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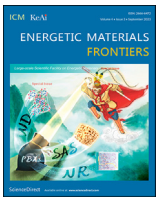

Recent progress in research on the dynamic process of high-energy explosives through pump-probe experiments at high-intensity laser facilities

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ABSTRACT

To accurately predict the detonation and safety performances of high-energy explosives, it is necessary to investigate their reaction mechanisms on different scales, which, however, presents a challenge due to the complex reaction kinetics of the explosives and a lack of experimental methods presently. This work introduces the time-resolved pump-probe experiments capabilities aiming at high-energy explosives based on large-scale laser facilities and presents the recent progress in research on the dynamic process of the explosives, obtaining the following understandings: (1) First, the micron-sized single-crystal 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) can be compressed to an overdriven detonation state at a laser facility, followed by the shock Hugoniot measurements of TATB; (2) Second, high resolution transient X-ray radiography makes it possible to achieve the dynamic imaging of the internal deformation, damage, and reaction dynamics of high-energy explosive under dynamic loading; (3) Third, the phase transformation and chemical reaction products of the shock-compressed explosives can be investigated using dynamic X-ray diffraction or scattering spectra; (4) Finally, the structural changes, molecular reactions, molecular bond cleavage, and intermediate product components of explosives under ultrafast pumping can be explored using ultrafast laser spectroscopy. Large-scale laser facilities can provide various flexible pump-probe methods, including laser shock loading, transient X-ray imaging, dynamic X-ray diffraction, and ultrafast spectroscopy, allowing a series of experiments to be carried out to evaluate different levels of ignitions from low-pressure to overdriven detonations. Furthermore, the facilities also enable *in situ*, real-time investigations of the internal deformation, phase transition, and ultrafast dynamics of explosives under dynamic loading at high spatial and temporal resolutions. The study of the reaction kinetics and mechanisms of high-energy explosives on microscopic-mesoscopic scales provides an efficient means to unravel the mystery of explosive reactions.

1. 1. Introduction

High-energy explosives, as one of the most important materials, release initial energy in an ordnance system. Their detonation and safety performances are the decisive factors affecting their damage effect, reliability, and safety. The widespread applications of condensed phase explosives make research on these performances increasingly important. The reaction kinetics of high-energy explosives is a critical factor that determines the initiation performance and safety of the explosives. The energetics of reaction kinetics involves a complex system consisting of intra- and inter-molecular bonds, numerous morphologies, different polymorphs, crystal defects, impurities, and compositional inhomogeneity.[1–3] To accurately interpret the detonation and safety performances of explosives, it is necessary to determine the reaction mechanisms and dynamic process of explosives on different scales,[4] which, however, poses a challenge due to the complex reaction kinetics of explosive and a lack of experimental methods.[5–8] On the one hand, under high-temperatures and high-pressure conditions, extremely fast and intense photophysical and photochemical processes occur in very thin reaction zones (micrometers to nanometers) and on ultrafast time

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scales (nanoseconds to femtoseconds). On the other hand, an entire detonation can be decomposed into several key steps, such as energy injection, energy coupling, electronic excitation, molecular excitation, structural phase transition, reaction growth, and product formation.[9] In recent years, the reaction kinetics of explosives have been explored using theoretical modeling and numerical simulations.[9] There is an urgent need to elucidate the inherent reaction mechanisms of explosives under dynamic loading and to verify the models and simulations through high-precision experiments.

The reaction kinetics of a detonation process results from the coupling of chemical reactions and shock waves. Microscopic and mesoscopic experimental research is a key means to solve the mystery of explosive reactions.[10] The mesoscopic research is generally associated with the propagation of the shock and detonation waves induced by an explosive column. However, it is yet to ascertain the mechanisms of hot spot initiation,[11,12] in which the sensitivity and chemical reaction rate of explosives are directly affected by factors such as cracks, pores, and crystal-binder interface.[13–16] Therefore, it is crucial to perform an *in situ* investigation of the detonation process using dynamic technology. In addition, the phase transition kinetics, reaction product composition, and other related issues should be further studied using X-ray diffraction or scattering spectra. Regarding the microscopic level, many scientists have always believed that subtle understanding at a molecular level is essential for predicting the behavior of explosives and evaluating their performance.[17] It has been recognized that the non-equilibrium state of explosive molecules in the transient detonation state plays a key role in the energy conversion process.[18] Dynamic technology is essential for research on nanoseconds or even femtosecond scales. Moreover, advanced X-ray radiography, X-ray diffraction, and small-angle X-ray scattering (SAXS) have been developed using the high-energy synchrotron sources or free electron laser facilities, such as the Advanced Photon Source (APS) at Argonne National Laboratory,[19,20] which provide dynamic methods for the *in situ* investigation of detonations. However, these facilities are very large and too infrequent to accommodate more than a small fraction of experiments. As a result, new experimental research on the microscopic and mesoscopic scales is still under development.

A high-intensity laser facility exhibits unique advantages and great potential for research on the ultrafast reaction kinetics of explosives for the following reasons: (1) The time scale of ultrafast reaction kinetics, which is in the order of nanoseconds or femtoseconds, is equivalent to the pulse duration of ultrafast laser. Therefore, ultrafast laser technology becomes an effective experimental tool for studying the reaction kinetics of explosives under impacts, making it possible to image these reactions; (2) A large-scale laser facility can load an extremely high temperature and high pressure on a sample. Moreover, it can generate ultra-short and highly intense X-ray beams and particle beams in backlight photography and diffraction experiments; (3) Laser-driven experiments are more competitive for small targets and feature high accuracy and serial loading pressures, providing valuable insights into the dynamic behavior of energetic materials. The pump-probe experiments are complex and challenging and should be carefully designed to minimize noise and ensure accurate measurements. Based on high-intensity laser facilities, experimental platforms that integrate wave-profile diagnostics, laser spectroscopy, transient X-ray imaging, diffraction, and electronic imaging technologies have been developed.[21–24] The experimental study of ultrafast reaction kinetics can be achieved using nanosecond-scale laser pumps and ultrafast laser probes, making it possible to investigate the crystal phase transition dynamics,[24] hot spot formation and evolution law,[25] short-pulse initiation mechanisms,[9] and product components of explosives.[26]

