Ultrafast imaging for uncovering laser–material interaction dynamics

Du Wang1 | Shubin Wei1 | Xiandan Yuan1 | Zhiyuan Liu1 | Yueyun Weng2 | Yuqi Zhou3 | Ting-Hui Xiao3 | Keisuke Goda1,3,4 | Sheng Liu1,2 | Cheng Lei1,3

1The Institute of Technological Sciences, Wuhan University, Wuhan, China
2The Key Laboratory of Transients in Hydraulic Machinery of Ministry of Education, School of Power and Mechanical Engineering, Wuhan University, Wuhan, China
3Department of Chemistry, University of Tokyo, Tokyo, Japan
4Department of Bioengineering, University of California, Los Angeles, California, USA

Abstract
The physical mechanism of the dynamics in laser–material interaction has been an important research area. In addition to theoretical analysis, direct imaging-based observation of ultrafast dynamic processes is an important approach to understand many fundamental issues in laser–material interaction such as inertial confinement fusion (ICF), laser accelerator construction, and advanced laser production. In this review, the principles and applications of three types of commonly used ultrafast imaging methods are introduced, including the pump–probe, X-ray diagnosis, and single-shot optical burst imaging. We focus on the technical features such as the spatial and temporal resolution for each technique, and present several conventional applications.

KEYWORDS
high-energy-density physics, laser–material interaction, laser processing, ultrafast dynamics, ultrafast imaging

1 | INTRODUCTION
The laser is one of the most important inventions in the 20th century, which has markedly improved scientific research capabilities and the quality of human life. With the continuous advancements in laser technology, the output power of lasers has significantly increased. For example, the widely used fiber laser has achieved 10 kW-level continuous wave (CW) output with a single fiber,1,2 and over 100 kW laser power through beam combining technology for broad applications in laser processing, laser weapons, and other fields.3 The high-energy pulsed laser has been developed with a maximum pulse energy of 1 MJ for the nanosecond laser4 and approximately 10 PW peak power for the femtosecond laser.5 The introduction of the high-power laser source allowed creating a series of new frontier interdisciplinary subjects, including inertial confinement fusion (ICF),6 laser-driven particle accelerator,7,8 strong field quantum electrodynamics,9 and laser material processing.10 For many of those topics, the interaction between the high-power laser and materials has resulted in new interesting and fundamental physics problems. When the laser intensity exceeds $10^{18}$ W/cm$^2$, the majority of the irradiated materials are ionized instantaneously and form high-energy-density states of material. This highly nonlinear process is accompanied by...
new physical phenomena and effects, which are commonly analyzed by large-scale numerical simulations to understand their internal mechanisms. Similarly, in the field of laser material processing, the fundamental scientific problems behind the material removal process remain unanswered. In ICF facilities, understanding the damage mechanism of optical elements under intense laser irradiation has been a persisting challenge for scientists. Therefore, direct imaging observations of the ultrafast dynamic process of laser-material interaction are of great scientific value for revealing its physical mechanism and validating the dynamics modeling.

In the process of laser–material interaction, the thermalization time of optically excited electrons is of the order of femtoseconds. Phonon relaxation occurs within several picoseconds, thermal diffusion and shock wave generation range from tens of picoseconds to nanoseconds, and the dynamics of molten pool occurs of a microsecond order. The frame rate of conventional array sensors, such as charge-coupled devices (CCDs) and complementary metal oxide semiconductors (CMOS), is not large enough for temporally resolved photography of the aforementioned ultrafast dynamics. The pump–probe technique is a commonly used optical detection method to observe the ultrafast events initiated by a laser beam, through imaging or non-imaging strategies. Due to its relatively simple and flexible optical setup, adaptive temporal and spatial resolution can be achieved when observing repeatable phenomena (Section 2). In high-energy-density physics, the laser plasma radiation spectrum is in the energy range of $10^7$–$100$ keV. Accordingly, several time-resolved X-ray imaging techniques have been developed as diagnostic methods (Section 3). A series of all-optical ultrafast imaging techniques based on single-shot laser pulses have been developed to capture these nonrepeatable ultrafast phenomena. Due to these advancements, the temporal resolution has been improved to hundreds or even tens of femtoseconds (Section 4). In the following three sections of this review, we introduce the applications of these imaging techniques involved in the dynamics of laser–material interaction. The scope of discussion mainly focuses on the application of high-power laser, including strong-field physics, damage mechanisms, and laser processing methods.

