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TOPICAL REVIEW — Synergetic Extreme Condition User Facility: Breakthroughs and opportunities for the research of physical science

Femtosecond laser user facility for application research on ultrafast science*

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The advent of chirped-pulse amplification (CPA) has greatly advanced the field of ultrafast and ultra-intense laser technology. CPA has become an indispensable platform for multidisciplinary research, such as physics, chemistry, life sciences, and precision metrology. The femtosecond laser facility at the Synergic Extreme Condition User Facility (SECUF) is a comprehensive experimental platform with an advanced femtosecond laser source for ultrafast scientific research. It will provide an ultrafast scientific research system having a few-cycle pulse duration, wide spectral range, high energy, and high repetition rate for multipurpose applications.

Keywords: femtosecond, carrier envelope phase (CEP), chirped-pulse amplification (CPA), ultrafast spectroscopy, optical parametric amplifier (OPA)

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1. Introduction

The development of the ultrafast laser has been one of the most important directions of laser science in recent years. It has become an indispensable platform and tool in numerous fields, such as physics, chemistry, life sciences, information, precision metrology, national defense, and manufacturing. It has also developed many new disciplines, such as high-energy density physics, laser particle acceleration, and optical atomic clocks. In 1999, Prof. A. Zewail won the Nobel prize for studying the kinetics of chemical reactions with a femtosecond laser.[1] In 2005, Prof. T. W. Hänsch and Prof. J. L. Hall shared the Nobel prize for the study of a femtosecond-laser frequency comb.[2] With the further development of the ultrafast laser, advanced amplification technology has been able to generate a peak power on the order of petawatts,[3–6] and a focused intensity greater than 10^{22} W/cm^2 has been achieved in a conventional laboratory. Using the extra-cavity compression technique, scientists have achieved shorter than 3 fs pulses in the visible spectrum, which correspond to just one optical period. According to the interaction between the femtosecond laser and inert gas, scientists obtained a single pulse shorter than 100 as[7] and an x-ray laser source.[8] By using an ultrashort and ultra-intense femtosecond laser with a peak power approaching the petawatts range, scientists can produce extreme conditions similar to those of celestial bodies or nuclear explosions in the laboratory environment,[9] thus obtaining the state of materials under the extreme conditions of a high energy density, temperature density, etc. Such a laser can also produce secondary particle sources, such as high-energy electron beams and ion beams with ultrafast characteristics and high brightness. Coherent radiation, such as x-rays, extreme-ultraviolet (XUV) lasers, super-continuum[10,11] and terahertz radiation,[12–14] can also be obtained using ultrafast lasers. Thus, this technology not only brings new vitality to the development of many basic sciences, interdisciplinary frontiers, and strategic high-tech disciplines but also introduces new disciplines and high-tech applications, including atomic and molecular physics, chemical reaction dynamics, life science, precision manufacturing, and modern national defense. The femtosecond laser subsystem at the SECUF is a comprehensive experimental platform based on a femtosecond laser source for ultrafast scientific research. It is a multi-purpose ultrafast scientific research device with a wide spectral range, high energy, and high repetition rate. Considering the latest advances in ultrafast phenomena and the latest femtosecond laser technology, the femtosecond ultrafast experimental subsystem will offer the following functions.

(i) Multi-angle observation function The ultrafast spectroscopic experimental device based on the femtosecond laser can observe weak bond interactions, synergistic effects, coherent energy transfer processes, and dynamic behavior in the complex molecular systems of bulk-phase water, molecular crystals, and two mixtures (solution). Through theoreti-

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cal simulation, scientists can obtain physical and chemical images.

(ii) Multidimensional light field regulation function Using ultrafast pulse laser shaping technology, the ultrafast spectral experimental device allows the study of quantum coherence regulation on the femtosecond scale, helps to deepen our understanding of quantum coherent energy transmission in the complex phase of the condensed phase, and provides an important basis for further study of the quantum regulation of the condensed phase.