The purpose of this study is to present recent advances, dynamic capabilities, and potential applications of research on explosives through pump-probe experiments at large-scale laser facilities. First, this study introduces the levels of ignition from low-pressure to overdriven detonation at a large-scale laser facility and presents the shock Hugoniot measurement under overdriven detonation. The average velocity of detonation waves is also promoted at a loading pressure of above 40 GPa. Second, the internal deformation, damage effect, and reaction dynamics under dynamic loading can be imaged *in situ* in real time using a high-intensity short-pulse laser. The high-resolution dynamic X-ray imaging is exhibited, providing a perspective view of the flyer loading and structure of the detonation waves. Third, the phase transition and product formation in the process of explosive reaction is further understood using X-ray diffraction or scattering spectra. Moreover, the crystal structure under dynamic loading is investigated using dynamic X-ray spectroscopy based on a bright monochromatic X-ray source of a high-intensity laser facility. Finally, the excited-state dynamics and up-pumping mechanisms of explosives are studied using ultrafast spectroscopy. Insights into the structural, geometrical, and chemical changes in excitation, which are essential for the study of ultrafast and complex reactions.

2. Worldwide large-scale laser facilities and the pump-probe technique

A high-intensity laser facility provides a powerful pump-probe capability for studying the material structure and properties of materials under extreme conditions.[27] There is an increasing need to study the dynamic process through pump-probe experiments at large-scale laser facilities. To date, many large-scale laser facilities have been built around the world, including the National Ignition Facility (NIF), OMEGA EP, Titan, Vulcan, and AWE.[28] Among them, the NIF at the Lawrence Livermore National Laboratory (LLNL) is the first megajoule-scale facility and is also the only megajoule-scale facility that is fully operational currently.[28,29] It has 192 40 cm × 40 cm beams that are capable of delivering a total of 1.8 MJ initially in a ~3ns shaped pulse @3w, 0.6 PW [a true petawatt (PW)-class laser in its own right, albeit delivered in multiple beamlines] configured for indirect beam drive. The advanced radiographic capability (ARC) at the NIF is designed as an advanced X-ray radiographic capability for NIF. The ARC at NIF employs one fourth (a quad) of NIF's beams to achieve tens of picoseconds of temporal resolution and has been operational since 2015. Each beam is split into two, resulting in eight PW-class beams, which are capable of delivering 0.4–1.7 kJ at pulse lengths of 1.3–38 ps (0.5 PW each) in the infrared. ARC has contributed greatly to kJ-class short-pulse lasers, such as the development of high-efficiency meter-scale dielectric gratings, single-shot precision diagnostics, and dispersion management. OMEGA-EP is a four-beam system with an architecture very similar to that of the NIF, coupled to a 60-beam long-pulse OMEGA laser system with a proven deliverable energy of 30 kJ at 351 nm.[28] Two of the EP beams can operate in the short-pulse mode, in which an optical parametric chirped-pulse amplification (OPCPA) front end is used to add high-energy, PW-class laser power to provide X-ray backlighting and proton radiography capabilities for inertial confinement fusion (ICF) experiments. The short-pulse laser, which operates at pulse widths of 0.6–100 ps, can deliver nearly 1 PW of power under the best compression conditions and energy ranging from 1.25 kJ to 2.3 kJ at pulse widths >10 ps. Titan is one of five lasers that make up the Jupiter Laser Facility at LLNL.[28] It is a PW-class laser coupled to a kilojoule beamline for a wide range of experiments. The short-pulse beamline delivers up to 300 J at a sub-picosecond pulse and offers a 50 J high-contrast green option. It is currently being upgraded for higher peak powers, and a third beamline is being added to it. Vulcan at the STFC Rutherford Appleton Laboratory is the first PW-class laser that can be used by the international plasma physics community as a dedicated user facility.[28] It is a high-power Nd:glass laser that has been in operation for over 40 years, and its flexible geometry allows for a wide range of experiments. It has two target areas: one with six 300 J (1053 nm at 1 ns) long-pulse beamlines combined with two synchronized short-pulse beams and a separate target area with high-energy PW capability (500 J in 500 fs) synchronized with a single long-pulse beamline. AWE is a Nd:glass laser system that

combines 10 long-pulse beamlines (500 J, 1ns@351 nm) with two synchronized infrared PW beams (500 J in 500fs).[28]

In China, typical large solid-state laser facilities suitable for pump-probe experiments include the Shenguang-II upgrade (SG-II-U) laser facility, the XingGuang-III laser facility, and the ten-thousand-Joule laser facility.[28,30] The SG-II-U laser facility, a technical integration line consisting of 8 ns laser beams and 1 ps beam, is part of the development of a high-power solid-state laser for related laser fusion and strong-field studies.[23] The eight beams can generate a total of 24 kJ at 3 W, and the ninth beam can generate 500 J at 10 ps. The Xingguang-III high--intensity laser facility produces femtosecond, picosecond, and nanosecond beams with wavelengths of 800 nm, 1053 nm, and 527 nm, respectively.[31] Using an optical synchronization technique that combines supercontinuum generation with femtosecond optical parametric amplifiers (OPAs), the Xingguang-III laser can generate beams from the same source to achieve precise synchronization. Two short-pulse laser beams, namely femtosecond and picosecond beams, make it possible to probe dynamic response information about materials in two spatial dimensions and on different time scales. These features make the Xingguang-III laser facility a unique system for pump-probe experiments. It creates extraordinary conditions for physics research and greatly enriches the categories of experiments. The ten-thousand-Joule laser facility in Mianyang City consists of 8 ns beams and the ninth beam with a tunable pulse width from several subnanoseconds to 20 ns.[32,33] The eight laser beams have a wavelength of 351 nm and a quasi-square-shaped temporal profile with a rise time of 200 ps and an FWHM time of 3 ns. Each of the beams is smoothed using a continuum phase plate (CPP) with a maximum energy of 800 J. It irradiates the target at an incident angle of 45°, producing a uniform focal spot diameter of 2 mm. The ninth beam can provide a peak energy of 3000 J and is suitable for X-ray generation.