## 2 | TIME-RESOLVED PUMP–PROBE IMAGING

The origin of time-resolved pump–probe (TRPP) detection technology can be traced back to over a century ago. Common optical pump-probe techniques include time-resolved reflection/transmission spectroscopy, transient absorption spectroscopy, time-resolved Raman scattering spectroscopy, time-resolved photoluminescence spectroscopy, surface/bulk second harmonic generation, time-resolved four-wave mixing, and time-resolved infrared/THz time-domain/X-ray spectroscopy. In addition to spectroscopy, pump–probe imaging is the most reported transient method used within the imaging technology field. Currently, researchers mainly from two fields are focusing on the use of pump–probe imaging technology to study the mechanism of laser–material interaction: laser material processing and ultraprecision machining of optical elements. The former focuses on the mechanism of material modification or ablation under laser irradiation with the goal of contributing to enhancing the optimization of processing parameters through the study of dynamic processes. The latter mainly provides high-quality and high damage threshold optical elements for high-energy laser devices (especially ICF), and ensures that the optical elements will not be damaged or fail under high-power laser irradiation. In this section, we introduce the principle of ultrafast pump–probe imaging and provide a detailed discussion on the two above-mentioned areas.

### 2.1 | Principle

A pump–probe imaging system consists of a delay stage, a laser source, a beam splitter, and an imaging detector, as shown in Figure 1. The pump and the probe branch are separated from the laser pulse by the beam splitter and irradiated to the area of the sample. In some applications, there are two lasers for pumping and probing, respectively, for example, a nanosecond (pump) and a picosecond (probe) laser. The pump beam is used to excite the sample and induce a dynamic phenomenon. Simultaneously, the probe pulse passes this interaction area and irradiates the detector, carrying the transient information. The delay between the pump and probe pulses can be adjusted by mechanical and electronic devices, which determine the various time slices of the total dynamic process.

Taking into account the transmission situation, the non-excited medium has an absorption coefficient $a_0$. Excited states mostly decay exponentially, and the absorption rate decreases to $a_0$ immediately after excitation according to

$$\Delta a(t) = \Delta a_0 e^{-t/\tau_\alpha}, \quad t > 0,$$

where $t$ is the time delay after excitation and $\tau_\alpha$ is the excited state lifetime. The light intensity $I(t)$ that changes with time satisfies the following equation:

$$I(t) = I(0)\left(1 + \Delta a_0 Le^{-t/\tau_\alpha}\right),$$

where $L$ is the light transmission length of the sample.

The relative change of the transmitted light intensity $\Delta T(t)$ and the time delay $t$ can be related by the following equation:

$$\frac{\Delta T(t)}{T_0} = \frac{I(t) - I(0)}{I(0)} = \Delta a_0 Le^{-t/\tau_\alpha}.$$

![Figure 1](image-url)  
Schematic diagram of the pump–probe imaging system.
The above calculation shows that the time-resolved information of the sample can be obtained by detecting the delayed intensity of laser beam at different times.

The pump–probe technique includes two core concepts. First, it maps the demand of temporal resolution to the demand of spatial resolution through spatio-temporal transformation technology, which ensures the femtosecond precision positioning capability at the beginning of the detection. Since the speed of light is constant, the propagation distance of light is proportional to the time, and the light path corresponding to a time of 1 fs is 0.3 μm. Although the existing mechanical and electronic equipment cannot distinguish the time of 1 fs, the spatial resolution of 1 μm is easy to achieve. The current capability of spatial resolution can be at the nanosecond level, corresponding to attosecond-level temporal resolution. Second, it uses ultra-short laser pulses instead of conventional continuous light to achieve an ultra-short shutter time of camera. In this case, the actual exposure time of the sensor is determined by the laser pulse duration. With the advent of the attosecond laser, the exposure time can theoretically reach the attosecond level. Therefore, the temporal resolution of the pump–probe technique could be thousands of times faster than the response time (picosecond level) of the fastest electronic equipment.

