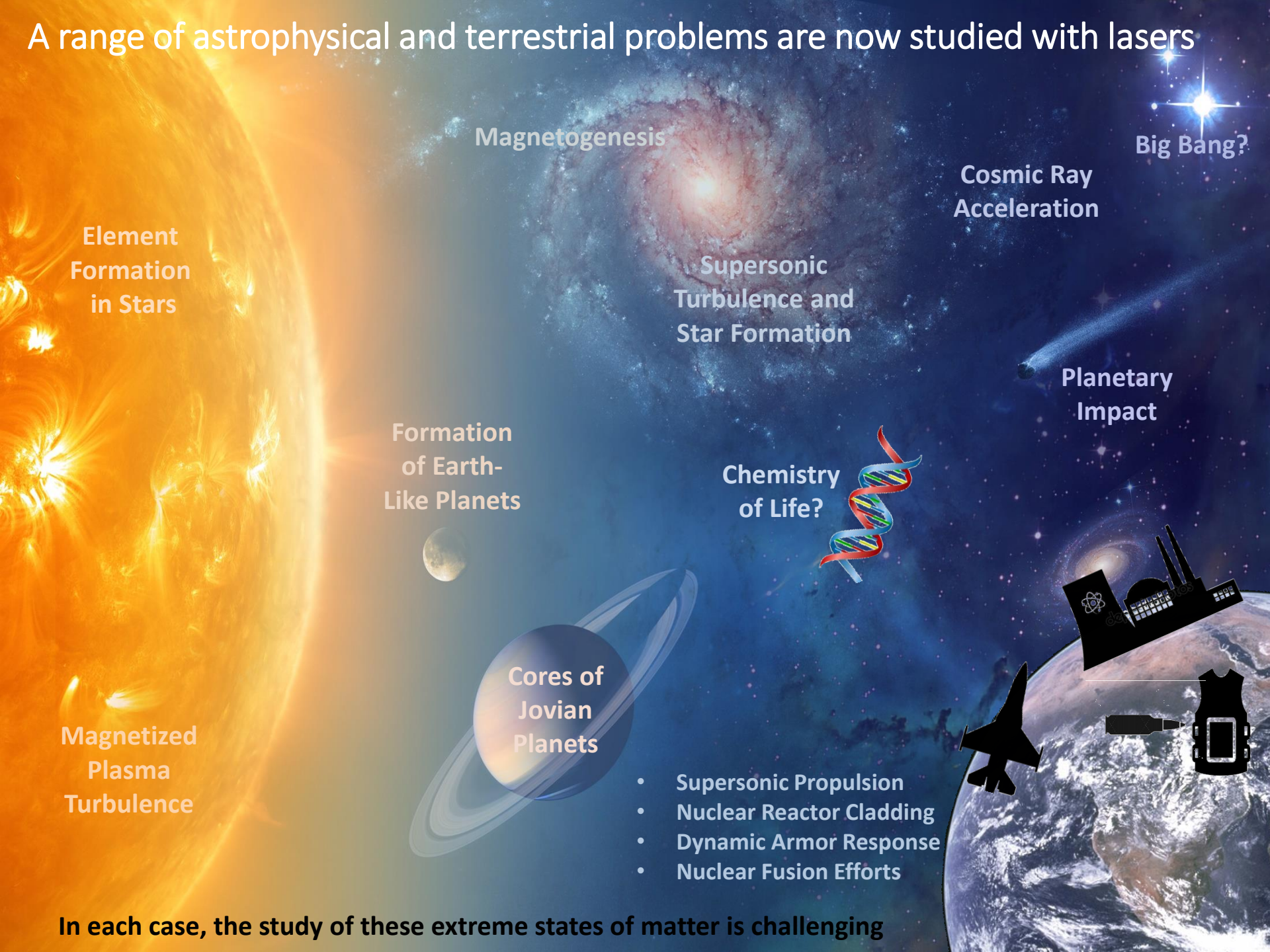
University of Nevada, Reno

Experimental Observation of Ion Acoustic Waves in Warm Dense Methane

Dr Thomas White

A range of astrophysical and terrestrial problems are now studied with lasers



In each case, the study of these extreme states of matter is challenging

Laboratory astrophysics and HEDP research at UNR

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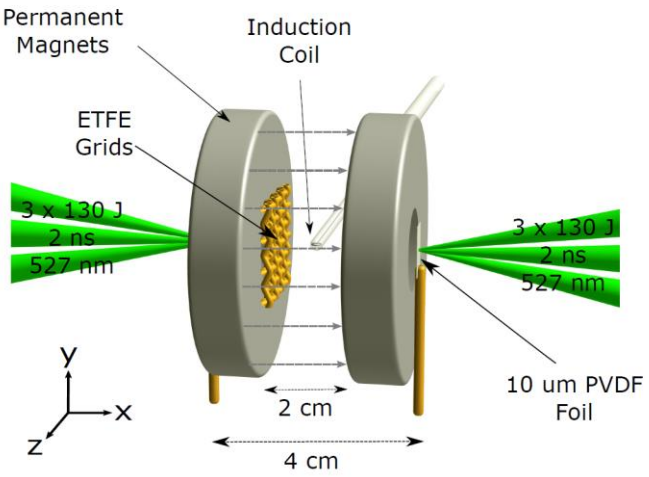
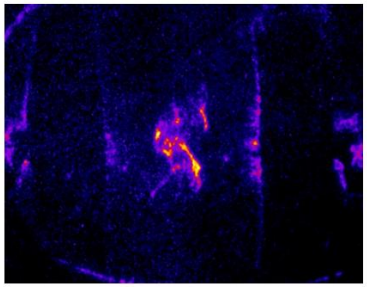
   A. Schekochihin and G. Gregori (Oxford)

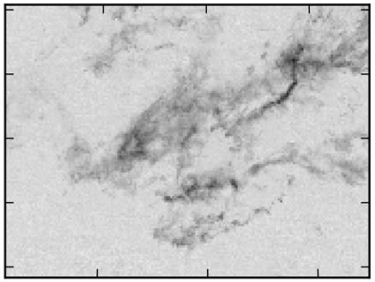


Jacob Molina, Cameron Allen, Thomas White, Emily Chau, Rebekah Hermsmeier, Matthew Oliver, Ryan Davis, Alex Angermeier behind the lens

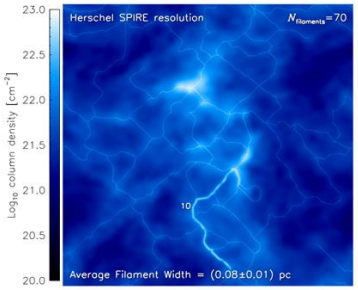
This Experiment



Taurus Molecular Cloud



Simulations (M=5)



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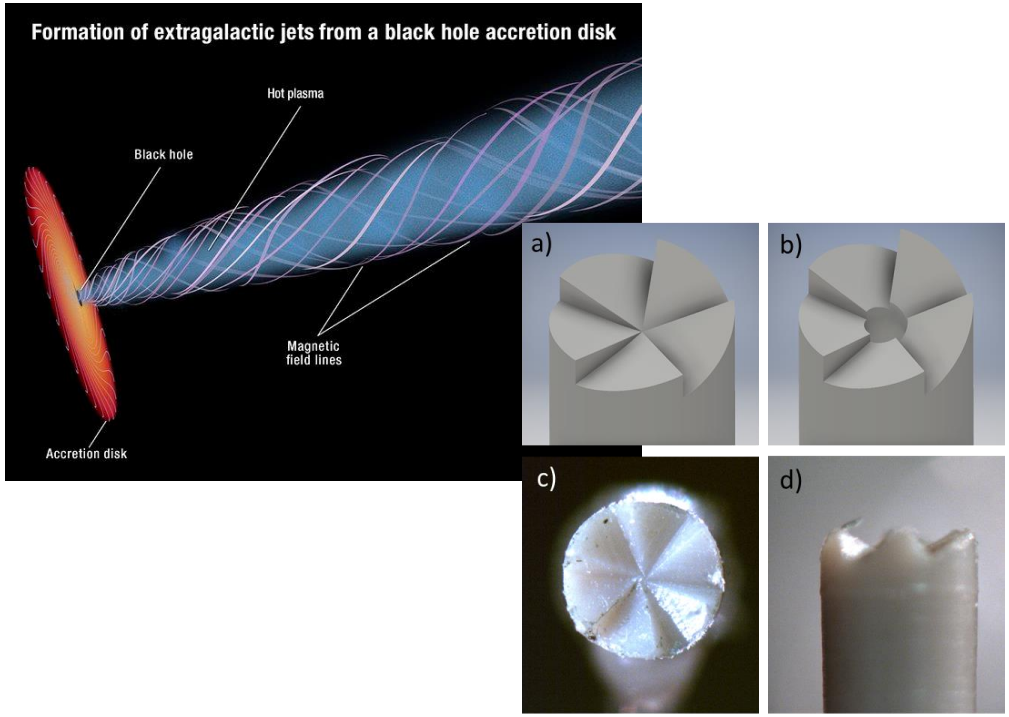

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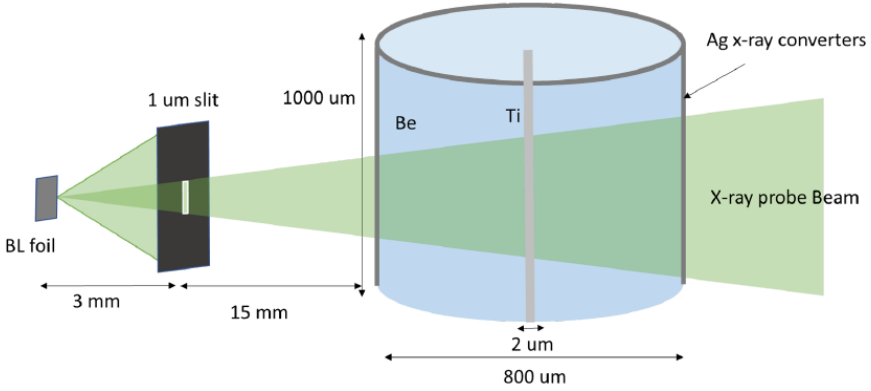