A large-scale laser facility can provide strong pump-probe capabilities for research on materials under extreme conditions, as shown schematically in Fig. 1. Specifically, laser “pumps” and laser “probes” are used to study the structure and properties of materials under high pressure.[21,23,24,27,29,32,34–37] In general, a laser "pump" refers to the high pressure and high density in the target driven by one high-intensity laser, while the laser “probe” usually refers to the high-precision diagnosis using bright X-rays or photons driven by the other high-intensity laser. There are several ways to achieve the high-pressure and high-density state of the target using a laser “pump”, where the laser energy is effectively converted into the internal energy of the material *via* ablation. Direct drive and indirect drive are two main methods that differ in the way that the material is ablated. Direct drive is achieved by laser ablation on the sample or an ablation layer, allowing shock waves to be generated by rapid plasma expansion.[23,32,37] The loading pressure, wave profile, and loading area of the shock waves are controlled by the laser intensity, pulse shape, and focal spot, respectively. For instance, triangular shock waves can be generated when a square-shaped laser is adopted, and the peak loading pressure can be adjusted by the laser intensity.[23] Laser ramp wave loading is always performed by plasma shock or temporally ramped pulse ablation, where the laser shape is modulated to a relatively long and special shape.[32] In addition, a laser-driven flyer can be launched by laser ablation or plasma colliding. This loading approach eliminates the need for additional preheating, thus producing a clean shock within target materials.[37] In short, the direct drive includes methods of direct laser shock loading, laser-driven flyer loading, and laser ramp-wave loading, as mentioned above. The indirect drive is achieved as follows: firstly, an Au cavity is irradiated using a laser to generate thermal X-rays, and then a sample is ablated by the X-rays to generate shock waves.[27,36] The laser irradiation of the Au cavity produces a spatially uniform distribution of thermal X-rays with a characteristic radiation temperature. The X-rays generated by the cavity ablate the ablation layer on the target surface and create a time-dependent, ramp-compression wave that propagates through the sample. Indirect loading is used in the case of ramped pulse ablation, where a designed composite ramped laser pulse shape with a long pulse width and high energy is used.

A laser “probe” provides high-precision parameter diagnosis of the high-pressure and high-density state of the target.[23,24,37,38] The techniques involved include transient X-ray spectroscopy, X-ray imaging, wave profile diagnosis, and optical spectroscopy. X-ray spectroscopy and X-ray imaging are the most important techniques for a laser “probe”. X-rays are generated by the interaction of a high-intensity laser and materials, in which the laser energy is effectively converted into X-ray energy. There are several mechanisms for generating X-rays from a laser, which are briefly stated as follows: (1) X-rays are generated by a nanosecond laser, primarily involving He_α emission and low-energy radiation.[29] This mechanism has a particularly high conversion efficiency of up to 10^{-2} and a particularly high photon yield for X-ray diffraction and imaging, with the photon energy typically less than 10 keV; (2) X-rays are generated by a picosecond laser, primarily involving K_α emission and bremsstrahlung radiation, in which electrons can be accelerated to an energy of MeV.[23,37,38] In this case, X-ray photons with energy greater than 10 keV can be efficiently produced, with a conversion efficiency of typically 10^{-4}–10^{-5} when the target is limited to a smaller size. The diameter of the source can be limited to about 10 μm, which can be used for high-resolution 2D radiography in a single-shot experiment; (3) X-rays can be generated from the betatron radiation when a femtosecond laser interacts with a gas target. In this case, the diameter of the source can be limited to several microns, which is suitable for high-resolution X-ray radiography, and the divergence can be several mrad. In

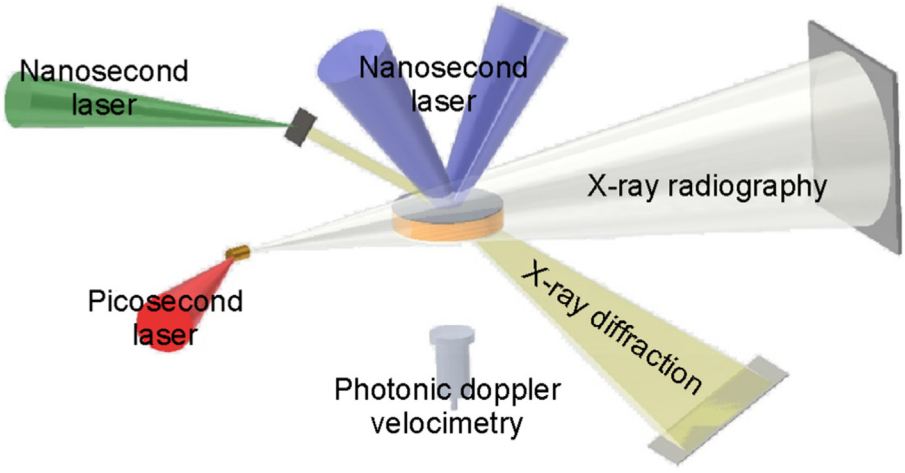

Fig. 1. Schematic view of the pump-probe capability of a high intensity facility, integrating direct laser shock loading, transient X-ray spectroscopy, X-ray radiography, and wave profile diagnostics.

particular, various X-ray sources can be used as laser "probes" in the pump-probe experiments for the study of explosives under extreme conditions. In addition, the Velocity Interferometer System for Any Reflector (VISAR) or Photonic Doppler Velocimetry (PDV) with an incident visible light can provide high-precision diagnostics of the interface velocity after the laser "pumping".[37] Optical imaging and spectroscopy are also widely used in pump-probe experiments, such as fast visible-light framing photography, gated infrared (IR) photography, time-resolved pyrometry, and optical emission spectrometers.[39]

3. Ignition levels from low-pressure ignition to overdriven detonation

The microstructures of explosives, such as different shapes, crystal defects, impurities, and compositional inhomogeneities, are usually on a micron scale. The reaction zone of an explosive that contains chemical reactions and shock waves is also very thin.[40] Research on the microstructural effect and reaction zone of an explosive under dynamic loading is critical to understanding the ignition of explosives under different conditions. However, it is inconvenient to perform such experiments on a large sample using conventional loading techniques such as light gas guns, explosive detonation, and magnetically driven loading experiments.[41] Explosives usually detonate at a low pressure, and the long distance of shock-to-detonation transition under low shock pressures brings challenges to the investigation of meso-scale dynamics driven by void collapse, hotspot formation, growth, and interaction. Experiments at high-intensity laser facilities are suitable for this case. Firstly, the loaded sample can be made very small, and the thickness of the sample can be only tens of microns. In addition, the loading pressure can be adjusted widely from a low-pressure ignition state to an overdriven detonation state, which can minimize the shock-to-detonation distance.[42]