2.2 | Dynamics in ultra-short laser processing

While many theoretical studies have been focused on the simulation of the ablation dynamics of metals, the experimental study of ablation dynamics over a long time period remains essential for the in-depth understanding of the ablation mechanism. Pump–probe microscopy was widely used to observe the surface ablation dynamics on various metals such as Ti, Au, and aluminum. In addition, the lattice dynamics of Au and Ni irradiated by laser in the first ten picoseconds can also be captured by pump detection technology. The complete dynamics process of femtosecond laser ablation of two industrial metal samples (aluminum and stainless steel) was observed for the first time by Winter et al. The results on surface dynamics of Al and stainless steel, the Newton rings generated at the time delay of 100 ps and their expansion with time. A time resolution of 100 ps, observation time of up to 3 ns, and eight different visualization time ranges could be achieved. By choosing a high-power laser to ablate Al, the dynamic process of femtosecond laser ablation with an energy injection much higher than the ablation threshold was achieved, and a time-delay image from 50 fs to 10 ns was recorded. The shadowgraphs recorded by Zhang et al. provide for the first time a direct dynamic picture of the interesting hybrid ablation process in an intuitive way.

The Coulombic explosion is one possible mechanism when a high-energy laser pulse irradiates a dielectric or semiconductor material. In addition, after the target is heated by an ultrafast laser pulse, a strong thermoelastic wave may be formed due to the sudden thermal expansion of the ablated area. This thermoelastic shock wave may cause spalling or chipping inside the target material and eventually lead to matter ejection. Currently, research on the dynamic process of femtosecond laser-induced plasma generation is mainly carried out by single-pulse irradiation. However, the plasma dynamics during multi-pulse laser ablation remains unclear. Therefore, by studying the laser-induced plasma dynamics during the ablation process of multiple femtosecond laser pulses of silicon, the structure–mater excitation of air plasma was directly observed in the femtosecond time scale, revealing the mechanism of the plasma and shock wave expansion. The two basic mechanisms are of great significance for gaining an in-depth understanding of the nature of the interaction between ultrafast lasers and matter. Figure 3 shows the time-resolved shadow map of laser ablation of silicon on the femtosecond time scale. The first pulse irradiation cannot excite the transient phenomena. However, the narrow dark region appeared when the probe delay of 300 fs and the dark region increased with the delay of the probe in the opposite direction of the laser.

The ablation process by the ultrashort laser pulse is affected by transient dynamics. The ultrashort laser pulse has a rapid cooling rate of up to 10^12 K/s in the air. However, when performed in liquid, the ultrashort pulse ablation process became extremely complicated and the ablation rate may differ on the time scale of femtosecond to picosecond. Therefore, the ablation process of iron in different liquids on the femtosecond and picosecond time scales was also observed. The results show that the ablation process in air and liquid is similar in the first 10 ps of laser ablation; however, after 10 ps, there is a significant difference from the ablation process usually observed in air. At the same time, due to the influence of the liquid environment, the reflectivity does not decrease due to scattering and absorption, but increases, and strongly depends on the liquid used. The above results indicate the influence of the liquid environment on the ablation process during laser ablation.

![Figure 2](image-url)

**Figure 2** Time-resolved measurements of the surface dynamics from pump–probe reflectometry (PPR) of Al in (A) and stainless steel in (B). Reproduced with permission. Copyright 2020, Elsevier
The ICF devices, such as the National Ignition Facility (NIF), have a large demand for ultraprecision optical elements, which are vulnerable to damage and failure in high-power laser systems. Research on the damage mechanism of transparent optical elements continues to be an important scientific subject. The small cracks that form in the optical element during the manufacturing process are generally considered to be the cause of damage. TRPP with a time resolution of nanosecond can capture the temporal changes in laser-induced damage. The defects absorb the laser energy and cause localized temperature rise, leading to the formation of plasma at the interface between the material and air. The plasma expands and produces shock waves and stress waves. Eventually, the shock waves and stress waves cause circumferential and radial cracks, thus leading to the appearance of a pit structure. The dynamic process was recorded by TRPP, as shown in Figure 4. The transmission optics did not show obvious damage when the delay of laser was under 2.5 ns. However, as the time elapsed, the center of the transmission element showed a fuzzy structure and became a pit gradually.