(iii) High peak power function The system provides a periodic laser pulse with a peak power in the near-terawatt range. After focusing, it can interact with matter on a sub-10 fs timescale. It can perform unprecedented extreme ultrafast nonlinear optical studies involving high solid-surface high-harmonic and attosecond pulses, ultrafast plasma kinetics, etc.

(iv) Ultrafast fluorescence measurement Femtosecond time-resolved transient fluorescence optical parametric amplification (OPA) spectrometers use ultrashort pulsed lasers to reveal the kinetics of molecular fluorescence transitions from the excited state to the ground state and are widely used in many research fields, such as luminescent materials, photoelectric conversion, bio-optical physics, and photochemical processes.

The femtosecond laser subsystem at the SECUF will integrate the most advanced femtosecond laser technology and ultrafast research methods. Upon its completion, it will satisfy the requirements of ultrafast detection of various systems and objects by users at home and abroad. This femtosecond laser subsystem can be used either for the study of ultrafast dynamics for important scientific issues or for the exploration of new directions.

2. Description of system

The femtosecond laser subsystem at the SECUF consists of a high-average power femtosecond laser source, a phase-locked few-cycle laser source, a mid-infrared OPA source, a liquid-phase femtosecond spectroscopy experimental unit, a gas-phase femtosecond spectroscopy experimental unit, an ultra-nonlinear optical ultrafast experimental unit, and a femtosecond time-resolved transient fluorescence optical parametric amplified spectroscopic experimental unit. The conceptual design of the overall system is shown in Fig. 1.

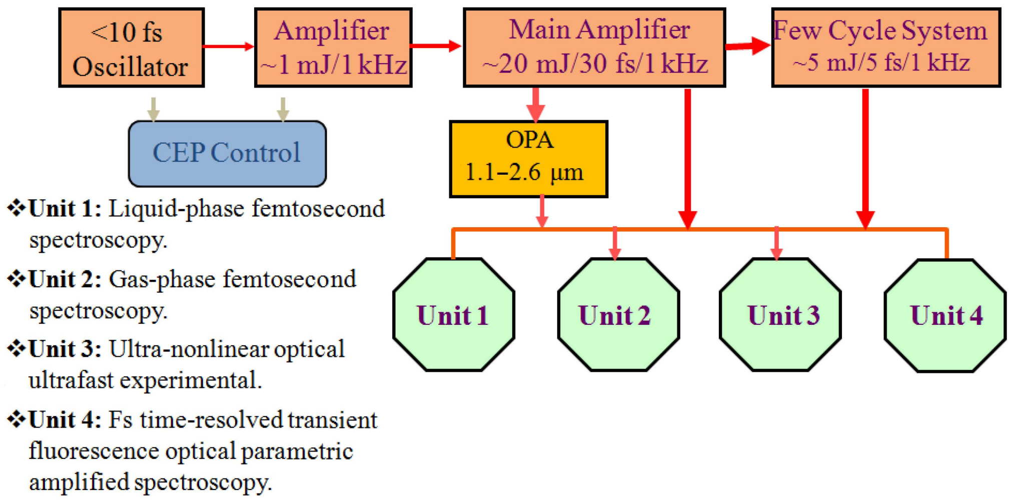

Fig. 1. (color online) Ultrafast femtosecond laser experimental subsystem structure.

The advanced femtosecond laser source is based on the chirped-pulse amplification (CPA) technique.[15] A sub-10 fs femtosecond Ti:sapphire oscillator[16] is used for the seed source. The parameters of this laser source are obtained via CPA, spectral broadening, pulse recompression, optical parameter wavelength tuning, carrier-envelope phase (CEP) locking, and other technologies. The basic technical route is to use an ultra-broadband femtosecond oscillator as the seed for CPA to obtain a laser pulse with a single-pulse energy of 20 mJ, a pulse duration of $<$ 40 fs, and a repetition rate of 1 kHz. The high-power femtosecond laser pulse output is injected into a waveguide fiber filled with helium gas to achieve spectral broadening and chirped-mirror compression to obtain a single-pulse energy greater than 5 mJ and a pulse duration of 7 fs. On this basis, the CEP of the oscillator and the amplifier is phase locked separately with a fast and a slow feedback control loop. In addition, a high-average power femtosecond laser is injected into an OPA to obtain a mid-infrared femtosecond laser with a wavelength-tuning range of 1.1–2.6 μm and a single-pulse energy of approximately 1 mJ@1.3 μm. Then, the laser is injected into each experimental unit.