Overdriven detonation is commonly achieved through high-speed plate impact, Mach-reflection detonation, and pool detonation, and the detonation behavior is different from that of low-pressure initiation.[43] An explosive can be overdriven by direct laser shock loading, and the equations of state for the explosive under the overdriven detonation can be studied via experiments at a high-intensity laser facility. Marshall et al. studied the shock Hugoniot measurements of single-crystal 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) that was compressed up to 83 GPa using direct laser shock loading.[42] The single-crystal TATB Hugoniot data showed significantly lower pressures than previous results on overdriven TATB formulations, as shown in Fig. 2, suggesting that single-crystal samples remained largely unreacted below 35 GPa over the short nanosecond-time scales inherent in laser-driven experiments.[42] The experiments on the overdriven high-energy explosive in a thin sample can also be performed at a high-power laser facility, such as the ten-thousand-Joule laser facility.

Short-pulse initiation is typically accomplished by exploding foil initiation and laser-driven flyers, among others. Laser-driven flyer initiation does not require additional preheating, thus producing a clean shock within the target materials.[44] Compared to gas guns and Z machines, the laser-driven flyer features a smaller size, a lower cost, and less destructive experiments. In addition, the laser-driven flyer, whose size is close to that of the plastic flyer in an exploding foil initiator, can be considered short-pulse initiation.[45–48] Studies on explosive detonation using minimal laser-driven flyers, which are attached to a transparent window, have been widely conducted since the flyers are strongly immune to electromagnetic disturbance and generate intense and short-pulse pressure pulse. Recently, Shui et al. conducted several experiments on laser-driven flyers at the ten-thousand-Joule laser facility.[33] In contrast to conventional flyers, which are launched by the expansion of plasma generated at the flyer-glue interface,[49] hypervelocity flyers

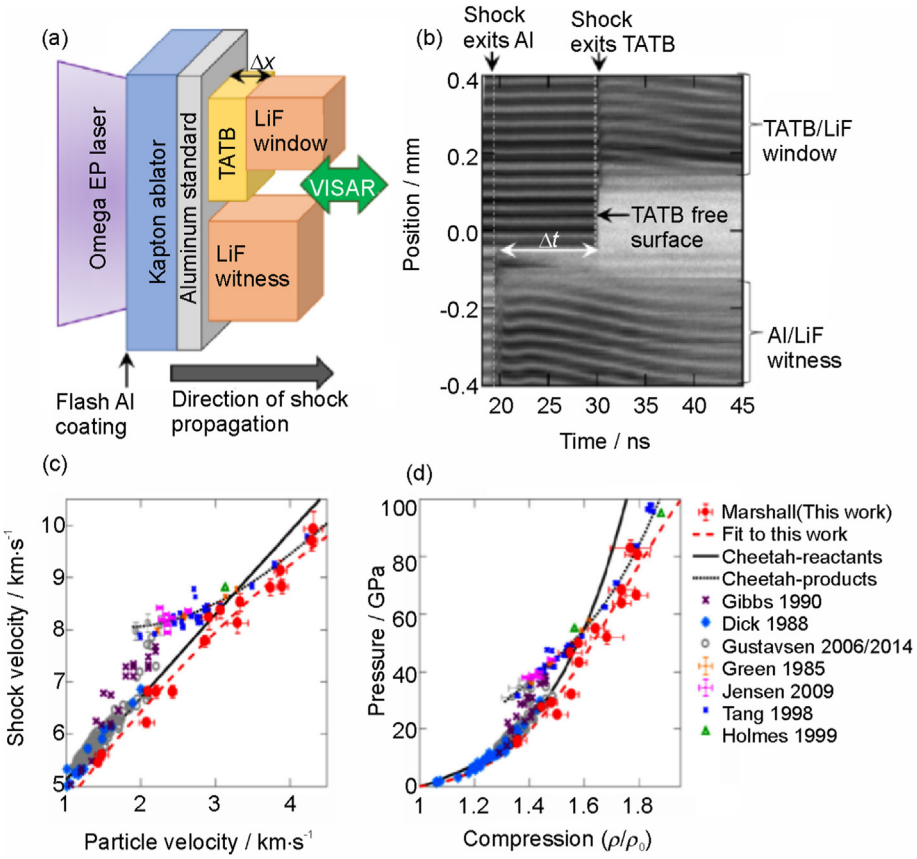

Fig. 2. (a) Schematic of the target, (b) VISAR image of a typical shot, and single-crystal TATB Hugoniot data and experimental data for pure TATB and TATB-based formulations in the (c) U_s-U_p plane and (d) P-($\rho = \rho_0$) plane.[42]

without a substrate have been launched using three methods developed.[33] In the first method, multilayer aluminum flyers, including an ablation layer and a heat shield layer ablated by a long tailored laser pulse, were slowly accelerated to a final velocity of 8 km s^{-1} using compression wave reverberation, without melting and vaporization. In the second method, an aluminum flyer and a multilayer flyer were ablated directly by a short pulse and were then accelerated to average velocities of 21.3 km s^{-1} and 55 km s^{-1}, respectively retaining good planarity. In the last method, a single-layer flyer was accelerated using the momentum transition of plasma collision, in which the plasma with a longitudinal density distribution was generated by strong shock unloading in the reservoir. Specifically, a single-layer aluminum foil and a tantalum foil were accelerated stepwise to 11.5 km s^{-1} and 6.5 km s^{-1}, respectively in the absence of X-ray and electron preheating, retaining good planarity and integrity. Flyers driven by the ramp-wave loading technique without a block can be accelerated to higher velocities at lower temperatures. Therefore, these flyers may have better motion and integrity, which are critical to research on explosives and the absolute equations of state measurements.