Over a longer temporal scale, particle spray is formed from the damaged area. The particles are ejected during the laser-induced breakdown process. These particles cause adverse effects on the entire system, including a reduction in the mechanical properties of the deposited coating and aggravating laser-induced damage to optical components. Therefore, observing the path of ejected particles has become the focus of research. TRPP technology is the main method used to observe the behaviors of ejected particles with a time resolution of <0.5 μs. The ejection process of particles is irrelevant to the laser parameters and mainly depends on the volume of the particles excited by the laser. The particle residues forming the initial pulse could be excited by subsequent pulses. With an increase in the number of pulses, the distance of particle ejection increases. These results provide sufficient valuable information about the trajectory of the ejected particles that can give insights to prevent damage to adjacent optical components.
Since the 1970s, X-ray imaging technology has developed rapidly and has been widely used in industrial flaw detection, weld inspection, and medical inspection. With the development of image digitization technology, higher spatial resolution, wider dynamic range, and real-time imaging capabilities without geometric distortion can be guaranteed under a large field of view. The X-ray high-speed imaging system is commonly used now in the imaging inspection field because it can quickly and dynamically monitor the internal structure, size, position, and dynamic changes of the detected object. Currently, X-ray high-speed imaging is also widely applied for the high-speed monitoring of the dynamic process of laser–material interactions, including molten pool monitoring, plasma evolution, and myoglobin structural dynamics. This section reviews the research on the use of X-rays to monitor dynamic processes at high speeds when lasers interact with materials.

### 3.1 Principle

The X-ray is widely used in the imaging research of the internal structure of metal and high-density plasma due to its penetration characteristics. Observation of the dynamic process requires time-resolved X-ray imaging technology. Generally, continuous photography with a μs–ms frame interval can be realized using a long pulse or continuous X-ray light source, such as a synchrotron radiation light source, combined with shutter exposure time control of the X-ray camera, as shown in Figure 5A. Alternatively, a plasma source excited by an intense laser pulse can generate an X-ray pulse with an ultrafast temporal resolution based on mechanisms such as Compton scattering, wakefield acceleration, and bremsstrahlung, which can achieve fs-ps temporal resolution. Figure 5B shows a typical laser wakefield accelerator-driven bremsstrahlung X-ray source for advanced radiographic imaging. The former method has been widely used in the field of laser processing and the latter ones are often used to capture ultrafast dynamic processes in the strong field physics of laser–matter interaction because it is convenient to realize time synchronization through the trigger of a laser pulse. However, due to the low repetition rate of intense laser pulses, it is often only possible to capture a single frame image.

### 3.2 Molten pool monitoring for laser processing

A critical research topic in high-power laser material processing is the evolution of the molten pool. The morphology and temperature of the molten pool have a huge influence on the dimensional accuracy, residual stress, and structural performance of the final component or system, and it is an object of significant monitoring importance. X-ray high-speed imaging plays a crucial role in molten pool monitoring.

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**Figure 4** Side-view transient images of the damage pit and temporal evolution of the damage process generated at 95 J/cm². The diameter of the final pit is 34 μm, and the depth is approximately 11 μm. Reproduced with permission. Copyright 2016, The Optical Society

**Figure 5** Schematic diagram of the X-ray imaging principle. (A) laser plasma. Reproduced under terms of the CC-BY license. Copyright 2017, The Authors, published by Springer Nature. (B) Synchrotron radiation source. Reproduced under terms of the CC-BY license. Copyright 2020, The Authors, published by IOP
As an important laser processing technology, additive manufacturing (AM) has gradually shifted its research focus to online monitoring of the melting process. The vapor depression (also known as keyhole) phenomenon is always an attractive problem in the metal laser melting processing field, and can be quantified using the ultra-high-speed synchrotron radiation X-ray imaging.\(^7\) Two different molten pool shapes were observed using this method, as shown in Figure 6. The results explained the previous uncovering of the change from the conduction mode to the keyhole mode: (i) the keyholes appeared in laser powder bed fusion within the range of the laser processing parameters used and (ii) according to the laser power density, there is an accurately defined threshold from the conduction mode to keyhole and the process follows the order of vaporization, sinking of the liquid surface, and deep and unstable keyhole formation.\(^7\)