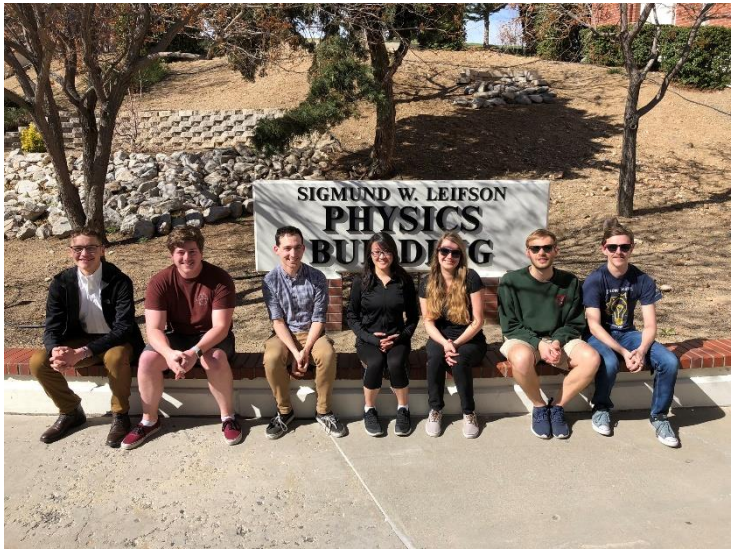

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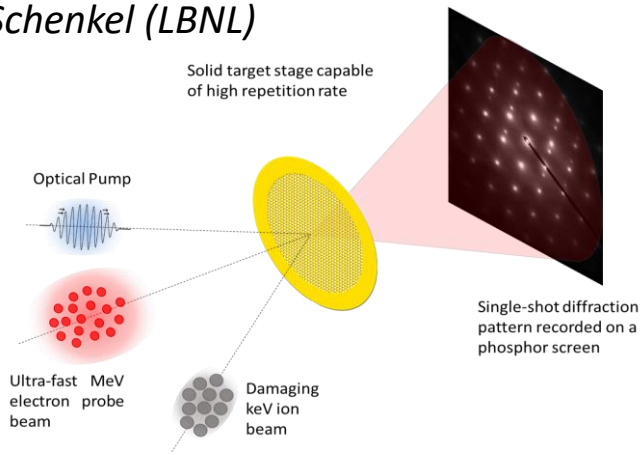

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5. Experimental measurement of ion acoustic waves in WDM (LCLS 2017)

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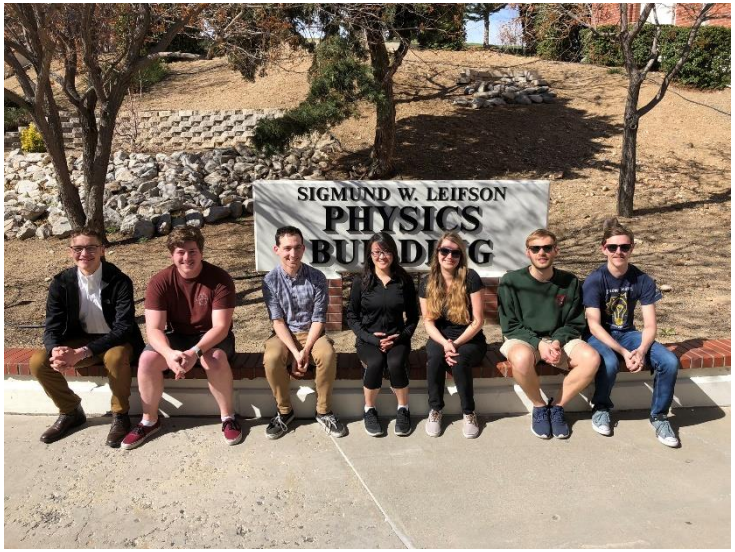

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6. Simulations of WDM/plasmas

   FLASH MHD, DFT-MD, Classical MD, Wave packet Molecular Dynamics

What constitutes an extreme state of matter?

Matter where a physical property (velocity, density, temperature, energy-density) that is beyond what we typically used to in everyday life.

High Energy Density Physics

"matter with an energy density > 10^{11} J/m^3" ➡ Pressure > 1 Mbar / 100 GPa $\sim \frac{\mathrm{eV}}{a_0^3}$

e.g. Warm Dense Matter

- T ~ 0.1 – 100 eV

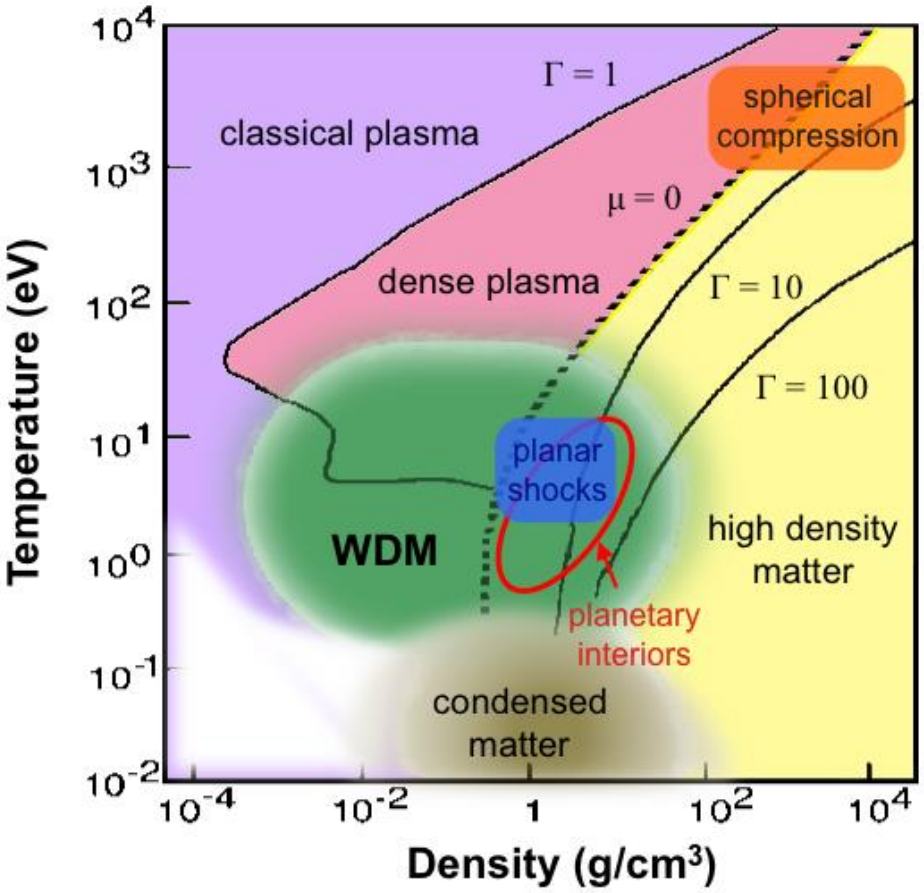
  'Moderate' temperature
- $\rho \sim 10^{-2} – 10^{2}$ g/cm^3

  Solid density
- $\Gamma \sim 0.1 – 500 \sim \frac{\mathrm{KE}}{PE}$

  Strongly coupled ions
- $T_{electron} \leq T_{fermi}$

  Non-negligible electron degeneracy
- $T_{electron} / T_{ion} \neq 1$

  Non equilibrium species



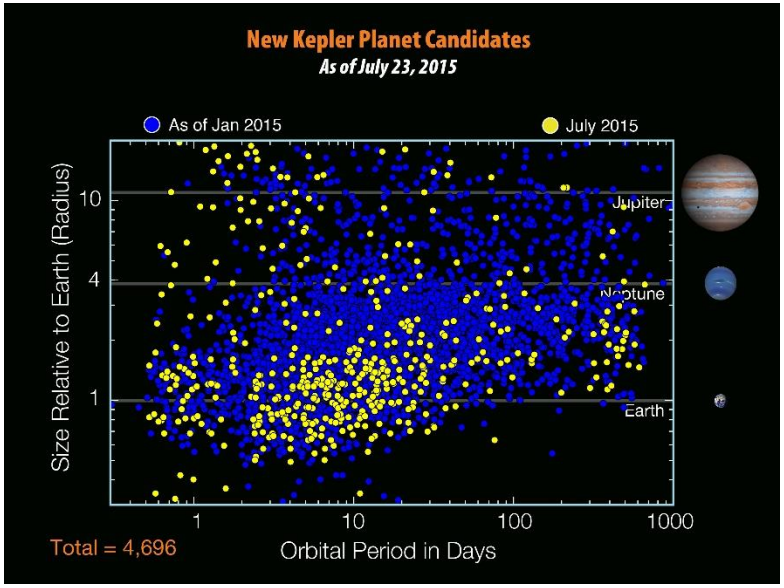
Example: Planetary interiors and exoplanets

Enigma of solar planets

Naptune and Uranus cannot be described by standard planet models

Exoplanets

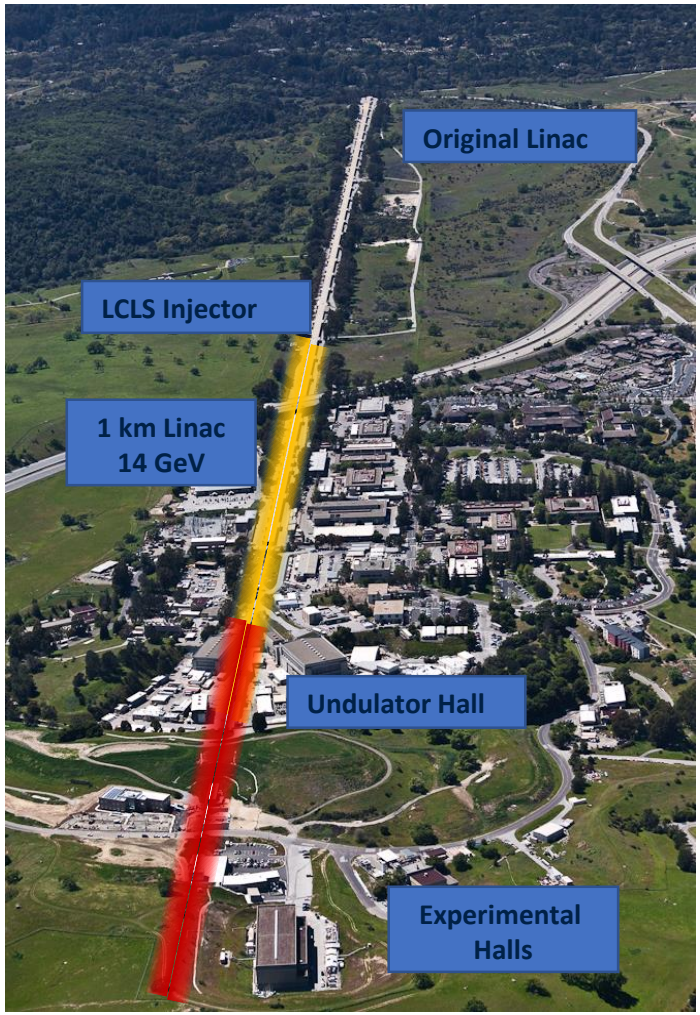
As of May 23, 2016, astronomers have identified 3412 such planets. Questions remain regarding potentially habitability?