The high-pressure dynamics of unreacted explosives is of great significance for the understanding of detonation and reaction mechanisms.[50] However, even a very low shock pressure can result in an apparent chemical reaction in explosives. The unreactive equation of state (EOS) is amenable to study using the quasi-isentropic compression technique (also known as laser ramp-wave loading) technique,[50] which can be performed at the magnetically driven and laser facility. The quasi-isentropic compression technique has been applied in many fields, such as the EOS, high-pressure phase transition, dynamic response of materials under a high pressure, planetary physics, and energetic material reactions.[22] A super high pressure of 5 TPa has been achieved by ramp compression at the NIF, making this method attractive and important.[21,27] Using ramp-wave loading, a quasi-isentropic compression experiment of aluminum based on reservoir (CH material) ablation was performed at the ten-thousand-Joule laser facility, yielding a peak pressure of more than 80 GPa, with a rise time of 10 ns and a strain rate of 10^8 s^{-1}.[32] Ramp-wave loading can be used to study the dynamics of unreacted explosives at pressures from several GPa to even higher pressures for the purpose of providing important experimental data on the unreacted explosives.

In general, experiments at a high-intensity laser facility are suitable for studying the microstructure effects and reaction zone of an explosive under dynamic loading. The pump-probe experiments at high-intensity laser facilities make it possible to study the overdriven detonation, short-pulse initiation, unreactive EOS, and microstructure effects of explosives. In the future, more experiments on this aspect can be performed at a laser facility.

4. Mesoscopic structure investigation under dynamic loading

The macroscopic performance of explosives is strongly related to the mesoscopic-scale reaction zone, which contains chemical reactions and shock waves.[1] The mesoscopic structures, such as cracks, pores, and crystal-binder interface, directly affect the sensitivity and chemical reaction rate of explosives. It is difficult to perform *in situ* real-time imaging of internal deformation, damage, and reaction dynamics under dynamic loading[20,51,52] since high temporal and spatial resolutions are required. Dynamic X-ray imaging, which can penetrate materials with a certain thickness, provide scenography of the dynamic process. Currently, advanced X-ray phase-contrast imaging (PCI) based on synchrotron radiation and X-ray free-electron lasers allow real-time *in situ* imaging with a temporal resolution of 100–0.01 ps and a spatial resolution of 1–10 μm. High-speed synchrotron X-ray PCI has been used to characterize the deformation and damage of plastic-bonded explosives under dynamic loading by ultrasound, split Hopkinson pressure bar, and gas guns. Sanchez et al. performed dynamic initiator experiments using X-ray PCI at the APS, which allows direct observation of slapper flight dynamics and plasma instabilities.[53] The results showed that Kapton "bubbled" and impacted the high-explosive pellet instead of tearing it. In addition, a shock wave was observed in pentaerythritol tetranitrate (PETN), and the expansion of the shock front might indicate a rapid response. Product expansion was also observed in PETN impacts. These data contribute to the understanding of the interactions between an initiator and high-energy explosives and provide insight into initiation mechanisms. This study provided an important physical understanding of small-size detonation flow fields and non-ideal characteristics. However, the facility is scarce and infeasible for laboratory use.

The detonation process of high-energy explosives can be studied *in situ* using dynamic X-ray radiography at a high-intensity laser facility. A high-energy X-ray source with high brightness and a diameter of a few micrometers can be obtained using a high-intensity short-pulse laser. In this sense, X-ray imaging with micrometer, femtosecond-picosecond resolutions can be achieved using this source,[38] thus providing an effective method for studying the dynamic properties of explosives. A laser plasma source consists mainly of Wakefield acceleration, Bremsstrahlung, and Compton scattering. The diameter of an X-ray source based on the Wakefield acceleration method can be minimized to less than 5 μm, which is suitable for high-resolution radiography.[54] Dynamic flyer imaging can be performed at a repetitive 1J femtosecond laser facility and has a spatial resolution of up to 5 μm, which is comparable to the PCI results at APS. Currently, X-rays from a picosecond laser with a microwire target can have energy of several tens of keV, which can penetrate materials with a thickness of several mm.[23] The dynamic imaging of a laser-driven flyer is shown in Fig. 3. According to this figure, the flyer has good motion and integrity, which are critical to explosive initiation.[37] In addition, it becomes possible to study the detonation wavefront and shock-to-detonation mechanisms in a column during the complex reactions of an explosive.

The X-ray projections of microstructures allow observation during the evolution of reactions. Roy et al.[25] studied hotspot ignition and growth through methods from tandem high-resolution numerical simulations to the visualization of micro-scale shock-induced reactions using nanosecond microscope imaging and optical pyrometry. As a result, they obtained the overall agreement that the time to complete the consumption of crystals with sizes of 100–300 μm during hot spot evolution was ~100 ns. In addition, natural speckles can be used for correlation analysis to track displacement/strain fields. X-ray digital image correlation (DIC) is more advantageous when internal features (e.g., explosive crystal particles) are used to generate speckles by PCI. X-ray DIC can be developed using a high-intensity laser facility to provide smaller speckles(50 μm or less) and a higher spatial resolution (10 μm or above).[25] Therefore, the deformation dynamics for local regions, especially for particle-matrix interfaces in plastic-bonded explosives, can also be resolved using X-ray DIC.

5. Phase transition and reaction kinetics under dynamic loading

Structural phase transition, reaction growth, and product formation are key compositional steps in the entire detonation process. Important factors in understanding the ignition and detonation mechanisms of explosives include the crystal structure, phase fraction, grain size, and chemical reaction products of the explosives during dynamic loading.[55] However, understanding the physical and chemical changes is a long-term challenge due to a lack of experimental capabilities. In addition, it is necessary to further understand the phase transition kinetics and reaction product composition of explosives using X-ray diffraction or scattering spectra. Dynamic X-ray spectroscopy based on a bright, monochromatic, and short pulse X-ray source serves as an efficient method to study the crystal structure under dynamic loading. High-temporal-resolution X-ray diffraction and scattering spectra have been used to unravel the complex reactions of explosives. Elissasios et al. measured the structure of the TATB under detonation-induced conditions using high-temporal-resolution X-ray diffraction at the APS, and the results suggested surprising resilience of TATB under high-pressure, high-temperature, and shock conditions.[56]