Martin et al. revealed the dynamics of entrapped pores deep within the material at the liquid–solid interface of the molten pool and reported that Marangoni convection drives entrainment of pores in the molten pool; they also observed the oscillation of the pores in the molten pool with a time resolution of 10 µs.\(^7\)

The research on the microdynamics of the molten pool in the AM process has been the focus of many investigations.\(^7\) The control mechanism of directional energy deposition additive manufacturing (DED-AM) was found by in situ and operational synchrotron radiation X-ray imaging and diffraction of nickel based superalloy IN718. The spatial imaging can quantify the molten pool boundary and flow dynamics in the solidification process utilizing the unique DED-AM process replicator. The spatial resolution accurately diffracts the time-resolved microstructure phase, and the rapid cooling rate completely inhibits the formation or recrystallization of the second phase in the solid phase. After solidification, the stress increases rapidly to the yield strength for cooling, which shows that, combined with the solidification crystallization range of IN718, the accumulated plasticity depletes the ductility of the alloy, resulting in liquefaction cracking. The change in shape of the molten pool photographed by X-ray is shown in Figure 7. This study reveals the mechanism controlling the formation of a highly nonequilibrium microstructure during DED-AM. During the whole period, the spatial resolution was approximately 100 µm, and the temporal resolution was less than 1 ms.\(^8\)

The flow evolution of the laser molten pool under gravity, surface tension, the Marangoni effect, and steam pressure is a complex process. Zhao et al.\(^6\) used laser powder bed fusion (LPBF) to process Ti-6Al-4V and other powders to observe the changing process of molten pool dynamics with the highest imaging recording rate in AM of 10 MHz. They observed the development of molten pool geometry under different laser powers and scanning speed conditions, such as depth, aspect ratio (depth/width), and nominal area, as shown in Figure 8. Simultaneously, they observed a change from a higher growth rate to a lower rate during the growth process of the molten pool, indicating that the strong and complex flow of molten metal in the later stage of laser heating tends to relax the sample temperature and maintain a relatively stable molten pool distribution. The time resolution of X-ray imaging is less than 22 µs. The phenomenon and mechanism of powder motion behavior changed with time, and ambient pressure in the process of laser melting was demonstrated in the schematic diagram of powder motion, as shown in Figure 9.\(^8\) Recently, they revealed four types of pore formation mechanisms in the DED-AM process, based on the in situ high-speed high-resolution X-ray imaging technique as shown in Figure 10.\(^8\)

The corresponding system for on-site monitoring of laser welding was developed by integrating high-speed X-ray imaging, an acoustic sensor, and machine learning. High-speed X-ray imaging was used to associate acoustic emission (AE) signals with actual processing events. Combined with the machine learning algorithm, the classification accuracy of images ranged from 74% to 95%. The effective spatial resolution of the X-ray image detector can reach 11 µm on combining AE with ML (machine learning), as shown in Figure 11, which has a broad application potential in field and real-time monitoring of the laser welding process.\(^9\)

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**FIGURE 6** Evolution of melt pool and vapor depression under stationary laser illumination. (A) Initial formation of a melt pool. (B) Formation of a small, stable vapor depression. (C) Steady growth of the vapor depression. (D) Instabilities form in the vapor depression. (E,F) Rapid change in the vapor depression shape. (G,H) Periodic fluctuation of the vapor depression. (I,J) Change of the melt pool shape from quasisemicircular to bimodal. Reproduced with permission.\(^7\) Copyright 2019, AAAS
FIGURE 7  Schematic diagram of the experimental methods and results. (A) Powder additive manufacturing process replicator, designed to reproduce the operation of commercial DED-AM systems. (B) Schematic diagram of field X-ray imaging of the DED-AM process. (C) IN718 multilayer thin-wall melt trajectory. Reproduced with permission.80 Copyright 2021, Elsevier