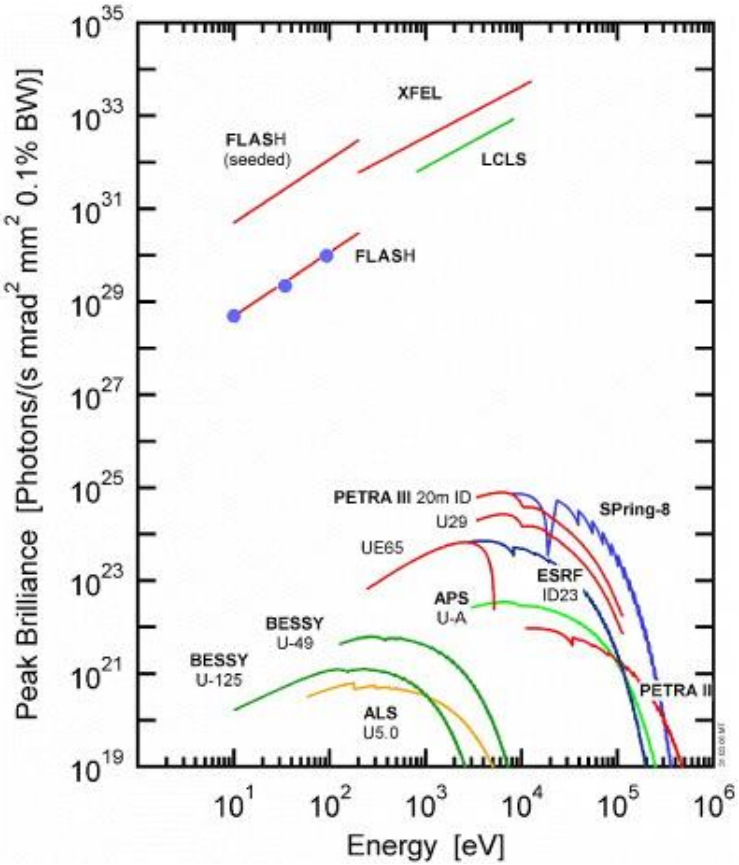
Interpreting observational data from astrophysical objects relies on knowledge of the state and evolution of warm dense matter

Diagnosing High Energy Density States

1. The states of matter exist only for a short time. We must use a short intense probe.



2. Due to high ionization/free electrons the dense matter is typically opaque to visible light. We must use more penetrating forms of radiation.



X-Ray Free Electron Lasers are the brightest X-ray sources on the planet. They are 10^9 times brighter than any synchrotron.

How do X-rays scatter off matter?

Diffraction (Elastic Scattering)

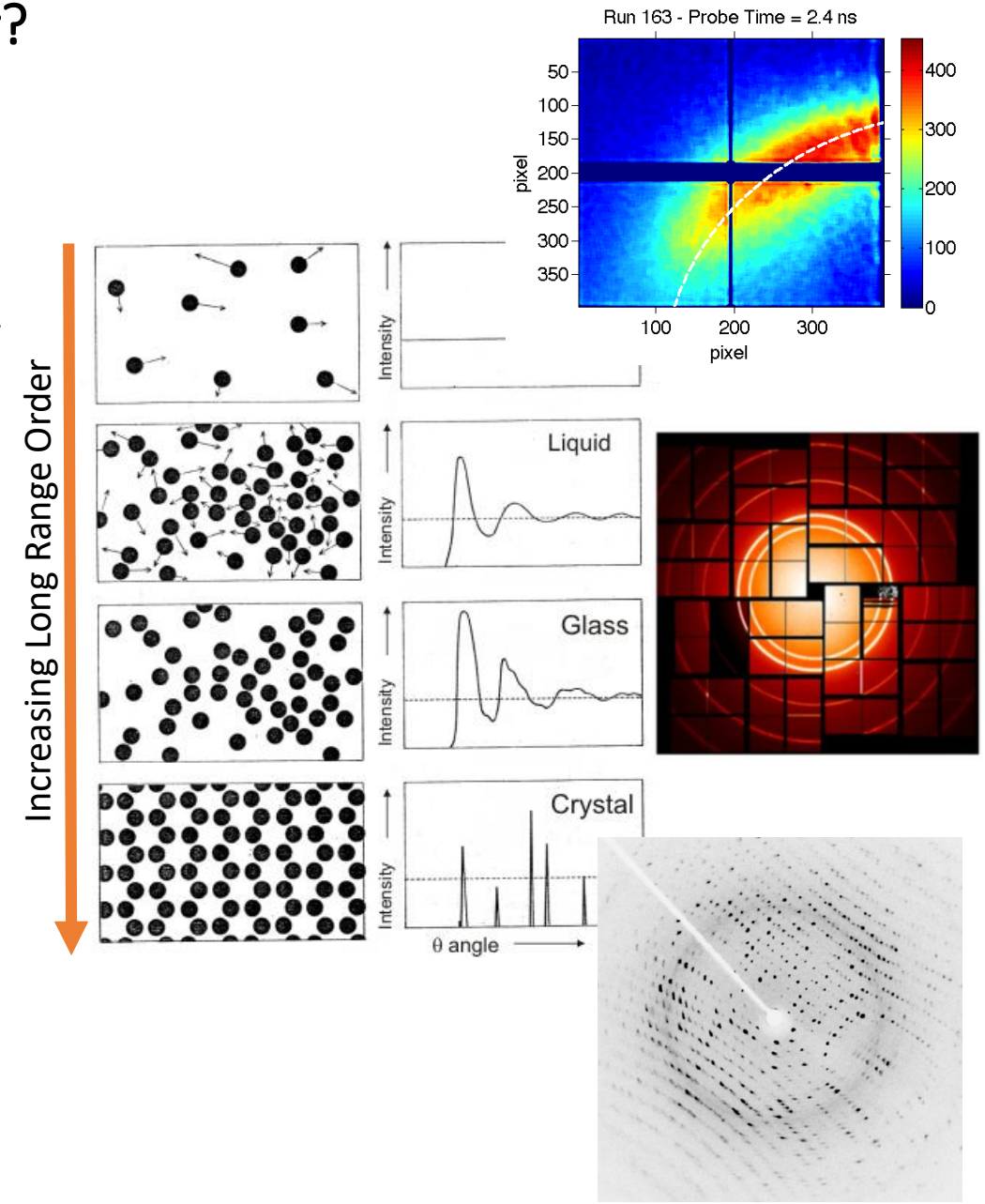
Scattered X-ray beams interfere constructively in some directions, producing diffracted beams (think Bragg's Law!).

More generally we can related the diffracted intensity to the Fourier transform of the density/atomic positions.

$$S(\mathbf{k}) = \frac{1}{N}\left\langle \sum_{jk} e^{-i\mathbf{k}(\mathbf{R}_j - \mathbf{R}_k)} \right\rangle \qquad k \approx \frac{4\pi}{\lambda_0}\sin\left(\frac{\theta}{2}\right)$$

This function is known as the static structure factor.

As well as being able to measure it directly it defines the microstructure of the material and feeds into thermodynamic variables => EOS



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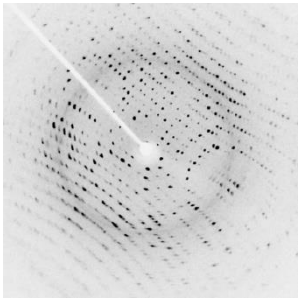
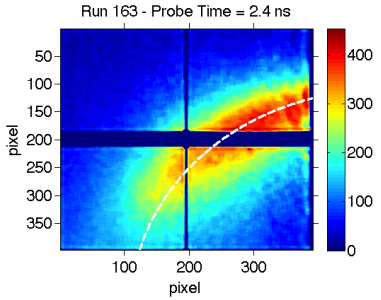
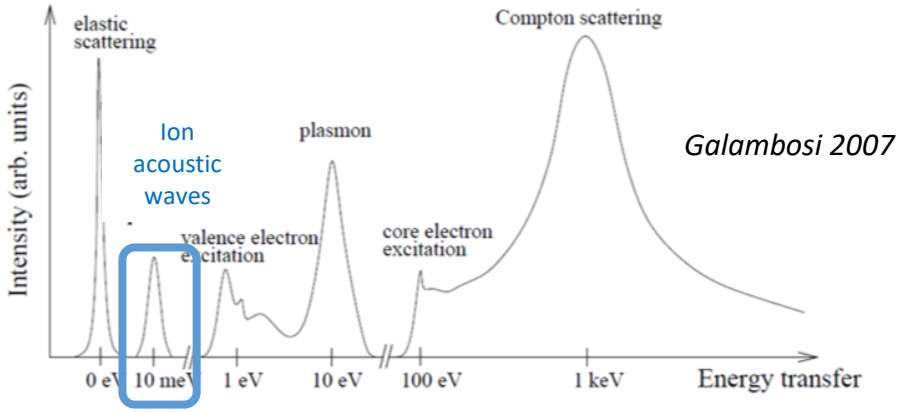
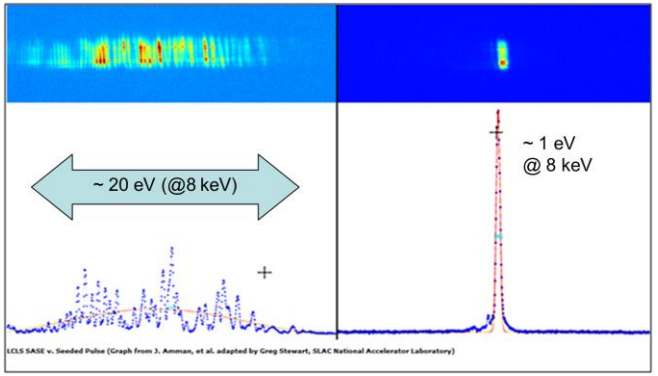


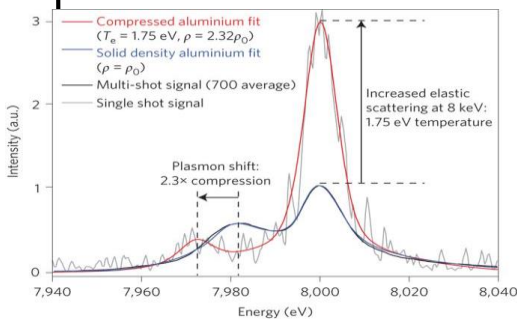


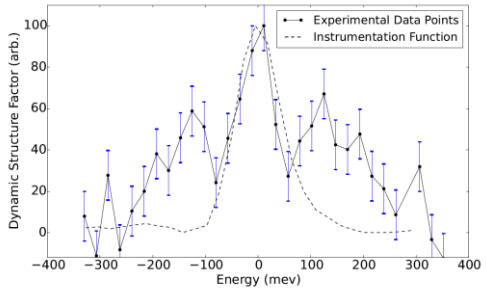
Inelastic X-ray Scattering



Galambosi 2007







A wealth of information can be obtained from the ion acoustic spectra

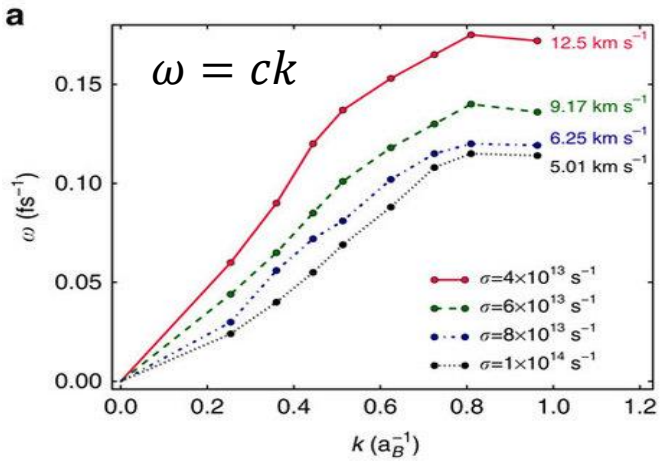
The width of the side Brillouin peaks is related to the plasma viscosity.