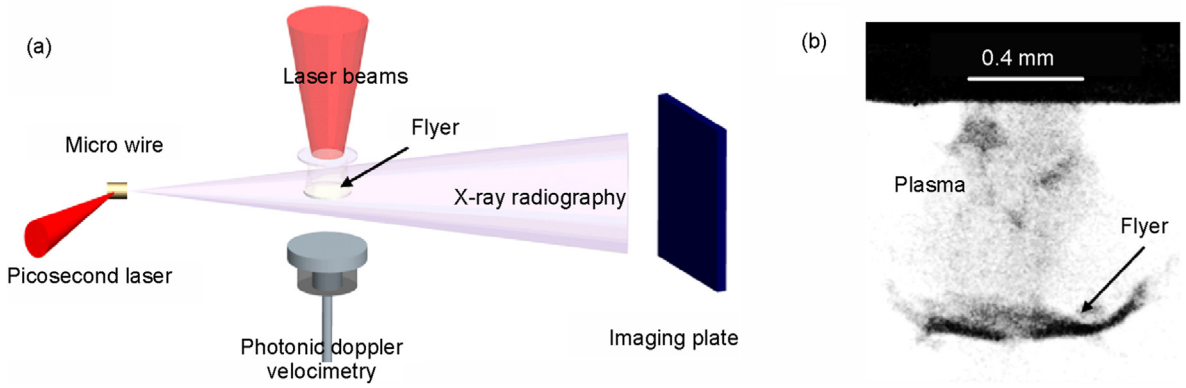

Fig. 3. (a) Schematic of the high-resolution X-ray imaging of a laser-driven flyer via the picosecond laser interaction with the microwire target, (b) the dynamic imaging of an aluminum foil flyer, snapshot at the time of 350 ns after laser loading. The thickness of 20 μm and the diameter of 440 μm are determined from the X-ray imaging.[37]

A high-intensity laser facility is an ideal platform for the development of dynamic X-ray diffraction technology due to the pump-probe methods and the accurate synchronization capability it provides. At this facility, a sample can be pumped with a nanosecond laser and probed with X-rays produced by laser-plasma interactions. Fernandez et al. introduced the phase transition and chemical reaction in shock-compressed TATB at the OMEGA EP laser facility.[26,57] Specifically, the TATB was shocked at pressures of 10–90 GPa using a continuous shock wave with a duration value of 20 ns. Then, the crystal structure of the shocked TATB was studied based on the X-ray diffraction generated by the He_α emission from the Fe foils irradiated by intense laser beams. The initial results indicated that the TATB single crystals remained as highly textured solids at pressures of up to 40–50 GPa. A change in the X-ray diffraction pattern was observed when TATB was shocked at a pressure of above ~55 GPa. A new powder line, corresponding to the diffraction peak of the (111) plane of diamond, was observed at a pressure of 90 GPa. Recently, the solid carbon products from chemical reactions were measured directly *in situ* using two X-ray diffraction beams on the similar target in less than 50 ns at the NIF, as shown in Fig. 4.[24,29,58] The experiment used a non-detonable amount (less than 7 mg) of TATB single crystals and captured the time evolution of the products under shock compression in excess of 150 GPa. This experiment helps to understand This experiment helps to understand the formation of reaction product formation under varied shock pressures. Based on the ten-thousand-Joules laser facility, the feasibility of studying the crystal structure of explosives in a single shot using time-resolved X-ray diffraction can be verified. The diffraction experiment had a temporal resolution close to 1 ns. The diffraction images of high-energy explosive based on laser-plasma X-rays had a high signal-to-noise ratio. Overall, the dynamic X-ray diffraction technology combined with a laser-plasma X-ray source has the potential for research on the microscopic mechanisms (e.g., the phase transition and solid reaction products) of explosives under dynamic loading. It helps to further understand the EOS and yields the strength and phase transition of high-energy explosives.

Carbon condensation is one of the most important processes in the detonation of carbon-rich high explosives. The accurate measurements of the nanodiamond size distribution and nucleation process provide direct access to the kinetics of carbon condensation relevant to explosive interior. X-ray diffraction (XRD) identifies crystalline and liquid correlations at the Angstrom level, while the SAXS is sensitive to feature sizes with an order of magnitude of 1–100 nm.[59,60] The SAXS, which is based on X-ray scattering theory, can probe particles or cavities with lengths from a few nanometers to micrometers.[60] The size distribution and growth process of nanodiamonds formed from shock-compressed TATB were consistently characterized by *in situ* SAXS at the APS and XFEL facilities, demonstrating the importance of the pressure (P) - temperature (T) state for the diamond formation kinetics. Preliminary experiments have been performed to study the solid carbon in the detonation products of carbon-rich high explosives at the APS.[59] Since this technology requires a highly collimated, mono-energy, high-brightness X-ray source, the feasibility of establishing the SAXS technology based on a laser-plasma X-ray source is still under consideration. High brightness is an

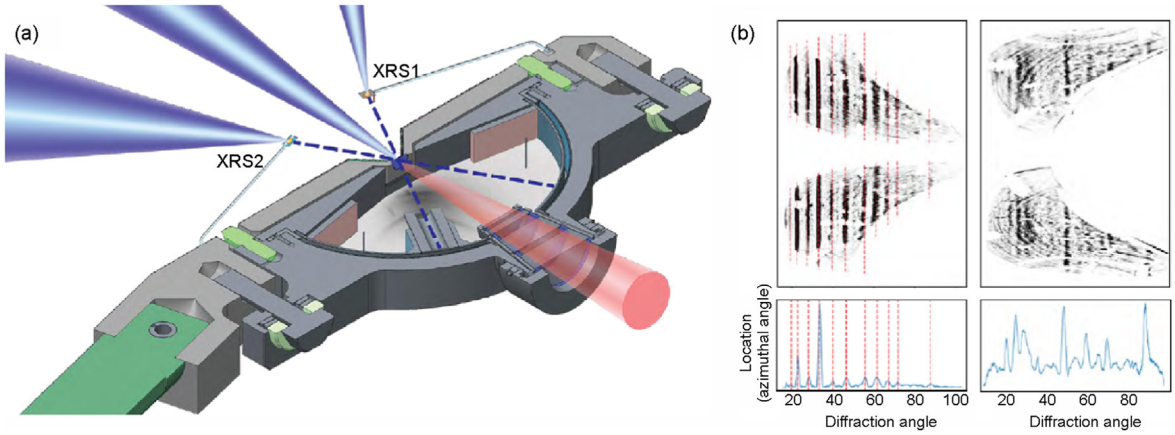

Fig. 4. (a) The TARDIS (Target Diffraction In Situ) body holds image plates used to collect X-ray diffraction signals. In this experiment, TARDIS provided structural X-ray diffraction information at two time points, as indicated by the dashed lines labeled XRS1—the early probe at 15 ns—and XRS2—the late probe at 49 ns. (b) In the diffraction data from the high-explosives shot at the early time point (left, XRS1 = 15 ns (ns)) and the later time point (right, XRS = 49 ns), the Livermore team observed uncompressed TATB at the early times (indicated by the dashed red lines) and evidence of reaction products (to be confirmed in future experiments) at the later time probe.[58]

advantage of a laser-induced X-ray source. However, it is difficult to improve the collimation and mono-energy of an X-ray source. X-ray mirrors are essential for the application of laser-induced X-ray sources, while there is no report of SAXS based on a laser facility. In the future, the increasing demand for carbon condensation research may lead to the establishment of SAXS at a high-intensity laser facility.