FIGURE 8  Experimental principle of molten pool shooting. (A) Dynamic evolution of the melt pool in laser powder bed fusion processes of Ti-6Al-4V. Reproduced under terms of the CC-BY license.61 Copyright 2017, The Authors, published by Springer Nature. (B) Laser AM process for the Al-Si10-Mg plate. Reproduced under terms of the CC-BY license.81 Copyright 2018, The Authors, published by International Union of Crystallography
3.3 Ultrafast imaging for intense laser plasma physics

In laser-driven ICF, asymmetry of the hot spot and the mixing effect of the shell material into the hot spot remain important factors restricting the further increase of the ICF implosion yield. Capturing an image of the hot spot with high temporal and spatial resolution is essential to analyze the asymmetry of the hot spot shape and the physical design of the symmetry control of the hot spot. The X-ray high-speed microscopic imaging system based on Kirkpatrick–Baez (KB) is widely applied in the field of ICF. A variety of KB microscopic imaging systems have been developed, such as the total reflection wide-band KB microscopic imaging system, the multi-layer film quasi-single energy response KB microscopic imaging system, and the multi-mirror structure advanced KB microscopic imaging system. The above imaging system has a large field of view (~1 mm), high spatial resolution (<5 μm), and high temporal resolution (<100 ps), as shown in Figure 12A,B. It has been used to observe the process of CH shell implosion, black cavity implosion at both ends, and glass shell implosion to push the target. Some of the experimental results are shown in Figure 12B.

X-ray framing recording technology is equipped with simple pinhole imaging or KB microscopic imaging or curved crystal imaging with high spatial resolution imaging capabilities, which can achieve a two-dimensional high-spatial resolution imaging diagnosis to observe the evolution of the hot spot morphology during implosion. To further improve the imaging time-resolution capability, an X-ray drift imaging system was developed by combining electronic drift technology. By accelerating the photoelectron in the drift zone, the "velocity dispersion" of the electron group was accomplished. The principle and schematic of the system are shown in Figure 13. The pulse was broadened by 20 times the width, and the time resolution was increased from 60 to 20 ps, with high spatiotemporal resolution diagnostic capabilities. This technique has been used in the research of hot spot morphology evolution, fuel movement state, and the degree of hot spot mixing. As the charge effect increases, the noise increases, and the signal-to-noise ratio of the acquired image is lower, which affects the spatial resolution of the image.

4 SINGLE-SHOT ULTRAFAST OPTICAL IMAGING

Conventionally, the pump–probe methods have allowed capture of the dynamics through repeated measurements. However, many ultrafast phenomena are either nonrepeatable or difficult to reproduce such as laser-induced shock waves, microfluidics, and photochemical reactions. Under these circumstances, single-shot ultrafast optical imaging techniques become necessary to overcome the limitations of pump–probe. Various ultrafast single-shot imaging schemes have been developed for capturing the two-dimensional images of nonrepetitive ultrafast phenomena in a time scale less than nanosecond. In this review, we mainly focus on the active illumination ultrafast imaging techniques that require an ultra-shot illuminating
FIGURE 10  Pores transferred from the feedstock powder to the build. (A–H) The sequence of X-ray images showing that the pores inside the feedstock Ti64 powder are transferred to the melt pool after the melting of the delivered powder in the DED process. Reproduced with permission. Copyright 2021, Elsevier

FIGURE 11  Real-time X-ray images of (A) Conduction welding, (B) stable keyhole; (C) unstable keyhole, and (D) spatter. Reproduced with permission. Copyright 2018, Elsevier
laser pulse in the system, while ignoring the receiving imaging methods. The latter methods do not need illumination light and can capture the self-luminescence phenomenon, such as the compressed ultrafast photography (CUP).95–97 Typical active single-shot ultrafast optical imaging methods include femtosecond time-resolved optical polarimetry (FTOP), frequency-domain holography (FDH), frequency-domain tomography (FDT), time-resolved holographic polarization microscopy (THPM), sequentially timed all-optical mapping photography (STAMP), and frequency recognition algorithm for multiple exposures imaging (FRAME).