The width of the central Rayleigh peak is related to the thermal diffusivity

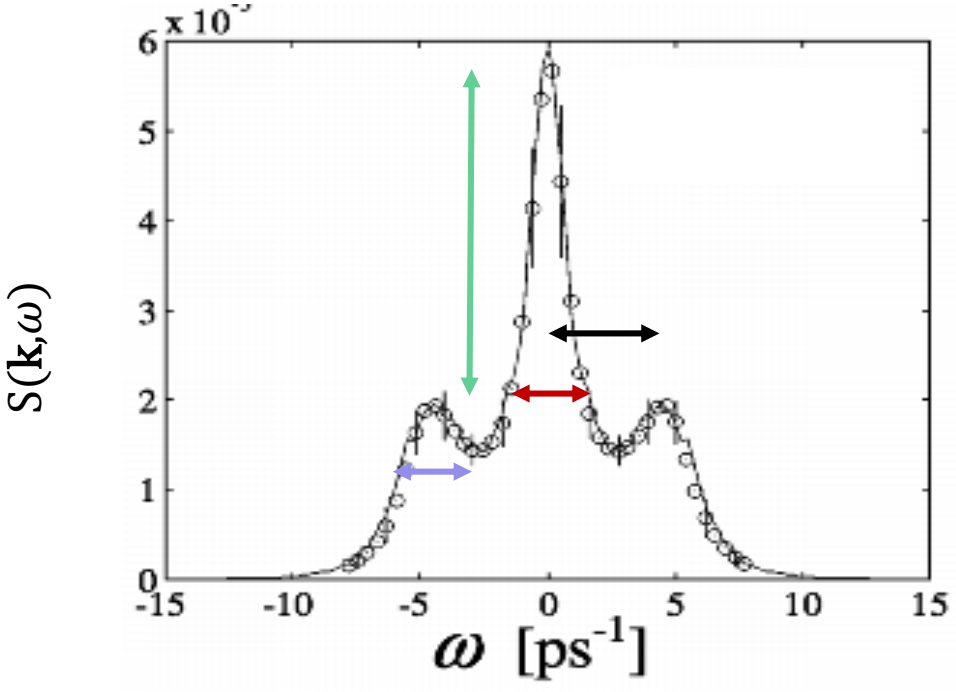
The ratio of the peaks give the adiabatic index or heat capacity ratio

$$\gamma = \frac{C_P}{C_V}$$

The position of the side peaks is related to the dispersion relation and the adiabatic sound speed



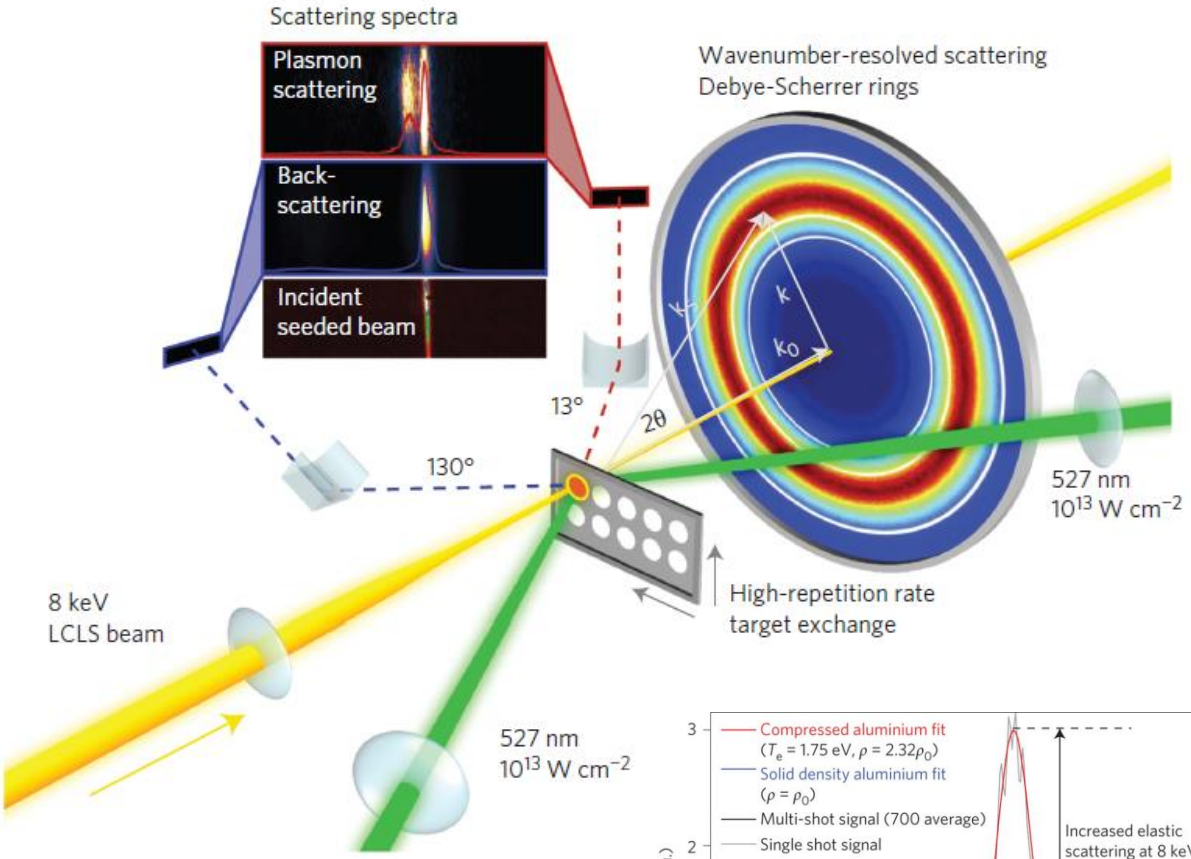
$$S(\mathbf{k},\omega) = \mathcal{F}\left[\frac{1}{N}\left\langle \sum_{jk} e^{-i\mathbf{k}(\mathbf{R}_j(t)-\mathbf{R}_k(0))}\right\rangle\right]$$

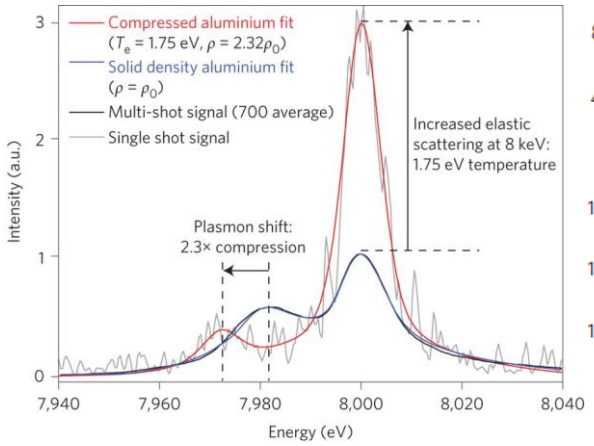


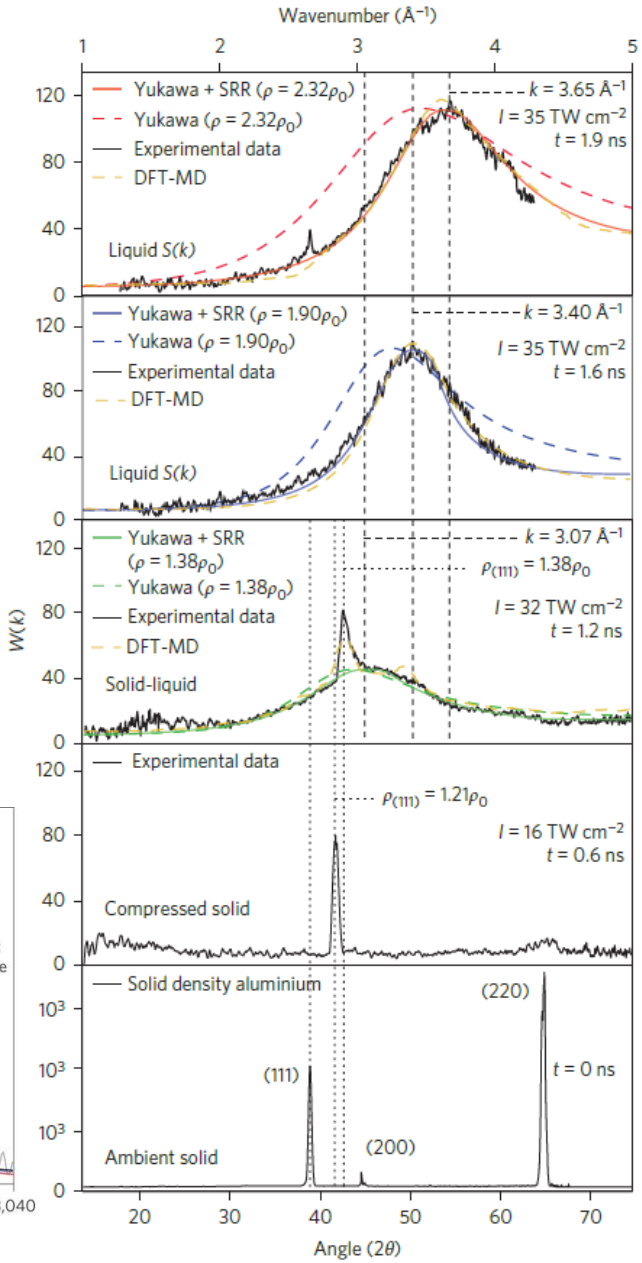
We can perform an experiment to observe these ion waves

Previous Work

Previous Work (L. Fletcher *et al.* Nat. Photonics, 2015)







