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# Investigating phase dynamics of materials under laser-induced extreme conditions

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## ABSTRACT

Shock compression driven by nanosecond-laser techniques generates extreme pressure and temperature conditions in materials, enabling the study of high-pressure phase transitions and the behavior of materials in extreme environments. These dynamic high-pressure states are relevant to a wide range of phenomena, including planetary formation, asteroid impacts, spacecraft shielding, and inertial confinement fusion. The integration of advanced X-ray diffraction experimental techniques, from laser-induced X-ray sources and X-ray free-electron lasers, and theoretical simulations has provided unprecedented insights into material behavior under extreme conditions. This perspective reviews recent advances in dynamic high-pressure research and the insights that they can provide, concentrating on dynamical phase transitions, metastable and transient states, the influence of crystal orientation, microstructural changes, and the kinetic mechanism of phase transitions across a variety of interdisciplinary fields.





## I. INTRODUCTION

The phase diagrams of materials under conditions of extreme pressure $P > 100$ GPa and temperature $T > 2000$ K are crucial for understanding planetary interiors and impact processes and for the development of advanced materials.[1–7] It is commonly accepted that unique equilibrium crystal structures are adopted by materials under given thermodynamic conditions (pressure, density, and temperature), irrespective of the state of strain.[8,9] With static compression (applied using diamond anvil cells, DACs) equilibrium states are achieved over timescales of seconds to hours, whereas dynamic methods (e.g., laser shocks at $>10^9$ $s^{-1}$ strain rates) induce kinetically arrested pathways owing to suppressed atomic diffusion and adiabatic trajectories. This leads to diverging phase boundaries, metastable phases, and interface artifacts that obscure direct comparison.

In the context of the laser shock approach, recent advances in experimental techniques such as time-resolved velocimetry,[9,10] *in situ* X-ray diffraction (XRD) at laser facilities,[11–14] and the employment of X-ray free-electron lasers (XFELs),[15,16] as well as the use of computer simulations enhanced by machine learning,[17,18] have helped to resolve these discrepancies by (1) mapping atomic-scale rearrangement pathways in real time, (2) identifying strain-rate-dependent metastable phases, and (3) quantifying phase transition kinetics—directly addressing nonequilibrium effects caused by timescale divergence. As illustrated in Fig. 1, these developments have revolutionized the high-pressure research field and have provided complex pictures of dynamical

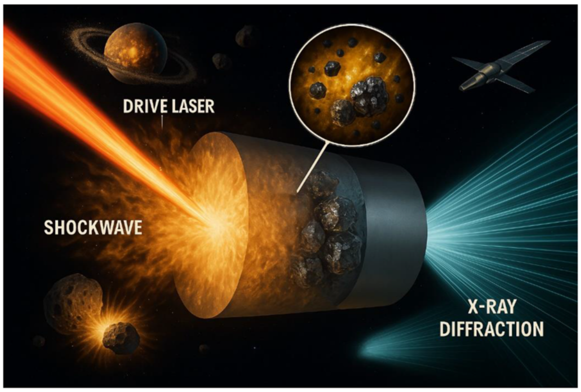


**FIG. 1.** Unlocking phase dynamics of matter under extreme conditions: laser-driven shock compression and X-ray diffraction are revolutionizing high-pressure research and materials science.

phase diagrams, enabling real-time measurements of materials compressed to planetary-scale pressures over nanosecond timescales.

## II. ADVANCES IN EXPERIMENTAL TECHNIQUES

The key advances in experimental techniques are time-resolved velocimetry and *in situ* XRD:

- Advanced techniques for time-resolved velocimetry such as line-imaging velocimetry have enabled high-resolution velocity measurements of dynamic material behavior.[10,19] These methods are crucial for deriving pressure–density relationships and other key physical properties essential for understanding material response at extreme conditions.
- The development of *in situ* XRD techniques at laser facilities and ultrabright X-ray sources such as XFELs[20,21] has provided unprecedented temporal and spatial resolution, enabling the investigation of materials compressed to extreme pressures over nanosecond timescales and has allowed real-time observations of phase transitions and structure crystallization, including grain density, maximum grain size, and crystalline fraction during dynamical compression.[22,23]

Despite significant advances, however, unresolved experimental challenges remain in XFEL-based dynamical compression studies, driving the need for improved methodologies in high-pressure physics. A primary obstacle is the time-evolving pressure gradient along the loading direction, which complicates efforts to observe uniform structural responses under identical loading conditions. While the use of thinner samples reduces this gradient, it introduces new complications: ultrathin specimens amplify interfacial contributions and cause signal discretization, leading to diffuse scattering that obscures true structural features and risks misinterpretation of material states. Concurrently, in laser facilities generating pressures exceeding 600 GPa, intense drive lasers produce strong background noise during nanosecond XRD measurements, overwhelming weak diffraction signals and limiting the accuracy of studies bridging static and dynamic compression regimes. Despite these hurdles, technological innovations and refined analytical approaches have enabled critical advances. Recent works has provided several key insights into the phase dynamics of materials under laser-induced extreme conditions.