### 4.1 Single-shot femtosecond time-resolved optical polarimetry

The main features of the FTOP are the optical polarigraphy technique. The ultrafast dynamics of the intense femtosecond laser pulses propagation was captured with the FTOP.98–102 Figure 14A shows the experimental schematic diagram. The laser beam is split into a pump and a probe pulse by a beam splitter. The probe pulse is frequency-doubled by a crystal, and is divided spatially into four daughter pulses by a four-step echelon. The pump pulse beam is focused into a fused silica cuvette filled with the target of CS₂, after passing through a delay line and a half-wave plate. The recorded FTOP images of the pump-pulse propagation in CS₂ are shown in Figure 14B. The frame interval was approximately 0.96 ps, corresponding to a frame rate of approximately 1.05 THz. FTOP uses the width of laser pulse to achieve high temporal resolution, but the number of frames and imaging field of view are limited, and can only photograph the ultrafast phenomenon with nonoverlapping trajectories.

### 4.2 Single-shot frequency-domain holography

Single-shot frequency-domain holography (FDH) is a single-shot ultrafast phase measurement technique with one-dimensional spatial resolution, developed based on an extension of the frequency-domain interferometry (FDI) method.103,104 The advantage of FDH is the single-shot measurement of optical phase shifts with femtosecond temporal resolution over a picosecond time scale. Figure 15 shows the original system schematic of FDH. A temporally stretched...
probe-reference pulse was used and covered the whole object, accumulating phase shift given by

$$\Delta \Phi_{pr}(r, \xi) = \frac{2\pi}{\lambda_{pr}} \int_0^r [1 - \eta(r, \xi, z)] dz,$$

where $\eta(r, \xi, z)$ is the pump-induced refractive index at a single time delay $\xi$; $r$ and $z$ denote the transverse and axial distances from the pump propagation, respectively; and $\lambda_{pr}$ is the wavelength of the probe pulse. A main contribution of FDH method is the single-shot visualization demonstration of laser-wake-field accelerator structures, capturing the evolution of multiple wake periods for the first time. In their experiments, two chirped and frequency-doubled pulses took holographic snapshots of the ionization front and wake, which was created by an energy of 1 J and a 30 fs pump pulse. The spatial resolution was close to 3 \( \mu \)m, and the temporal resolution was 30 fs.

4.3 | Single-shot FDT

Tomography imaging is based on the measurement of radiation transmitted through an object along different directions, which enables noninvasive imaging of hidden stationary objects from sequentially measured projections. FDT can image a wide range of nonlinear propagation phenomena, including filament formation in gases and the evolution of plasma wakefields. Figure 17A shows schematic representation of single-shot FDT. Tomographic methods are adapted to visualize the instantaneous structure and evolution of a laser-induced object propagating through a transparent Kerr medium. Due to the nonlinear refractive index dependence and the plasma generated by the intense pump pulse, the transient refractive index structure changes in fused silica glass at the speed of light. A three-layer structure BBO (\( \beta \)-barium borate) crystal was used to generate 15 frequency-doubled pulses, and among these, 5 pulses with different projection angles were selected as the probe pulses, as shown in Figure 16A. Then, the probe pulses overlap spatially and temporally at the target location and the dynamic process is measured by a spectral imaging interferometer. Snapshots of animation of nonlinear laser propagation in a fused silica glass are shown in Figure 16B. There constructed snapshots have revealed the dynamics of self-focusing within 7.4 ps and laser filamentation at 9.8 ps. A steep-walled index hole generated at 12.2 ps indicates that the plasma had induced a negative index change and offset the laser-induced positive nonlinear refractive index change.