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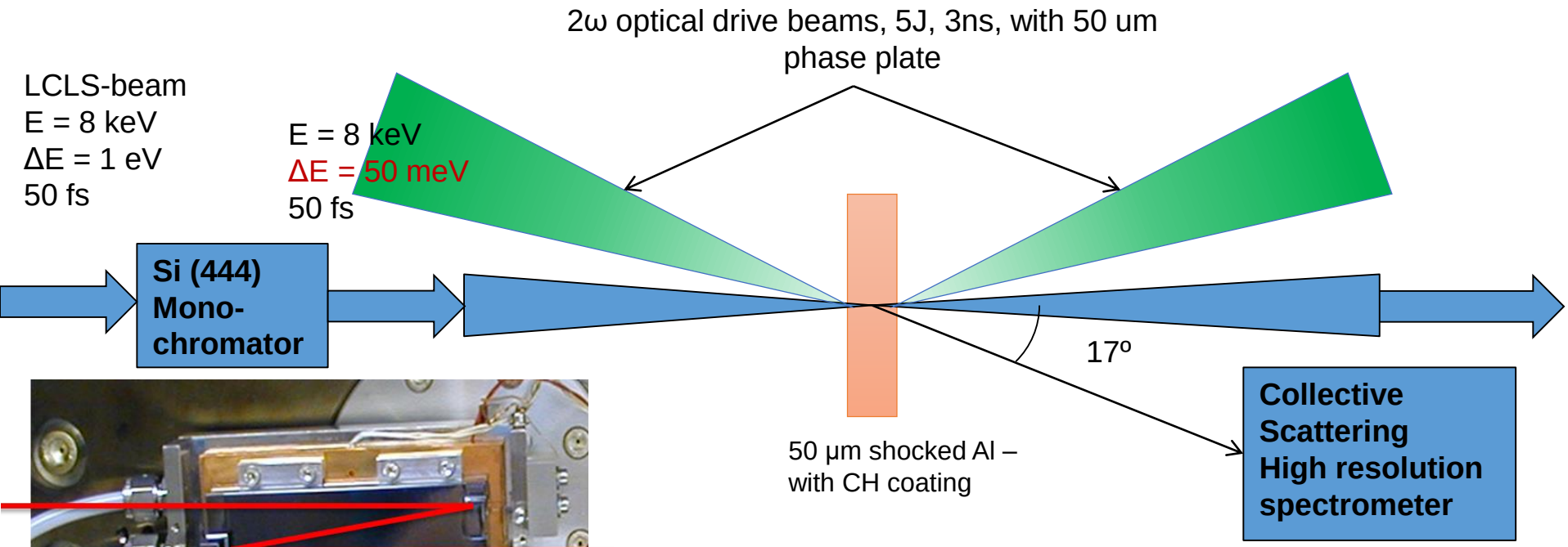

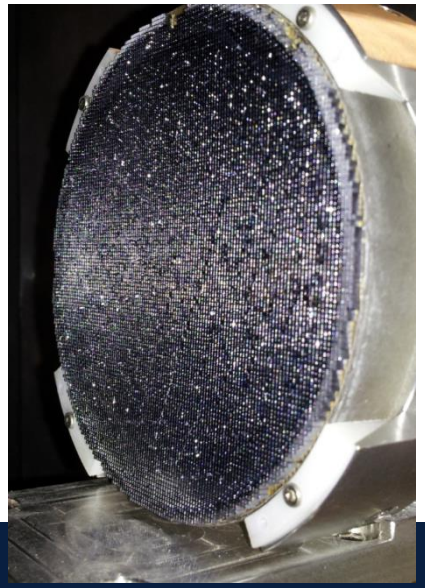

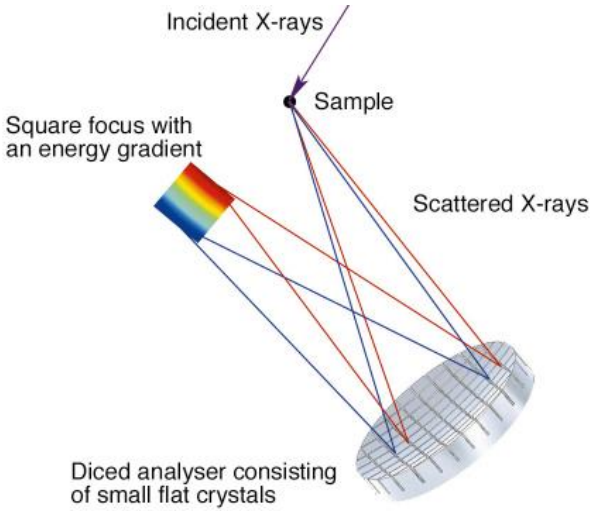

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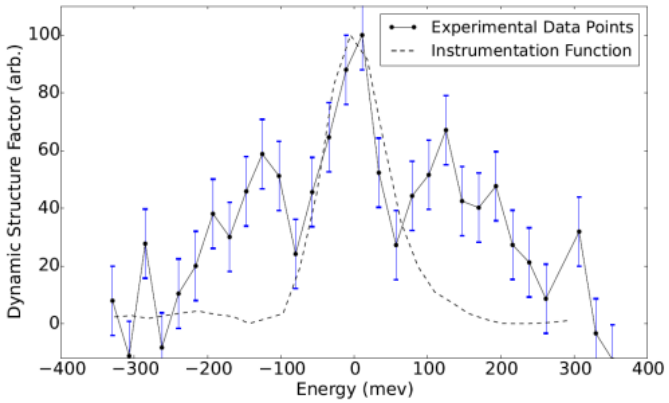
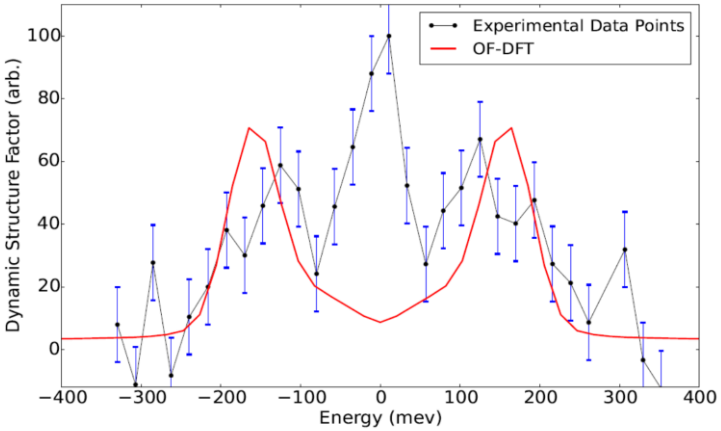


The first experimentally measured spectrum of ion-acoustic waves in a dense plasma. Signal is derived from ~ 10 shots ~ 100 photons. From this spectrum we are able to calculate a number of physics properties.

	Units	Fitted Result	95% Confidence
k	Å^{-1}	2.1	
γ		1.79	$1.18 - 2.39$
D_T	m^2/s	0.021	$0.003 - 0.040$
c_s	km/s	11.21	$10.26 - 12.3$
η	mPa s	1.71	$0.61 - 2.65$

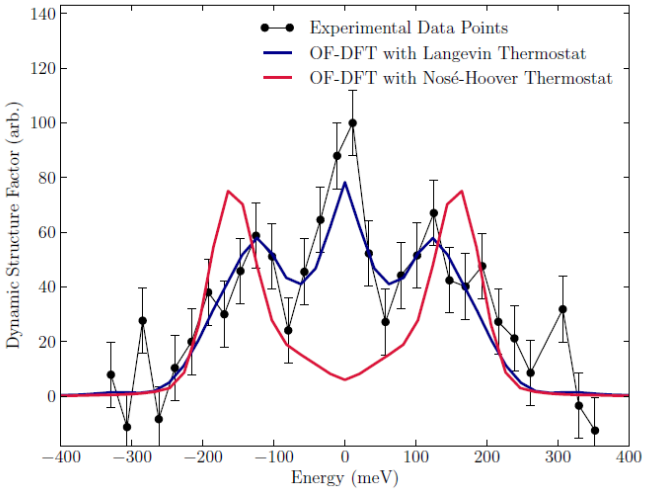


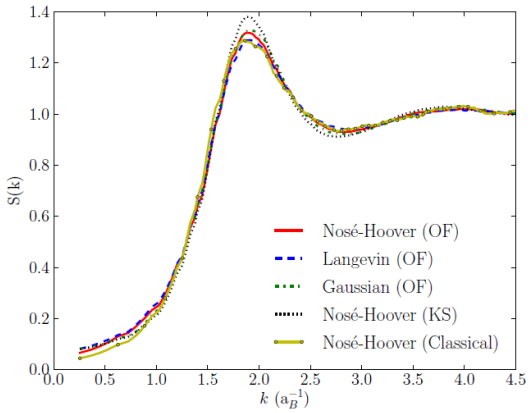
Comparison of the OF-DFT dynamic structure factor with the scattered spectrum correctly predicts the position of the two side peaks. This suggests that the strength of the inter-atomic potential is correct.

The central peak? (Mabey et al. Nat. Comms, 2017)

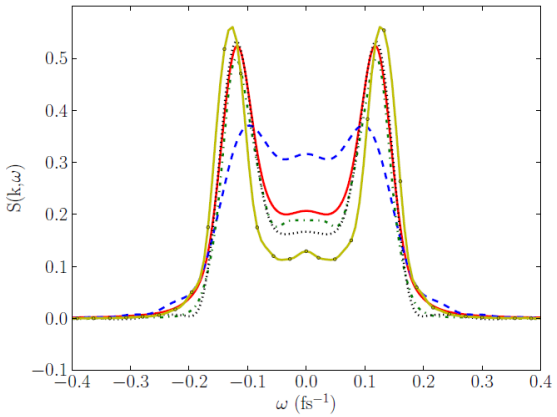
The appearance of the central peak Brillouin peak in the experimental results is not described by the current theory. Mabey *et al.* Nat. Comms 2017 suggests that the neglect of electron-ion interactions in the Born-Oppenheimer approximation can explain the missing central feature. This is currently an open question I will come back to later.

$$\boldsymbol{F} = m\boldsymbol{a} = -\nabla V(\boldsymbol{r}) - \sigma m v + \sqrt{2\sigma m k_b T} R(t)$$

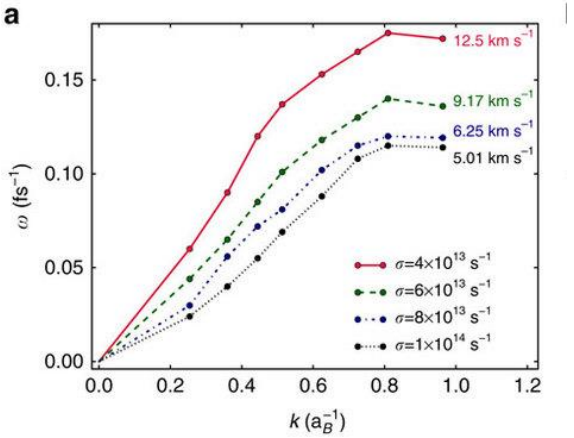




Thermodynamic variables stay the same (EOS)



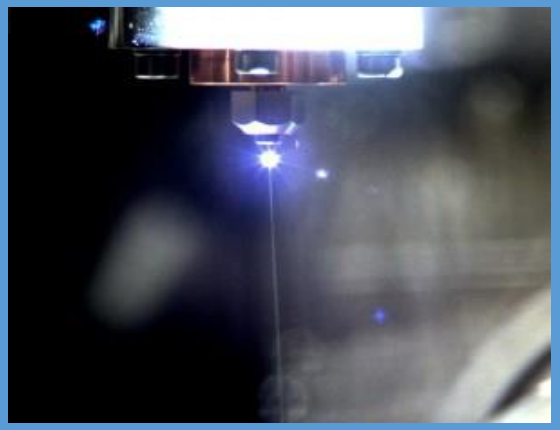
Transport variables change (Thermal diffusivity, viscosity, sound speed, stopping power, e-i equilibration)



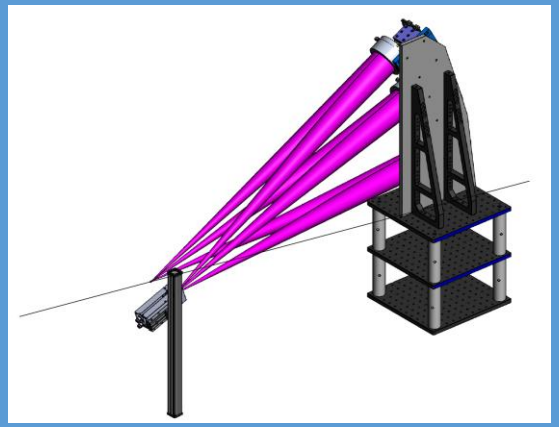
Current atomistic simulations neglect electron-ion collisions and do not correctly predict transport properties?