## III. KEY INSIGHTS

### A. Phase transitions and metastable states

Phase transitions under dynamical compression are governed by nonequilibrium thermodynamics and kinetic pathways distinct from those under equilibrium conditions. The Hugoniot equations—derived from conservation of mass, momentum, and energy—define the thermodynamic states achievable behind shock fronts. These states often involve metastable phases due to rapid compression rates (nanoseconds). For example, Hwang *et al.*[20] revealed the complex structural dynamics of iron under shock compression and expansion process, identifying dynamical phase transitions ($\alpha$, $\gamma$, and $\varepsilon$ phases) in subnanosecond time. Similarly, the discovery of the metastable Sb–I′ phase in antimony and body-centered cubic (bcc) structures in gold or copper under dynamic compression highlights the emergence of transient states that challenge traditional phase diagrams.[21,24,25] Diamond retains its metastable structure at an unprecedented 2 TPa, resisting the theoretically predicted transition to BC8 carbon.[26] This persistence highlights the strength of its tetrahedral bonds and nearly doubles the highest pressure at which metastable carbon phases have been characterized via XRD.[27] These findings underscore the importance of dynamical phase diagrams in capturing nonequilibrium processes.

### B. Orientation-dependent responses

Single crystals exhibit orientation-dependent mechanical responses under shock compression, owing to the uniaxial nature of the shock.[28,29] Molecular dynamics simulations reveal that shock compression along different crystallographic directions results in distinct transition pathways, explaining why single-crystalline samples exhibit orientation-dependent experimental responses under dynamic loading.[30,31] This behavior highlights the need for advanced models and simulations to capture the full complexity of material responses under dynamic loading.

### C. Phase transition dynamics

Shock compression uniquely generates structural defects such as twinning, dislocations, and stacking faults in metals[32,33] and silica.[34,35] Additionally, shock-induced shear deformation can trigger amorphization in covalently bonded materials such as Si, Ge, and $B_4C$,[36] as well as in metals such as zirconium[37] and $Fe_2O_3$.[23] These phenomena, observed through *in situ* XRD and shock recovery studies and simulated based on advanced potentials, provide critical insights into the deformation mechanisms associated with the microstructural changes that occur at high strain rates under extreme conditions. The recovery of materials generated during laser-driven shock compression experiments remains an emerging challenge, particularly for high-pressure regimes (>100 GPa).[38,39]

Phase transformations under dynamical loading usually follow two dominant pathways.[40] Displacive/martensitic transitions occur through cooperative atomic shifts with preserved neighbor relations, exemplified by shock-induced graphite → diamond conversion. By contrast, reconstructive transitions require complete bond reconfiguration via diffusion-dependent nucleation, as demonstrated in amorphous $SiO_2$ mediated stishovite crystallization.

For reconstructive transitions, homogeneous nucleation obeys the Volmer–Weber model, where nucleation rates scale with undercooling ($\Delta T$) and atomic mobility. Grain growth exhibits two distinct regimes: initial interface-controlled linear growth followed by diffusion-limited coarsening. Recent quasi-isentropic compression experiments—including nanosecond freezing of water into ice VII—validate how classical nucleation theory reconciles seemingly contradictory heterogeneous and homogeneous nucleation modes under extreme driving forces.[41]

Time-resolved diffraction (XRD/XFEL) provides direct experimental validation of these transition modes. Laser-driven shock experiments coupled with time-resolved diffraction reveal complex kinetic pathways, such as in coesite, where a transient dense supercooled liquid first crystallizes into semi-disordered d-NiAs-type silica before pressure-dependent transformation to either seifertite (>70 GPa) or stishovite, with unexpected back-transformation to coesite upon pressure release.[42] Similarly, melting studies demonstrate heterogeneous nucleation dominance in shock-induced melting.[43] Renganathan *et al.*[44] quantified how melting timescales decrease with increasing pressure, establishing a universal kinetic framework for molten-phase evolution under dynamic compression.

Recent findings have demonstrated that dynamic compression enables the discovery of stable phases under both equilibrium and nonequilibrium conditions. Time-resolved experimental observations have necessitated the use of atomic-scale simulations to fully decode these dynamic processes. These advances provide key insights with novel implications, opening promising avenues for future research.