6. Ultrafast dynamic research at a molecular level

Molecular explosives are subjected to high temperatures and high pressures on a nanosecond to femtosecond time scale when they are ignited for detonation. The ignition process, which involves key steps such as electronic excitation and molecular excitation, is expected to occur at the wavefront on a nanometer-micrometer scale. Ultrafast laser spectroscopy, which adopts the pump-probe experimental method, can achieve a high synchronization accuracy on a femtosecond scale. The ultrafast dynamics of explosive reactions can be studied based on changes in spectra and enjoys unmatched advantages in detecting the structural changes, molecular bond cleavage, molecular reactions, and intermediate product components of explosives.[61] Numerous important results have been obtained regarding the geometric change and energy transfer process of explosives, including the following aspects:

The fundamental questions of the shock initiation of explosives at the molecular level include the role of electronic excitation in the shock initiation, vibrational non-equilibrium, and the temperature in the kinetics of the chemical reaction. McGrane et al.[17] conducted experiments to understand the molecular-level energetic materials under shock initiation. They estimated the role of electronic excitation in the shock initiation using the femtosecond ultraviolet/visible absorption spectroscopy, simultaneously measure the temperature and chemistry in the Stokes and anti-Stokes spectral regions using the femtosecond stimulated Raman spectroscopy, and presented typical experimental results of 1,3,5-trinitroperhydro-1,3,5-triazine (RDX), which help to justify or refute assumptions that must be made in the process modeling. These effects have significantly advanced the understanding of the dynamic responses of energetic materials at a molecular level.

Electronic excitation drives molecules to enter an excited state, which is thought to occur on an ultrafast time scale. This driving process is a key reaction pathway in the decomposition of explosive molecules. It occurs in nanoseconds but plays an important role in energy transfer. Chu et al. investigated the excited-state dynamics of the HNS and TATB using femtosecond transient absorption spectroscopy, as shown in Fig. 5.[62,63] According to this figure, an equilibrium between the S_1* state, the rapidly populated S_1 state, and the long-lived T_1 state, and a cascading de-excitation model was established based on the experimental investigation. Moreover, the electron transfer process of TATB agreed well with the spectral change. These results suggest that some typical nitro explosives behave similarly in the excitation and relaxation processes, where nitro groups are activated by the excitation energy and then have a relaxed structure in the excited state.[64]

It is believed that the decomposition products in the electronically excited state partly reflect the products in the detonation transient state and play a crucial role in energy conversion. In this sense, the non-adiabatic unimolecular decomposition pathways of energetic materials