New Experimental Design

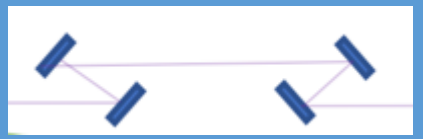
Higher Repetition Rate

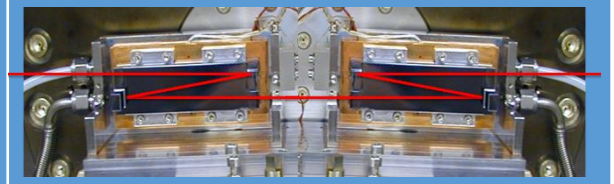


Multiple Analyzers



Simultaneous Diffraction/XRTS Measurement



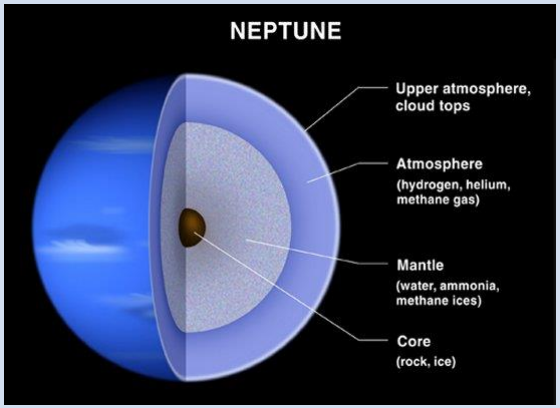


Lower Noise/Single Photon Counting

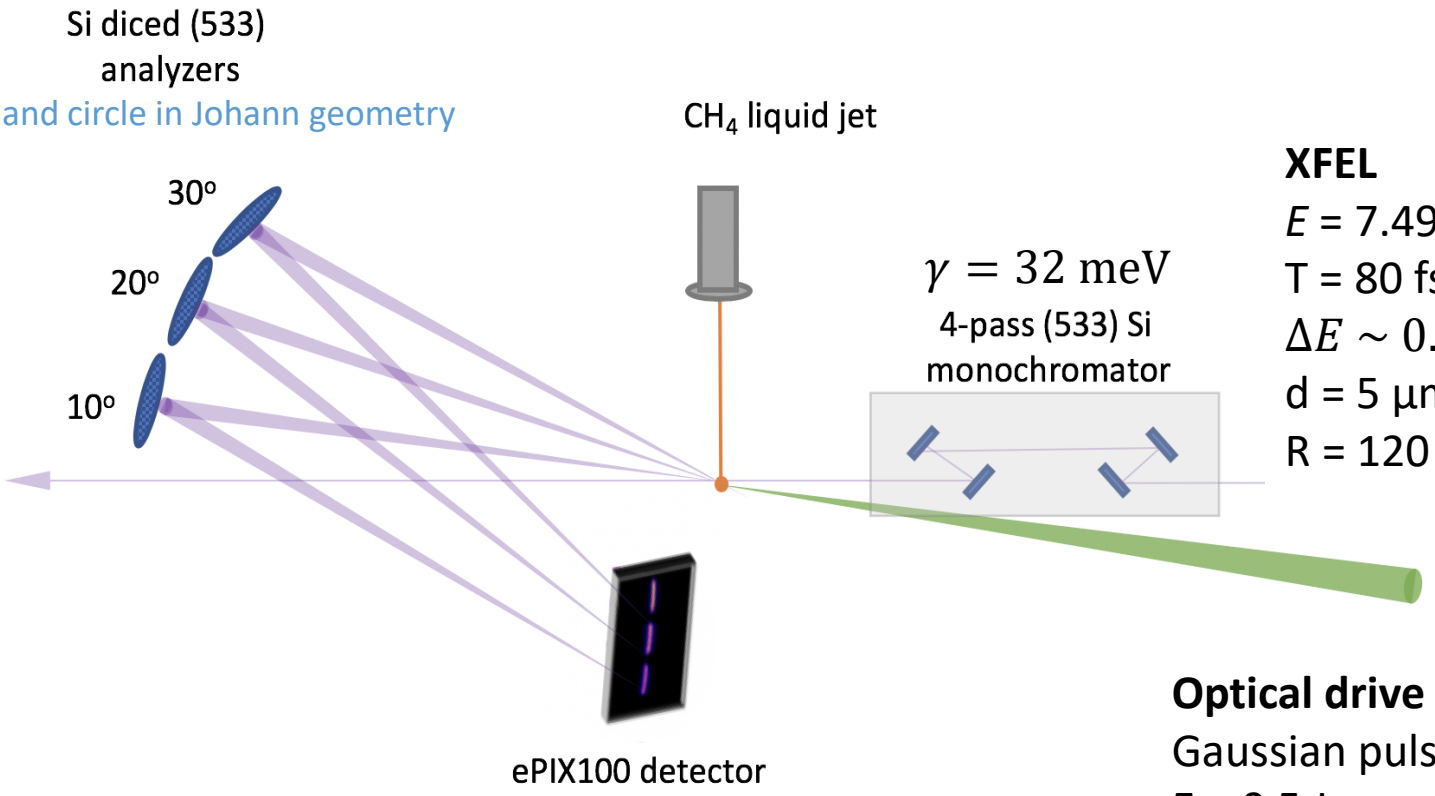
Higher resolution/less noise

Si(4,4,4)
↓
Si(5,3,3)

More Astrophysically Relevant



New Experimental Setup



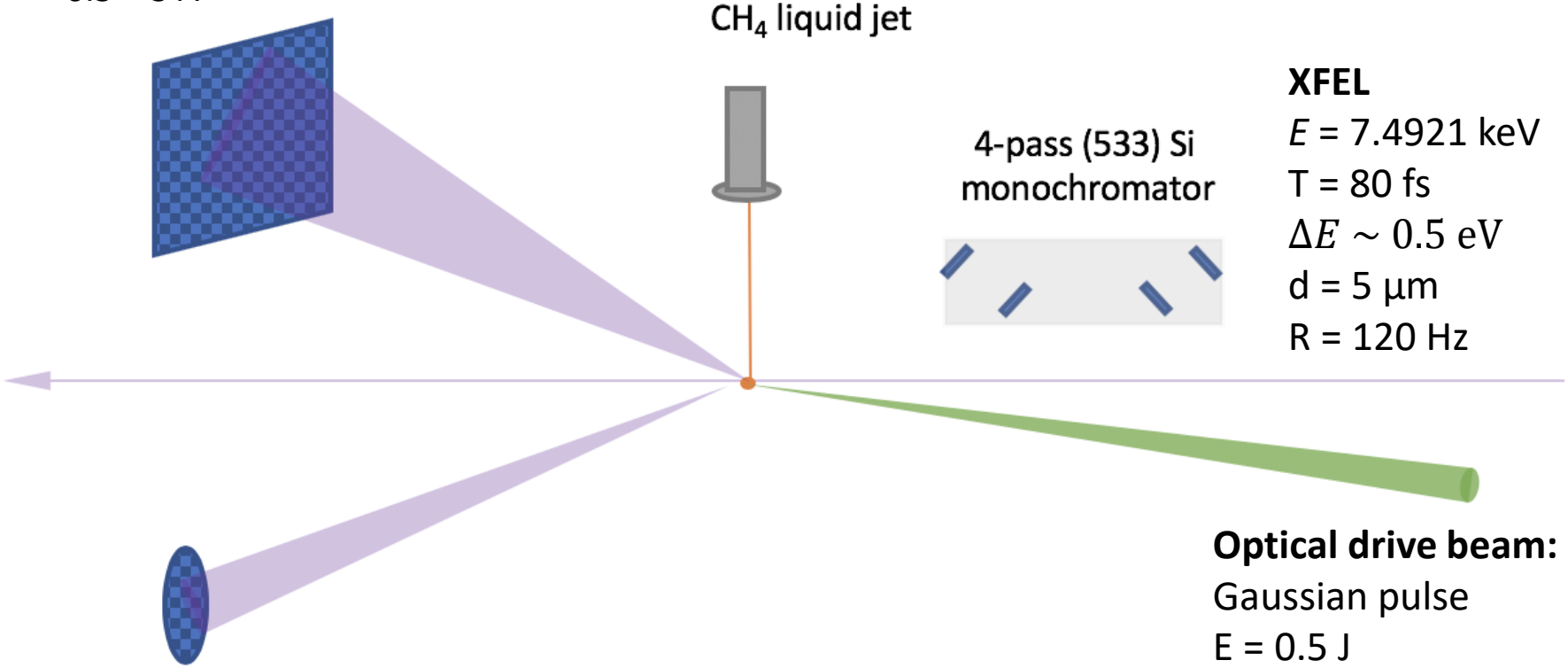
XFEL
E = 7.4921 keV
T = 80 fs
$\Delta E \sim 0.5$ eV
d = 5 µm
R = 120 Hz

Optical drive beam:
Gaussian pulse
E = 0.5 J
t = 150 ps
FWHM = 100 µm
λ= 800 nm
R=5 Hz

ePix (50 µm pixel size) &
diced Si(533) crystal with R=1
m & θ=87.5° $\Rightarrow$ ~50 meV @
7495 eV

