Time-domain experiments in diamond anvil cells

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Challenges:
- Materials characterization under extreme conditions of high \( P-T \)-strain rate
- New materials synthesis under extremes including non-equilibrium conditions

New pulsed laser and X-ray techniques:
- Pulsed laser heating
- Ultrafast laser pump-probe techniques
- Combined X-ray synchrotron-pulsed laser experiments
- Laser driven shock compression in the DAC

Themes:
- Metals thermal EOS and melting: Pt
- Simple diatomics- molecular dissociation: \( \text{H}_2, \text{D}_2, \text{N}_2, \text{O}_2 \)
- Minerals: MgO
- Noble metals-thermal conductivity: Ar
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Scientific challenges: bridge the gap between static and dynamic experiments in P-T-strain rate conditions reached & probed

Phase diagram of hydrogen

Extreme P-T conditions are relevant for:
- warm dense matter
- new materials synthesis
- fast chemical reactivity
- materials strength
- melting curves
- planetary interior

Here we propose to combine static and dynamic experiments in the DAC by performing
- pulsed laser heating
- laser driven shock in the DAC
Melting phenomena and properties of fluids at high P-T condition

Shock & static experiments disagree by 1000’s K

Dramatic decline of melting line?!
Diagnostics of melting is scarce

Problems with static methods
- Instabilities (e.g., diffusion)
- Chemical reaction
- Indirect criteria and lack of positive observations

New techniques are needed to enable accurate measurements of melting phenomena

Improved laser heating techniques:

Time-resolved X-ray & optical techniques:
- Diffuse peak in XRD
- XAS spectroscopy
- Elastic, optical, and vibrational properties
Pulsed versus continuous laser heating in the DAC

Pulsed laser heating: experiment

Finite element calculations, maps

Temperature histories (FE calculations)

- Measurements are very challenging (small volume, strong thermal radiation)
- Uniform in space and time heating in the DAC require longer pulses
Time-domain experiments in laser heated DAC: thermal radiation & chemical reactivity suppression

**Timing for pulsed heat + pulsed Raman operation:**

**Measurement**

**Pulsed heating (ns and μs):** we discriminate spatially and temporally (by measuring ~5-10 μs after the arrival of the heating pulse).

A. F. Goncharov & J. Crowhurst (2005); Goncharov et al., 2008; Goncharov et al., 2010
Rapidly collected Raman spectra show modified intramolecular bonds above 40 GPa.

Subramanian et al. PNAS, 2011
X-ray diffraction combined with pulsed laser heating

- Pulsed fiber laser: 1-50 μs, 5-20 KHz
- Spectrograph & Intensified gated CCD detector
- Laser
- Thermal radiation
- X-ray detector
- Sector 13: GSECARS
- Time-resolved detector: Pilatus

Goncharov, Struzhkin, Prakapenka, Kantor, Rivers, Dalton
Pulsed laser heating in the DAC: μs timescales

GL: A. Goncharov, V. Struzhkin, A. Dalton; GSE CARS: V. Prakapenka, M. Rivers, I. Kantor
Optical Pump-Probe System for Time Domain Thermoreflectance experiments use a double modulation approach

\[ \Delta T = \frac{(1 - R)Q}{C_{Al} A d_{Al}} \]

D. Dalton, A. Goncharov, W.-P. Hsieh, D. Cahill
We are developing a new coherent Antistokes Raman and broad band spectroscopy systems

Fianium Laser
Wavelength: 1064 nm
Rep Rate: Single Shot to MHz
Energy: 2 \( \mu \)J, Pulse Width: <10 ps
Peak Power: \( \sim 20 \) kW

D. A. Dalton, McWilliams
Broadband Optical Spectroscopy will enable single shot study of optical properties at the extreme environments attainable in the DAC.

Supercontinuum Generation (SG) results in a very bright, white light source.

The supercontinuum data was collected in a single shot manner at ~180 nJ/pulse into the fiber.

Tungsten lamp (~3000 K) data collected at $10^3$ longer accumulation time.

D. A. Dalton & S. McWilliams
Time-domain optical spectroscopy in the diamond-anvil cell.

Transient extreme conditions; diamond anvil cell combined with pulsed laser heating.

Ultrasfast absorption spectroscopy using super-continuum optical probe.

Goncharov, McWilliams, Dalton, Geophysical Lab
First Sweep of the Supercontinuum using a streak camera
Coherent Anti-Stokes Raman Spectroscopy (CARS) will be used for time resolved chemistry in the DAC.

CARS has better conversion efficiency than Raman. CARS can discriminate from fluorescence and thermal background. CARS does have non-resonant background.

\[ \omega_{\text{pump}} = \omega_{\text{probe}} \]
\[ \lambda = 532 \text{ nm} \]
\[ \omega_{\text{Stokes}} \lambda = 532 \text{ nm} - 2 \mu \text{m} \]
\[ \omega_{\text{CARS}} \lambda = \sim 300 - 532 \text{ nm} \]
\[ \omega_{\text{molecule}} = \omega_{\text{pump}} - \omega_{\text{Stokes}} \]

\[ I_{\text{CARS}}(\Omega) \propto \left| \chi_{\text{CARS}}^{(3)}(\Omega) \right|^2 I_p^2 I_s. \]
Broadband Coherent Anti-Stokes Raman Spectroscopy (CARS) is planned to perform single shot study of optical properties at the extreme environments attainable in the DAC: first tests at CIW

CARS spectra with supercontinuum at CIW

CARS spectra with supercontinuum

Methanol 0.5 GPa

Dalton, McWilliams, & Goncharov
Broadband Coherent Anti-Stokes Raman Spectroscopy (CARS) is planned to perform single shot study of optical properties at the extreme environments attainable in the DAC: first tests at CIW.

CARS spectra with supercontinuum at CIW

Nitrogen 22 GPa

Dalton, McWilliams, & Goncharov
Laser driven shock compression in the DAC: samples are dynamically compressed in the DAC

- Precompression in 100 GPa range is possible
- Preheating and precooling if needed
- Ultrafast experiments can be small scale: *Table top system*, unlike currently better known technique of laser shocks which involves large laser facilities (such as NIF)

Armstrong and Crowhurst, LLNL
Laser shocks in the DAC can generate and detect 10s GPa shock waves (and low pressure acoustic waves) in materials under precompression of 10s GPa
First shots on deuterium

- Shock and particle velocities of ~12-13 km/s and ~1 km/s for precompression ranging up to 36 GPa, giving a shock pressure ~10 GPa.

- Possible phase transition over the duration of the probe window

M. Armstrong, J. Crowhurst, LLNL
Outlook: the field is mature
We are looking for new opportunities which will be given by new generation synchrotron sources

- Pulsed laser techniques have a great abilities to:
  - access unavailable previously extreme P-T conditions
  - overcome problems of containing and probing chemically reactive and mobile materials
  - study vibrational, optical, elastic, transport properties under extreme conditions

- The full potential of these techniques will be reached with further development of ultrafast (ps to fs) pump-probe & single-shot techniques coupled to pulsed laser heating and laser shocks in the DAC.
  - perform experiments in a time domain to access the time scale and dynamics of phase transitions & chemical reactions

- We are looking forward for developing new combined X-ray – optical techniques at synchrotron beamlines (e.g., ERL, Petra III, XFEL, NSLS-II)