4.4 | THPM

According to the angular multiplexing holographic technique, THPM can capture ultrafast phenomena that appeared in polarization-sensitive transparent materials. Figure 17A shows schematic representation of the THPM system. A frequency-doubled probe laser pulse was separated into two pulses as a pump and a probe, respectively. The interference holograms between the reference and signal pulses were captured by a CCD camera, which
loaded different spatial carrier frequencies by different spatial filters. Then, the phase and amplitude distributions of two orthogonal polarization components could be reconstructed by an imaging algorithm. The THPM technique can capture the amplitude and phase distributions of two orthogonal polarization states of an ultrafast dynamics at two different times with a one-shot measurement. The main principle of STAMP is to map the temporal and spatial information into a linear chirped laser pulse by using a temporal mapping device (TMD) and a spatial mapping device (SMD). The TMD allows different snapshots of different frequency components, and the SMD separates these snapshots to be recorded by one camera. The frame number of STAMP is determined by the separable spectral component. In the original research of STAMP, the dynamics of lattice vibrational waves induced by the femtosecond laser pulse was photographed at a frame rate of 4.4 trillion frames per second. However, only six frames were captured. Various optical structures have been constructed to increase the frame number. The 5- and 25-frame burst imaging by utilizing spectral filtering STAMP (SF-STAMP) was realized, in which their 4f imaging configuration consists of a diffractive optical element (DOE) and a band-pass filter (BPF). The ultrafast 2D-burst images of the crystalline-to-amorphous transition.
phase transition of Ge$_2$Sb$_2$Te$_5$ (GST) with a sub-picosecond temporal resolution were captured, as shown in Figure 18B. Compared with the periscope array, DOE can achieve a larger frame number while the manufacturing is still complex. Besides, a branched 4f system with a slicing mirror as the spectrum slicer for STAMP was developed to increase the number of frames while preserving its pixel resolution. Saiki et al. demonstrated a STAMP system using a slicing mirror and a branched 4f configuration, which achieved 18-frame imaging in a 3 × 3 manner with two image sensors.

4.6 | FRAME

FRAME is another spatial frequency division technique. The ultrafast 2D videography was first realized by FRAME with spectral compatibility, high temporal, and spatial resolution. In FRAME, the time scale of the frame interval can reach the femtosecond level. Figure 19A shows the schematic diagram of FRAME. FRAME encodes different carrier frequencies to detect daughter pulses by intensity modulation achieved with a Ronchi grating, and then deciphered in a post-processing step.
To demonstrate this capability, the diverging laser pump pulse propagated in the Kerr medium was reconstructed.\textsuperscript{122} As shown in Figure 19C, the FRAME imaging system monitored the dynamic process in which the laser beam passes through CS\textsubscript{2} liquid. The frame size was 1002 pixel × 1004 pixel, and the imaging speed was 5 Tfps. The final spatial resolution of FRAME was approximately 15 lp/mm, and the field of view exceeds 7 × 7 mm. By using 3D interpolation method, Figure 19D shows how the wave front of pump pulse changes when propagating through the CS\textsubscript{2} liquid.\textsuperscript{118}

## SUMMARY AND OUTLOOK

In summary, the progress of laser technology has led to unprecedented high-energy-density physical phenomena and new theoretical mechanisms, enabled the progress of ICF, laser accelerators, laser advanced manufacturing, and other fields, and made the theoretical research on the interaction between high-power laser and material increasingly attractive. A variety of ultrafast imaging technologies for different applications are being rapidly developed to help researchers observe and understand these transient phenomena. The focus of this paper was on reviewing the principles and applications of three types of commonly used ultrafast imaging methods including pump–probe, X-ray diagnosis technique, and single-shot optical burst imaging. The process above the damage threshold was the main focus. Each ultrafast imaging technique has its own advantages and limitations. For example, the penetrability of X-ray contributes to the internal imaging of plasma, but, for the most part, only one single frame image can be captured for ultrafast photography and the equipment is complex and expensive. Visible light photography based on a femtosecond laser is relatively convenient to implement under ordinary laboratory conditions, but the signal is easily disturbed and distorted in a complex electromagnetic environment.

Several representative ultrafast imaging technologies mentioned in this review are summarized in Table 1 in terms of classification, temporal resolution, and applications.

This review serves as a reference for researchers in related fields, despite difficulties in covering all the ultrafast imaging technologies. It is expected that the single-shot burst imaging technique, such as FTOP and STAMP, will become a reliable measurement method, while there are still some parameters, such as the number of frames, that need to be improved. It is also noteworthy that the micro- and nano-structures in MEMS (Micro-Electro-Mechanical System) and NEMS (Nano-Electromechanical System), such as high-frequency resonators in 5G and 6G, micro/nano beams, which may be used for biological detection, will also utilize the ultra-fast imaging of both the motion and internal structures and defects, which will be worthy of further study.

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### CONFLICT OF INTEREST

The authors declare no conflict of interest.

### DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

### ORCID

Du Wang http://orcid.org/0000-0002-0098-7613
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