New Experimental Setup

CSPAD:
$2\theta \sim 5^{o} - 55^{o}$
$0.5 - 3\ A^{-1}$



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d = 5 µm
R = 120 Hz

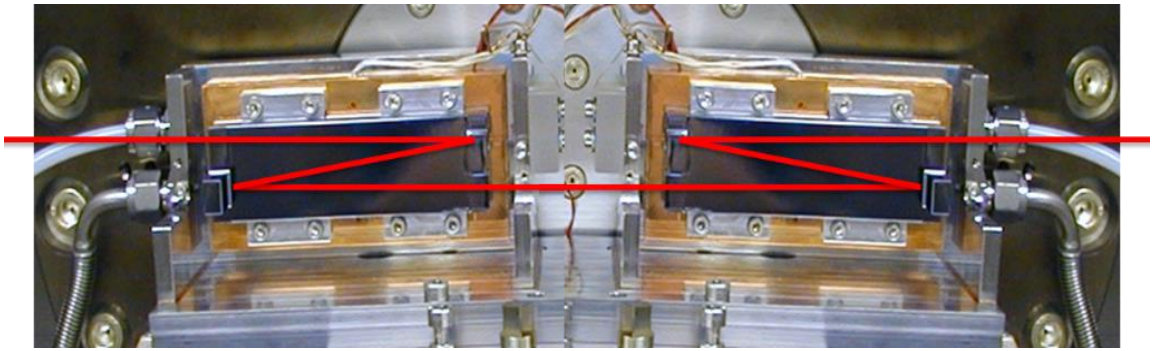
Optical drive beam:
Gaussian pulse
E = 0.5 J
t = 150 ps
FWHM = 100 µm
λ= 800 nm

XRTS

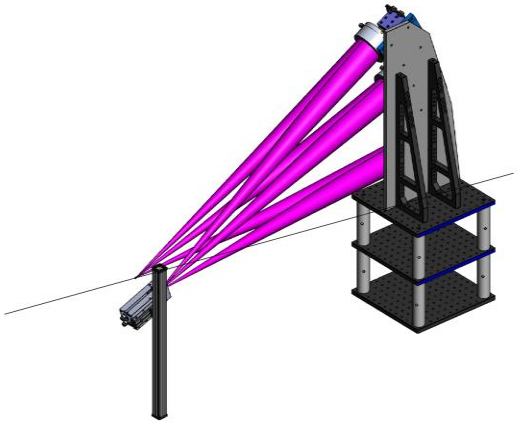
Achieving meV bandwidth

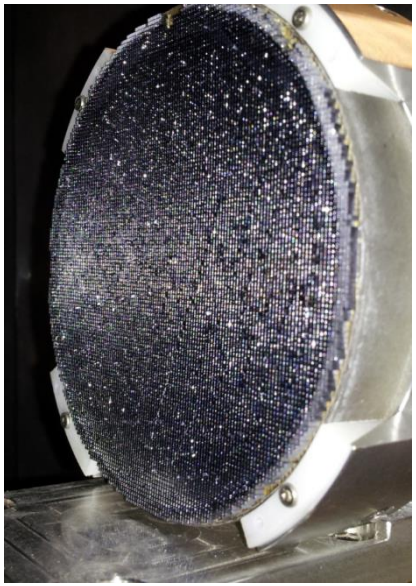
Resolving the dynamic structure factor requires meV resolution

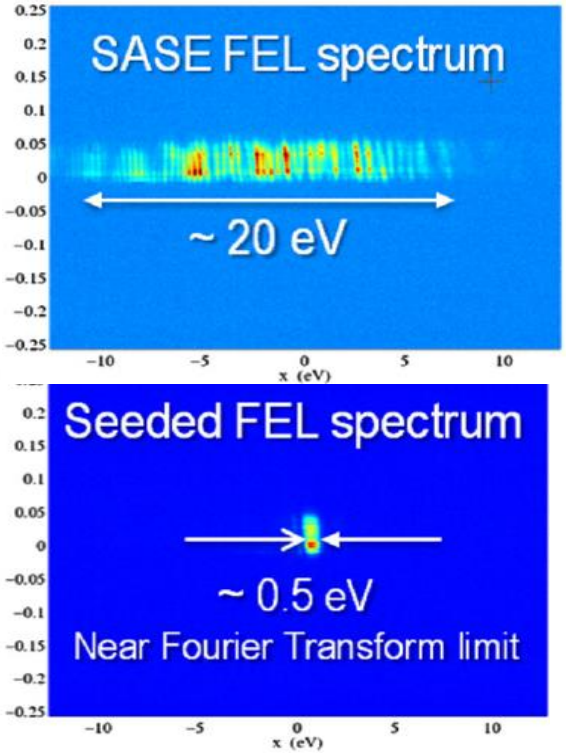
- Self seeded mode
- Double Monochromator
- High resolution X-ray analyzer using Si 533



Quadruple pass monochromator

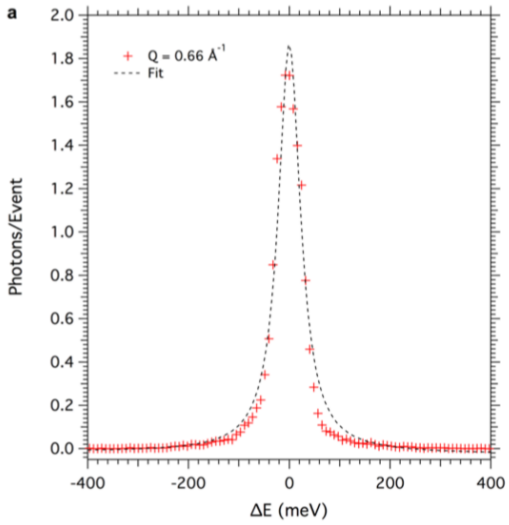






10.000 cubes of 0.7x0.7x2.3 mm^3 to enable collection of sufficient solid angle

Achieving meV bandwidth



Resolution measured from quasi-elastic scattering from 25 µm PMMA:

10° - Q = 0.66 A^{-1} $\Delta E = 53$ meV

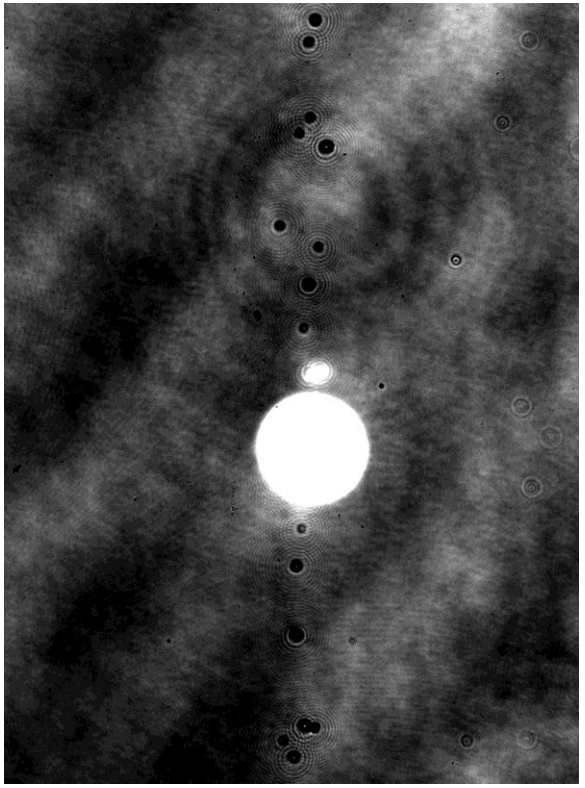
20° - Q = 1.32 A^{-1} $\Delta E = 61$ meV

30° - Q = 1.96 A^{-1} $\Delta E = 51$ meV

Approximate Photon Number

- SASE Beam ~ 10^{12}
- Seeded Beam ~ 10^{11}
- Monochromized Beam~ 10^{10}
- Scattered Photons~ 10^{4}
- Photons on analyser ~ 10
- Photons on detector ~ 1

Achieving higher rep. rates



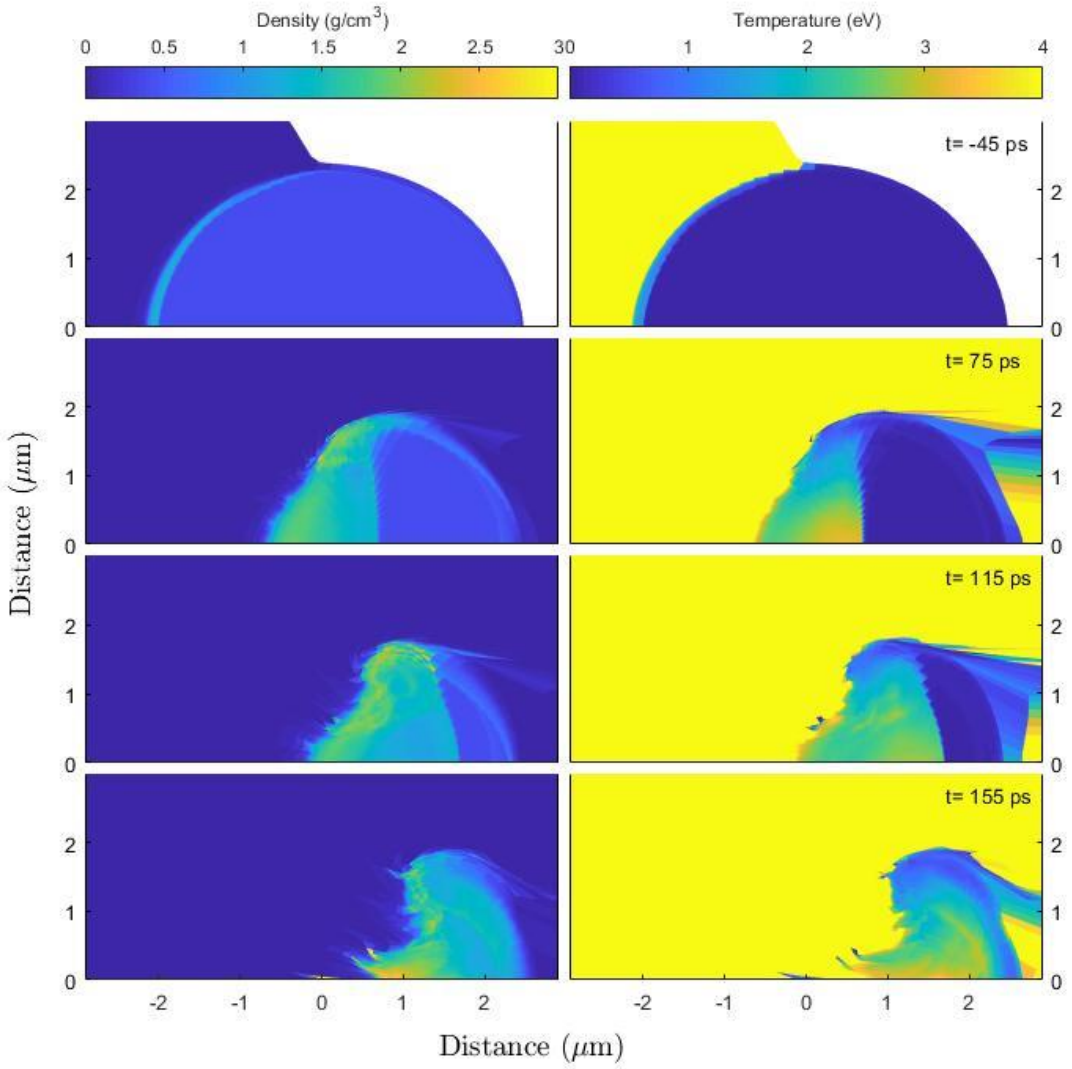
Diameter - 5 µm
Density – 0.4 g/cc
V = 50 – 100 m/s

