. J. Zhou, M. Xin, W. Liu, F. X. Kaertner, and **G. Q. Chang**, “Relative intensity noise and timing jitter of Raman solitons,” paper AM5A. 36, Advanced Solid-State Lasers, Shanghai (2014)
 - [13] W. Liu, D. Schimpf, T. Eidam, J. Limpert, A. Tuennermann, F. X. Kaertner, and **G. Q. Chang**, “Pre-chirp managed amplification (PCMA) in fibers to 100 W with 60-fs output pulse duration,” paper AW4A.2, Advanced Solid-State Lasers, Shanghai (2014)
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- [5] H. –W. Chen, J. K. Lim, Shanhui Xu, Zhongmin Yang, **G. Q. Chang**, and F. X. Kaertner, “3 GHz few-cycle ultrafast source at 850 nm,” postdeadline paper FW6B.2, Frontiers in Optics, Orlando (2013)
- [4] H. –W. Chen, H. Zia, J. K. Lim, **G. Q. Chang**, and F. X. Kaertner, “Yb-fiber oscillator based, few-cycle ultrafast source at 850 nm,” paper WA1-1, CLEO PR, Kyoto (2013) (best paper award)
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- [2] J. K. Lim, H. –W. Chen, **G. Q. Chang**, and F. X. Kaertner, “Stable frequency comb derived from a narrowband Yb-fiber laser: pre-chirp management for self-referenced fceo stabilization,” paper CM2I.7, CLEO/QELS, San Jose (2013)
- [1] **G. Q. Chang**, H. –W. Chen, J. K. Lim, S. H. Xu, Z. M. Yang, and F. X. Kaertner, “3 GHz, femtosecond Raman soliton source,” paper CM2I.4, CLEO/QELS, San Jose (2013)