3. Experimental setup and design considerations

The femtosecond laser at the SECUF consists of an advanced femtosecond laser light source, a liquid-phase femtosecond spectroscopy experimental unit, a gas-phase femtosecond spectroscopy experimental unit, an extreme-nonlinear optical ultrafast experimental unit, and a femtosecond time-resolved transient fluorescence OPA spectroscopy experimental unit. Details regarding each unit are presented in this section.

3.1. Advanced femtosecond laser source

i) High-average power femtosecond laser source

The high-average power femtosecond laser source includes a sub-10 fs oscillator, a stretcher, a regenerative amplifier, a multi-pass amplifier, a compressor, a synchronized controller, and several sets of pump lasers. The oscillator generates high-quality seed laser pulses with a pulse duration of approximately 10 fs. Then, these pulses are stretched to 200 ps by a Martinez stretcher and injected into the regenerative laser cavity for pre-amplification.[17] The pumping source is a commercial all-solid-state Q-switched green laser with an average power of 20 W and a repetition rate of 1 kHz. The energy of the laser pulse from the regenerative amplifier is larger than 4 mJ. Then, the laser pulse is injected into the second-stage multi-pass amplifier. The multi-pass amplification pump source comprises four commercial all-solid-state Q-switched green lasers with an average power of 25 W and a repetition rate of 1 kHz. The pump laser and the amplified laser have optimized delay, coupled matching, compensation for thermal effects in the amplification process, and other means. Eventually, energy of 30 mJ is obtained with good beam quality. Finally, to achieve compression, the amplified laser pulse is input into a compressor that is opposite to the stretcher. By optimizing the dispersion compensation, a pulse duration of approximately 40 fs and energy of over 20 mJ are obtained, corresponding to an average power of over 20 W. The design of the laser source is shown in Fig. 2.

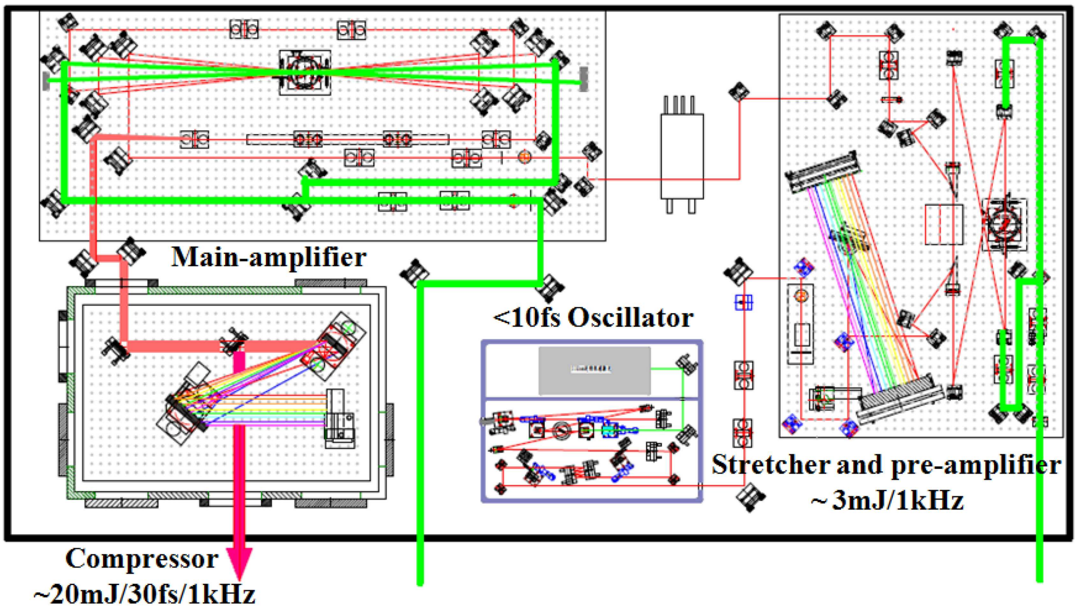

Fig. 2. (color online) Design of the high-average power femtosecond light source.