## IV. IMPLICATIONS

### A. Metallic hydrogen

While dense hydrogen exhibits fascinating phenomena,[45,46] the metallic state of solid hydrogen remains experimentally elusive, despite contentious claims.[47–50] Realizing and verifying this novel state at high pressures above 400 GPa represents arguably the most transformative milestone that condensed matter physics could achieve. A deeper understanding of solid phases, structures, and optical/electronic properties of metallic hydrogen/oxygen would revolutionize high-pressure physics and shed light on the interiors of astronomical bodies such as Jupiter, Saturn, brown dwarfs, and white dwarfs.[2,51–55]

### B. Superionic and novel phases

Investigating materials such as iron alloys and water under extreme conditions of pressure and temperature provides insights into superionic and novel phases believed to exist in planetary interiors.[56–59]

### C. Liquid phase behavior

Liquid–liquid phase transitions (LLPTs) have been predicted and documented in diverse elemental systems such as lithium,[60] potassium, and hydrogen under extreme pressure. The associated insulating, semimetallic, and metallic behavior in elements such as hydrogen, water, carbon,[61] silicates, and iron and its alloys have implications for the fundamental understanding of material behavior under ultrahigh-pressure conditions[62,63] and of the dynamics of planetary interiors.

### D. Spin-state transition

The decoupling mechanism between spin-state transitions and metallization processes in iron oxides under extreme conditions has been investigated.[64] Advanced multiscale modeling and experiments in time-resolved X-ray emission spectroscopy under dynamic compression may reveal the kinetics of spin crossover phenomena and explain seismic anomalies in Earth's deep mantle and guide predictions for exoplanetary interiors.

### E. Chemical demixing

A series of post-perovskite (PPV) phase transitions accompanied by chemical demixing is predicted to occur in the Mg–Si–O system under extreme conditions.[4,65] However, direct experimental observations remain elusive, despite their critical importance for understanding the internal structure and composition of super-Earth exoplanets. Diamond formation in hydrocarbon and energetic material systems [e.g., 2,4,6-triamino-1,3,5-trinitrobenzene (TATB)] has been demonstrated.[66] Elucidating these shock-driven transitions by multiscale modeling will refine icy-giant evolution models and allow predictive fabrication of nanocrystalline diamonds and next-generation energetic materials.[66–68]

### F. Behavior of *f* electrons in elemental metals

Strongly localized electrons can undergo delocalization, valence changes, or transitions between magnetic and electronic states, influencing phase transitions, superconductivity, and material properties. Dynamical compression experiments on lanthanides and actinides have probed the unique electronic and structural behaviors of *f* electrons under extreme conditions[69] and have provided fundamental insights into strongly correlated electron systems and their applications in advanced materials.[70]

### G. Framework for kinetics and modeling

Ultrafast XFEL experiments can directly resolve phase transition kinetics—quantifying nucleation barriers and crystalline sizes, validating classical nucleation theory, and distinguishing homogeneous vs heterogeneous pathways through picosecond-scale probing measurements. By integrating XRD with Raman/extended X-ray absorption fine structure (EXAFS) spectroscopy, transient states with critical coordination changes can be tracked. Machine-learning potentials combining *ab initio* accuracy with molecular dynamics scalability will enable predictive simulations of multicomponent systems[71] [e.g., bridgmanite ($MgSiO_3$), Fe–Ni–S alloys, and high-entropy alloys]. Furthermore, the incorporation of atomistic kinetics into continuum hydrocodes will allow the prediction of phase evolution in impact and inertial confinement fusion (ICF) scenarios—a critical capability for advancing future fusion energy technologies.

## V. SUMMARY AND PERSPECTIVES

Research into phase dynamics under shock compression holds significant potential for enhancing our understanding of planetary interiors, impact processes, and the development of novel advanced materials. Future work in this area should emphasize the integration of interdisciplinary methodologies, blending cutting-edge ultrafast diagnostic techniques with computational modeling powered by AI and machine learning to predict and examine complex, novel material behaviors. Tackling challenges such as reproducibility and transient phenomena will be also crucial for progress in this field.

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## AUTHOR DECLARATIONS

### Author Contributions

**Liang Sun**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Bo Chen**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Zhongjing Chen**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Jiayu Dai**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Wenge Yang**: Conceptualization (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **Toshimori Sekine**: Conceptualization (equal); Supervision (equal); Writing – review & editing (equal). **Ho-Kwang Mao**: Conceptualization (equal); Supervision (equal); Writing – review & editing (equal).

## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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