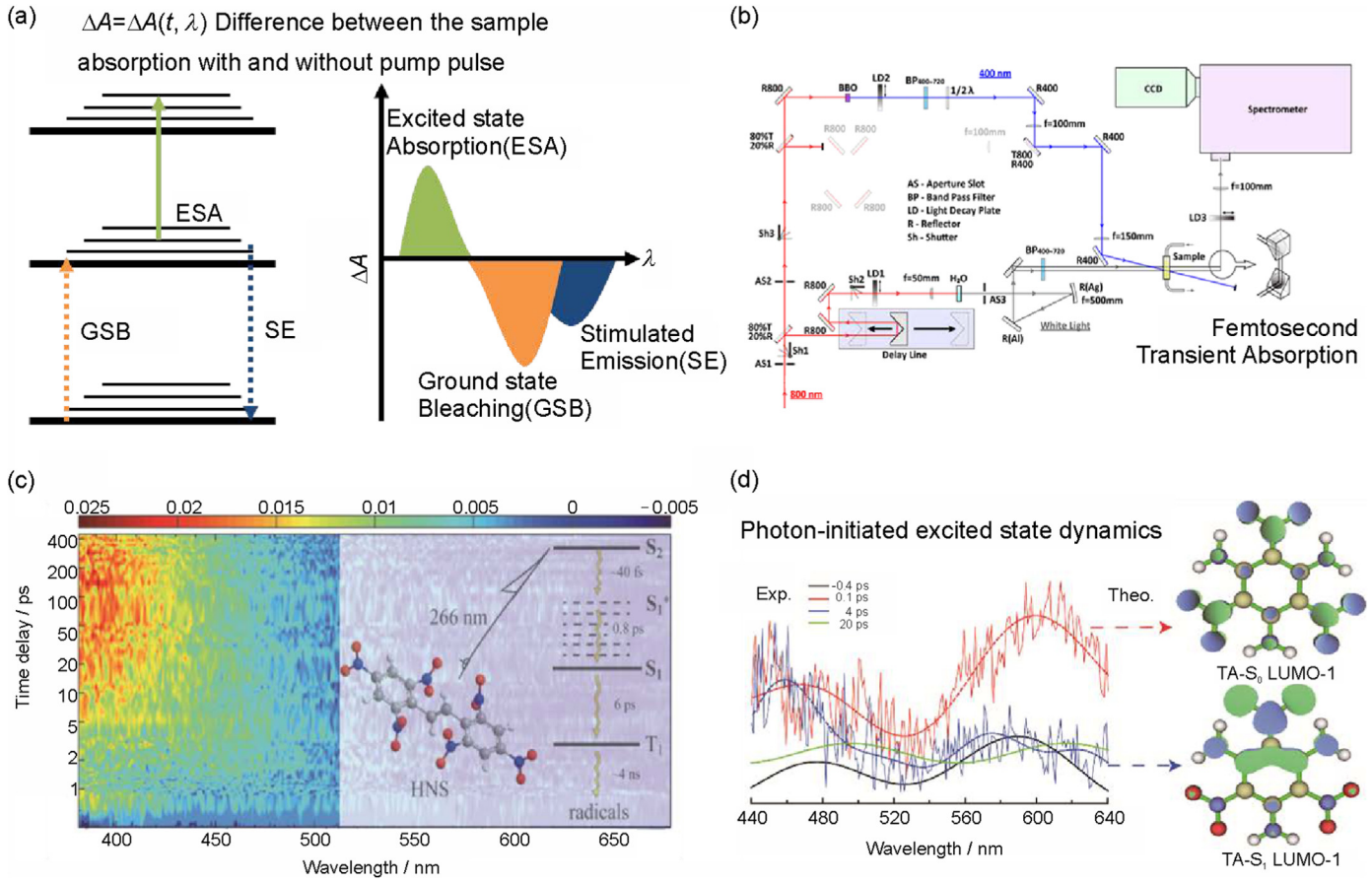

Fig. 5. (a) The principle of transient absorption spectroscopy, (b) the schematic of femtosecond transient absorption spectroscopy, (c) the transient absorption of HNS and its de-excitation model. An equilibrium between the S_1* state, the rapidly populated S_1 state, and the long-lived T_1 state is obtained. The lifetimes of S_1* and S_1 are about 0.8 ps and 6 ps, respectively. The T_1 state has a lifetime of about 4 ns. A cascading de-excitation model of HNS is established.[62] (d) Time-resolved absorption of TATB and electron transfer process.[63] The initial S_1* state can be observed at an excitation of 400 nm and then evolves into the S_1 state. The electron is transferred from the C-ring and NH_2 groups to the NO_2 groups after the vertical excitation. The initial S_1* state relaxes to the S_1 state through the structural relaxation of an active nitro group.

in the excited state are important for understanding the detonation mechanisms of explosives.[65,66] Berstein et al. studied femtosecond time-resolved spectroscopic techniques used to experimentally determine the decomposition mechanisms and dynamics of energetic species, which are consistent with multiconfigurational methods for modeling non-adiabatic molecular processes of energetic molecules. Nitramine- and furazan-based energetic species have been studied and discussed in detail.[65] The model systems have molecular structures similar to those of the energetic species and are critical to the understanding of the decomposition behavior of larger and more complex energetic molecules.

It is a mystery how the shock energy ignites an energetic material. Femtosecond laser multiphonon scattering has been used to study the doorway-mode up-pumping mechanisms.,[67–72] revealing that a single phonon mode is excited and then energy is redistributed to the other modes. For example, a 3-phonon theoretical analysis of energy up-pumping in RDX has been presented, helping to show how energy transfer dominates.[73]

Ultrafast laser spectroscopy is a powerful tool for studying the structural, geometrical, and chemical changes upon electronic or vibrational excitation. It is essential to unravel the inherent physical and chemical mechanisms of nitro explosives at a molecular level, which might contribute to understanding ultrafast and complex photo-initiated reactions and detonation physics.

7. Conclusions

It is crucial to understand the reaction mechanisms and the reaction process of explosives on different scales, which, however, presents a challenge due to the complex reaction kinetics of the explosives and a lack of powerful methods presently. The pump-probe experiments at large laser facilities provide various flexible load and probe combinations for the study of the reaction kinetics and dynamic process of high explosives in a wide temporal and spatial range. This paper introduces the recent progress made in research on high-energy explosives based on high-intensity laser facilities.

First, the micron-sized explosives are driven to a tunable pressure range from low-pressure ignition to overdriven detonation by laser loading. This is suitable for the study of the microstructural effect and the reaction zone of explosives under dynamic loading. Then, further experiments on overdriven detonation, short-pulse initiation, non-reactive EOS, and microstructure can be carried out using the pump-probe experiments at a high-intensity laser facility.

Second, mesoscopic structures, such as cracks, pores, and crystal-binder interface, directly affect the sensitivity and chemical reaction rate of explosives. High-resolution transient X-ray radiography makes it possible to achieve *in situ* real-time imaging of internal deformation, damage, and reaction dynamics under dynamic loading. The detonation process of high-energy explosives can also be studied *in situ* using dynamic X-ray radiography at a high-intensity laser facility. This technique can be used to study the microstructural evolution of high-energy explosives under dynamic loading and is of great importance for the performance optimization of explosive foils, as well as the design of new and reliable initiating devices.

Third, important factors in understanding the ignition and detonation mechanisms of explosives include the crystal structure, phase fraction grain size, and chemical reaction products of explosives under dynamic loading. These are likely to be studied using dynamic X-ray diffraction at a high-intensity laser facility.

Finally, ultrafast laser spectroscopy provides powerful methods to study the structural, geometrical, and chemical changes upon electronic or vibrational excitation. It is essential to unravel the inherent physical and chemical mechanisms of nitro explosives at a molecular level, which might contribute to understanding ultrafast and complex photo-initiated reactions and detonation physics.

Therefore, many studies on the reaction kinetics of shock and detonation processes can be conducted at high-intensity laser facilities. In the future, pump-probe experiments can be used to study complex reactions involving the coupling effect of chemical reactions and shock waves in order to obtain in-depth understandings of bond breaking/formation, local energy populations and their redistribution, changes in structure and stoichiometry, phase separation, and kinetics under dynamic loading. These experiments represent a significant challenge since it is essential to develop a new generation of *in situ* diagnostics for angstrom- to millimeter-scale lengths. The ultimate goal of pump-probe experiments that combine both optical and X-ray (or other particles) probes is to achieve the femtosecond imaging of chemical reactions at material surfaces and interfaces or buried within a compressed sample with an atomic-scale spatial resolution.

Author contribution

Gen-bai Chu, Writing—review and editing; Tao Xi, Shao-yi Wang, Min Shui, Writing—original draft; Yong-hong Yan, Guo-qing Lv, Yao Wang, Ming-hai Yu, Xiao-hui Zhang, Fang Tan, Carrying out experiments; Jianting Xin, Liang Wang, Yu-chi Wu, Jing-qin Su, Data analysis and discussions; Wei-min Zhou, Project planning and experimental design.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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