ii) Few-cycle femtosecond laser pulses and CEP-locked components

Traditionally, the differentially pumped hollow fiber is effective for generating few-cycle laser pulses. Compared with the statically gas fill scheme, it has the advantages of a high transmission rate and a wide spectral-broadening range. First, the laser is focused into a waveguide fiber filled with a certain gas for broadening the spectrum. The spectrum is broader than an octave, and then a high-dispersion chirped-mirror combination compressor is used to accurately compensate the dispersion. The stretched laser pulse is compressed to few-cycle to achieve a laser-pulse output with energy greater than 5 mJ and a pulse duration shorter than 7 fs. For few-cycle laser pulses, the locking of the CEP is achieved by locking the femtosecond Ti:sapphire oscillator and the amplifier separately. In the oscillator part, the difference frequency generation (DFG) technique is used to produce 1.5-μm infrared light of the broadband femtosecond laser via a periodically poled lithium niobate crystal.[18–20] The spectrum produced by the two parts contains the carrier-envelope offset phase signal of the output pulse obtained via the beat frequency method. The phase-locked loop is fed into the acousto-optic modulator, which can control the pump laser power to realize the CEP locking of the oscillator, yielding a "fast phase-locked loop." The CEP locking of the amplifier uses the f–$2f$ spectral interference technique. The interference spectrum of the f–$2f$ spectral interference system is obtained by using the spectrum broadening of the foregoing waveguides. The phase-drift information in the amplifier is obtained by analyzing the interference spectrum, and the feedback control is obtained by changing the insertion of the prism in the compressor, eventually causing the phase locking of the amplified pulse, which is called a "slow phase-locked loop." After the phase of the oscillator and the amplifier is locked, the CEP-locked high-energy laser pulse can be obtained.

iii) High-energy mid-infrared laser source

A multi-stage OPA is pumped by a 20-W high-average power femtosecond laser. First, the super-continuum is generated by injecting pump energy of approximately 1 mJ into a crystal. The wavelength range is obtained as 400–600 nm by controlling the power density of the crystal, and then the pump energy is injected into a multi-stage OPA pumped by 15-mJ femtosecond laser pulses. The bandwidth and gain in each stage of the OPA are controlled independently. The stability of the spectrum and energy is maintained when the parametric fluorescence is suppressed. A tunable wavelength of 1.1–2.6 μm is obtained. By using a prism pair and a chirped mirror to compensate for the dispersion, a femtosecond and widely tuned mid-infrared laser can be obtained. By optimizing the crystal angle and the pumping parameters, the maximum power of the optical parametric laser is achieved at the wavelength of 1.3 μm, with a maximum pulse energy of larger than 1 mJ and a pulse duration of approximately 100 fs.

3.2. Liquid-phase femtosecond spectroscopy experimental unit

The liquid-phase femtosecond spectroscopy experimental unit[21] is a femtosecond spectroscopic user experimental platform that combines linear infrared and Raman spectroscopy with femtosecond pump-detection technology, including the combination of infrared spectroscopy, Raman spectroscopy, and absorption spectroscopy. This system includes an OPA unit, a pump-detection unit, a Raman spectrometer, an infrared spectrometer, a temperature-control system, and a vacuum pump unit. Thus, the platform allows the measurement of the time behavior of complex molecular dynamics and is particularly suitable for the study of condensed-phase multi-particle systems in which weak bond interaction plays an important role. The ultrafast spectroscopic and kinetic experimental platform employs OPA lasers as a pump source and super-continuum white light as a probe source. This platform can be used for the study of various types of micro/nanostructures (liquid or thin film) or complex molecular complex systems. Combined with ultrafast pulse laser shaping (amplitude and phase modulation), CPA, compression, and other technical means, it can also be used to research quantum coherence in the time domain. The overall design of the experimental unit is shown in Fig. 3.

