### X-ray lasers, laser-plasmas, and high harmonics: what's best for creating and probing high-energy-density matter?

# Image: state state

absorption spectroscopy

emission spectroscopy





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#### scattering



imaging



# X-rays penetrate materials and are differentially absorbed and scattered by atoms



# Processes in excited or warm, and dense, materials create a phase space under extreme conditions

#### Warm & Dense Matter (WDM)

- Excited state materials
- High pressure materials
- Planetary cores
- Inertial confinement fusion (early time)

#### Hot & Dense Matter (HDM)

- Laser heated plasmas
- Stellar interiors
- Inertial confinement fusion (late time)



Basic Research Needs for High Energy Density Laboratory Physics, DOE (2010)

#### Simulations of electronic structure changes of warm and dense matter can be validated using x-ray absorption spectroscopy





Changes in structure and electronic bonding are observed near x-ray absorption edges using a synchrotron and ps streak camera detector probes laser-heated matter BERKELEY **Beamline at ALS x-ray synchrotron** Streak camera Tunable broadband x-ray pulse x-ray absorption data 70ps 200~1500eV BW ~ 5% 1000 Timing **Ti:sapphire laser** 990 fiducial 800/400 nm, 100 fs, 10 m 980 L2 970 X-ray streak camera detector 960 Energy 13 ps time resolution 950 [e\] 940 930 920 Target Probe x-ray pulse 910 Cu, 70nm from ALS synchrotron 900 -30 -20 -10 20 30 50 10 Time [ps] Spectrograph Laser pump pulse

**Dynamic x-ray absorption spectroscopy** 

# Changes in L-edge x-ray absorption in heated Cu reveal changes in DOS and electron temperatures



Time and temperature dependent electron-phonon coupling accompany electronic structure dynamics



Intense and short x-ray laser pulses can create more uniform hot & dense matter





- Short penetration depth (10-100 nm)
- Valence electron ionization
- Ionization by hot electrons and collisions
- Heating by electron transport and collisions
- Expands in fs



- Long penetration depth (10 -100 μm)
- Core electron ionization
- Ionization by ionizing x-ray flux and collisions
- Heating by large photon energy
- Expands in ps

Uniform, single-state physical data are essential for tests of theory (temperature, pressure, conductivity, opacity)

# An important consequence of intense x-ray illumination is saturation, creating a homogeneously heated sample

• Essential to create homogeneous WDM in a well-defined state (LTE)



# LCLS instrument for Materials in Extreme Conditions (MEC)



# X-ray emission spectroscopy at MEC reveals conditions of X-FEL heated aluminum



# Absorption in solid aluminum is wavelength dependent



Multiple absorption channels at near edge photon energies

# Two types of emission are observed from XFEL-pumped Al: K-alpha from ion stages and resonance scattering



## Dynamics include sequential photo-ionization & Auger decay; ionization depression affects dynamics



Vinko et al Nature 482, 59-62 (2011)

Ciricosta et al PRL 109, 065002 (2012)

# High fluence x-ray laser pulse opens inner shell processes that are inaccessible via single photon process



Cho et al. PRL 109, 245003 (2012)

# High fluence x-ray laser pulse opens inner shell processes that are inaccessible via single photon process



Spectral line broadening is observed at resonances due to high intensity and opacity effects



#### The Ka line widths for different x-ray pulse conditions



Rabi frequency for 1s-2p<sub>3/2</sub>

$$\Omega = \frac{\mu E}{\hbar} = 1.7 \times 10^{15} \text{ Hz}$$
$$\left(\Gamma_{K\alpha} \sim 2.3 \times 10^{13} \text{ Hz}, \Gamma_{KLL} \sim 5.7 \times 10^{14} \text{ Hz}\right)$$

**Power broadened spectral line** 

$$I(\omega) = \frac{\hbar\omega}{2\tau} \frac{\Omega^2}{\Omega^2 + 1/(2\tau^2) + (\omega - \omega_0)^2}$$
$$\gamma_{\omega} = (1/\tau)[1 + 2\Omega^2\tau^2]^{1/2}$$

# Collisional ionization rates also control population dynamics (utilize collisional radiative code SCFLY)

#### **Rates for Atomic and Collisional processes**



# Elastic and inelastic x-ray Thomson scattering reveals plasma properties



Collective modes of dense matter can be probed by varying scattering angle

#### Reveals

- temperatures
- densities
- ionization
- velocity distribution



# Thomson scattering depends on several factors

Three contributions to the Dynamic Structure Factor:

ion feature electron feature bound-free feature  $S(k,\omega) = |f_I(k) + q(k)|^2 S_{ii}(k,\omega) + Z_f S_{ee}^0(k,\omega) + Z_c \int \tilde{S}_{ce}(k,\omega - \omega') S_s(k,\omega') d\omega'$ 

 $Z_f$  =free e<sup>-</sup>;  $Z_c$ =core e<sup>-</sup>;  $f_i$  = ion form factor; q(k)= free & valence e<sup>-</sup> screening cloud



# Thomson scattering used to probe compressed Be



kJ laser pulses create plasma K- $\alpha$  x-rays radiating into  $4\pi$ 

1 mJ x-rays collected in small solid angle & scatters from shocked & heated material

implies that 1 mJ x-ray XFEL pulses can be used to scatter from excited material





# Equation of state measurements of CH plastic at Gbar pressures using the National Ignition Facility

T. Doeppner, A. Kritcher, D. Swift, J. Hawreliak, B. Bachmann, P. Celliers, D. Chapman, G. Collins, J. Eggert, S. Glenzer, D. Kraus, O. Landen, S. Le Pape, H. J. Lee, P. Neumayer, J. Nilsen, R. Redmer, S. Rothman, C. Keane, R. Falcone



Lawrence Livermore National Laboratory • National Ignition Facility & Photon Science

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LLNL-PRES-645847

Compressed CH sphere on NIF ICF platform is used to study Gbar-scale pressures using radiography & Thomson scattering



NIE

# Gbar experiments will be important for testing and validating EOS models

- Gbar pressures occur in ICF and center of brown dwarfs
- Pressure & temperature ionization may affect the compressibility (shell effects)
- Will benchmark models at 100 Mbar 1Gbar







NIF



NIF

# First x-ray scattering data were recorded at 9.33 ns (peak emission + 0.48 ns)



NIF

# We have reached a pressure of 720 Mbar



- Both NIF data sets (RT & cryo) are stiffer than the Sesame and LEOS Hugoniots
- N130701 data is consistent with Nova data (Cauble et al., PRL 1998)
- Possibly first observation of atomic shell effects
- Future shots will lower drive to rule out adverse impact from preheat

X-ray techniques uniquely probe matter under extreme conditions

#### 1. Absorption spectroscopy

- reveals electronic structure, bonding, local order
- utilize synchrotrons, FELs, HHG, Betatron radiation (?)
- 2. Emission spectroscopy
  - reveals pathways among highly excited states
  - utilize FELs
- 3. Inelastic scattering
  - reveals temperatures & densities of electrons & ions
  - utilize plasma backlghters, FELs
- 4. Diffractive scattering and imaging
  - reveals structure
  - utilize FELs
- 5. Ionization mass spectroscopy
  - reveals mass products of evolving molecular systems
  - utilize HHG sources at high rep rate

# Many international collaborators

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# Thank you