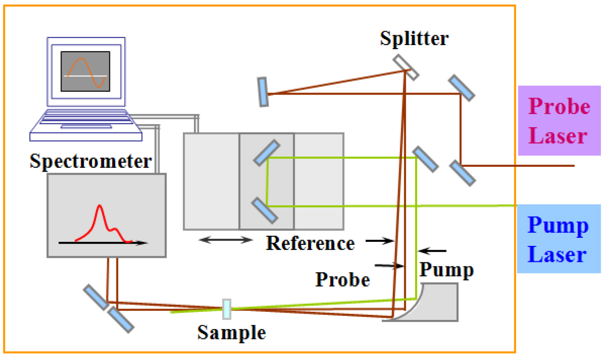

Fig. 3. (color online) Diagram of the liquid femtosecond spectroscopy experimental unit.

3.3. Gas femtosecond spectroscopy experiment unit

This experimental unit combines pump-detection technology with electronic[14] ion-momentum (energy) resolution measurement to achieve non-adiabatic dynamics of the gas-molecule excited state, quantum control, and other aspects of the study. The femtosecond lasers are divided into pump and probe lasers and focused on the molecular beam in the electron and ion velocity imaging device. Simultaneously, time-resolution and momentum-resolution measurements of the molecular fragments are performed.

The experimental unit is composed of a wavelength-tunable laser source, a pump-detection delay device, and an electron and ion velocity imager. Electron and ion velocity imagers are mainly composed of two parts: a beam source chamber and an ionization chamber. In the beam source chamber, a 1-kHz pulse valve produces the molecular beam through supersonic diffusion. The molecular beam enters the ionization chamber through a high-speed nozzle. In the ionization chamber, the molecular beam intersects the two femtosecond laser pulses perpendicularly. One of the laser beams excites the molecule to the electronic excited state (as a pump), and the other laser excites the excited state. The molecules or reaction products are ionized (as probes). The ionized electrons or ions are focused by the ion lens velocity onto the micro-channel plate detector to generate a two-dimensional electron or ion map and recorded by a charge-coupled device camera. The overall design of the experimental unit is shown in Fig. 4.

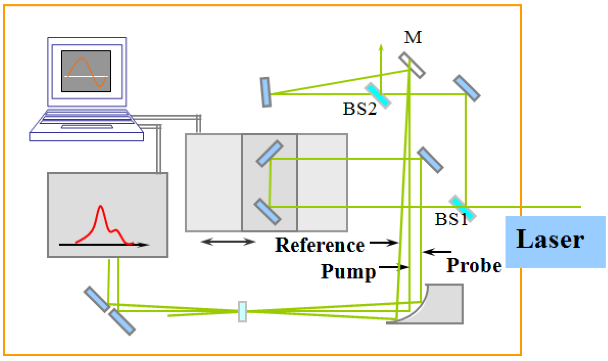

Fig. 4. (color online) Diagram of the gas-phase femtosecond spectroscopy experimental unit.

3.4. Extremely nonlinear optical ultrafast experimental unit

This experimental unit is designed as an extreme-nonlinear optical ultrafast research platform based on the CEP-locked high-energy few-cycle laser (5 mJ/7 fs/1 kHz, CEP-locked). The high-energy CEP-locked few-cycle laser interacts with a solid surface to generate high-order harmonics. Because this harmonic generation is limited to an ultrashort time, the extreme-nonlinear process can be measured by adjusting the CEP of the driving pulse.

The experimental platform includes a CEP-locked few-cycle laser source, interacting target chambers, XUV spectrometers, vacuum chambers, target components, and ultra-high vacuum pump units. The CEP-locked few-cycle laser is focused onto a solid target by a small-F-number off-axis parabolic concave mirror. The solid target is mounted on a four-dimensional platform and turned one position at a time to ensure that each occurrence is a new interaction point. The high-order harmonics are detected via XUV spectrometry. The design of the whole system is shown in Fig. 5.

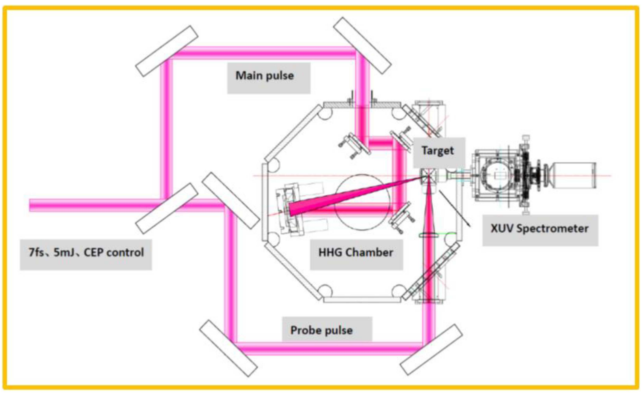

Fig. 5. (color online) Diagram of the extreme-nonlinear optical ultrafast experimental unit.

3.5. Femtosecond time-resolved transient fluorescence OPA spectrometer experimental unit

The femtosecond time-resolved fluorescence OPA experiment[22–24] system is a transient fluorescence measurement technology based on the non-collinear OPA principle.[25,26] It not only can measure the entire broadband spectrum at the same time but also has a gain of nearly 10^6 with respect to the fluorescence signal. It can be used to achieve femtosecond time-resolved measurement of weak broadband fluorescence signals. The measurement sensitivity is 30 fluorescent photons for a 100-fs timescale.

The experimental unit mainly includes femtosecond laser sources, non-collinear OPAs, fluorescence OPA devices, pump-detection devices, and data-measurement systems. Using the high-average power femtosecond laser as a light source, the pump laser is divided into two beams by a beam splitter. One of the beams is frequency-doubled to generate a pump light with a center wavelength of 400 nm for the fluorescence parametric amplification system. The other beam is used as a collinear or non-collinear OPA pump source. The wavelength is tunable, which can be exploited to excite samples to produce fluorescence. The fluorescence is amplified in a barium-borate crystal via a non-collinear OPA process, and data acquisition of the fluorescence-amplification signal is performed using a related instrument. The entire system design is shown in Fig. 6.

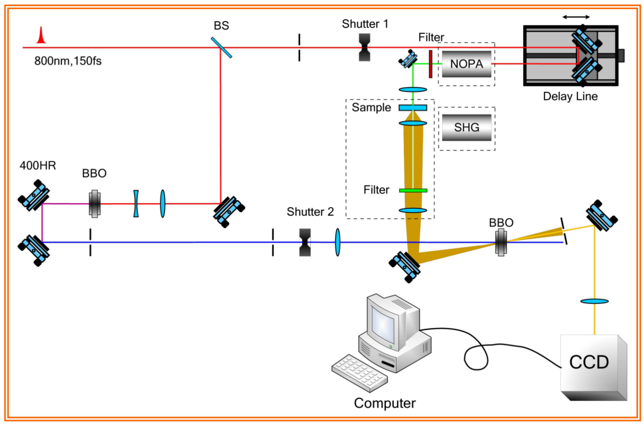

Fig. 6. (color online) Diagram of the femtosecond time-resolved transient fluorescence optical parameter amplification spectrometer experimental unit.

4. Conclusion and perspectives

A comprehensive physical dynamics research system based on the femtosecond laser technique for physical transients in physics, chemistry, materials, biology, medicine, and other disciplines will eventually be established. Considering the most important issues in the field of ultrafast science, several important research directions are selected, such as the pulse types, wavelengths, and acquisition methods for the final signals, to satisfy the requirements of multidisciplinary frontier development for the research of physical structural dynamics. Fully considering the requirements of scientists, the femtosecond laser subsystem at the SECUF will be developed as a system of scientific advancement and a perfect user-service system to attract experts and scholars in various fields to perform experiments, eventually becoming a world-class ultrafast scientific research center.

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