+ Uncompressed Short Pulse Driver at 5 Hz

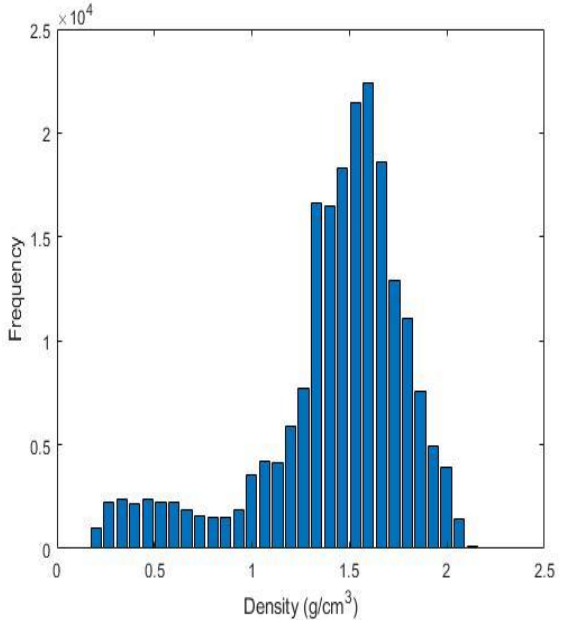
Using the EPIX and the Single photon counting code we are able to select photons in a narrow energy range reaching the detector

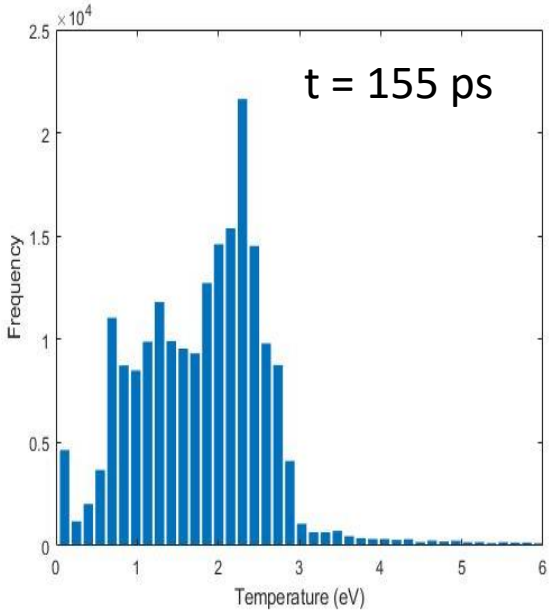
McBride RSI 2018

Expected Conditions

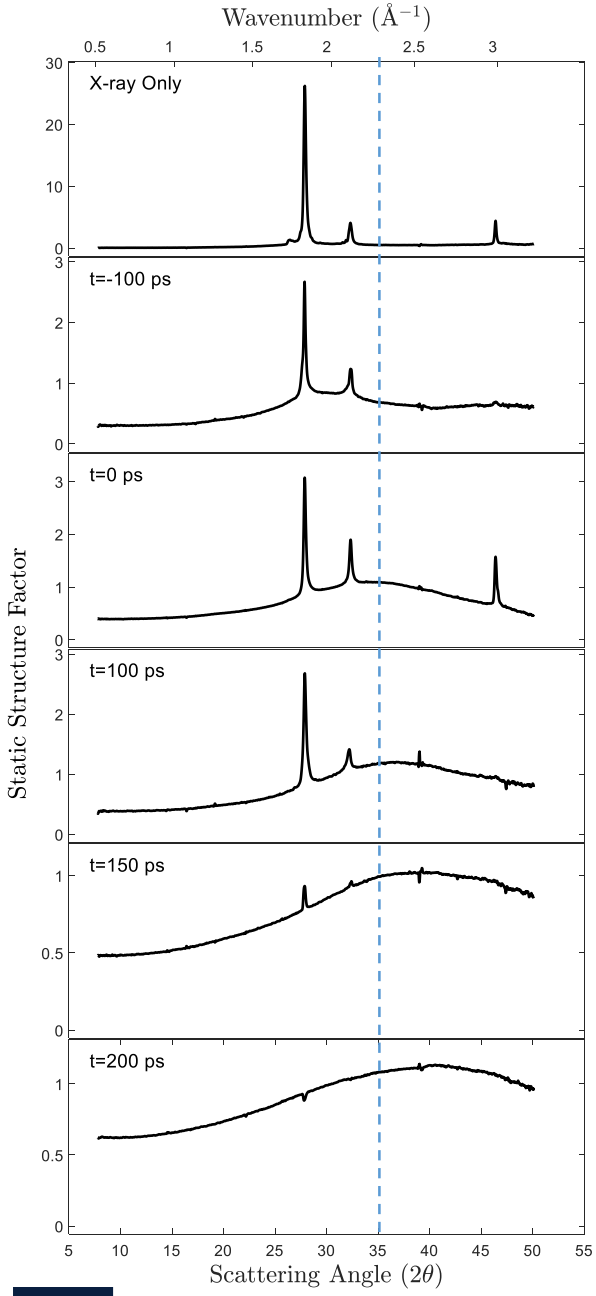


3D Radiative Hydrodynamic Simulations



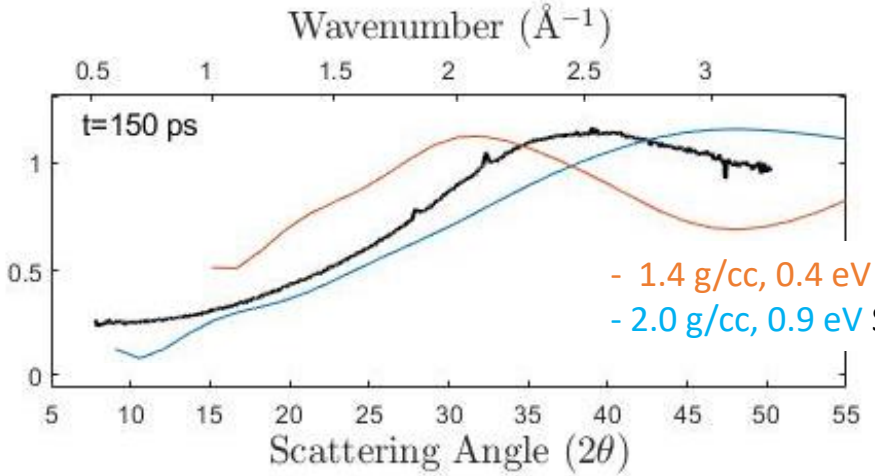


Results (X-Ray Diffraction)



- Figures show the static structure factor as measured on the CSPAD –between 2θ=5 and 2θ = 55 degrees.
- With no drive beam we see two clear Bragg peaks demonstrating the methane froze between leaving the jet nozzle and being probed.
- At early times we clearly see the coexistence of solid and liquid phases, the intensity of the Bragg peaks decreases and a liquid peak begins to appear.
- At late time the ion–ion correlation peak shifts to higher k suggesting further compression.
- Ideally, we would compare these structure factors to those calculated with an atomistic code. However, methane at these temperatures and densities is extremely hard to simulated due to strong bonds between the ions. i.e., OF-DFT fails in this regime.

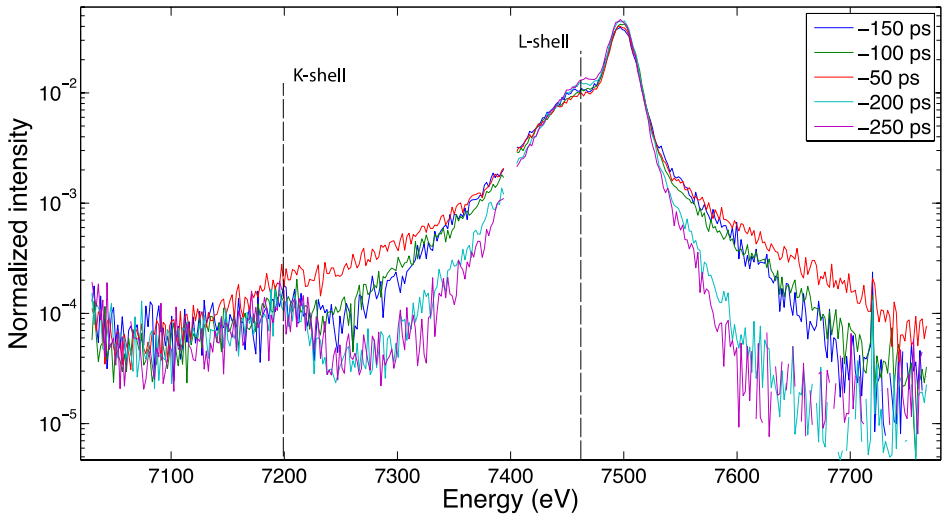
From DFT simulations we predict the density to be between 1.4 and 2.0 g/cc

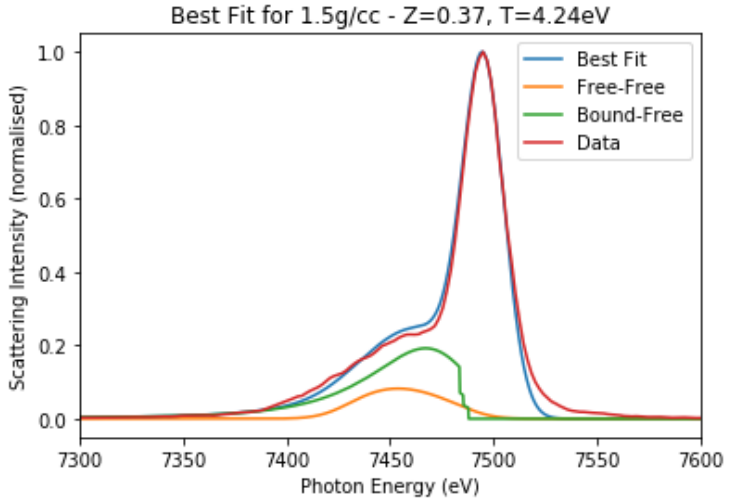


- 1.4 g/cc, 0.4 eV Li et al
- 2.0 g/cc, 0.9 eV Sherman et al

We are in the process of running our own DFT-MD simulations. Extremely time-consuming.

From XRTS we predict the temperature to be above 2 eV, but its poorly constrained





Non-collective regime $\alpha \sim 0.5$ Degeneracy parameter $\theta \sim 0.1$

Results: Ion Acoustic Spectrum

Dispersion relationship

We are in the process of running our own DFT-MD simulations. Extremely time-consuming.

Unfortunately, the peaks aren't as pronounced as in the aluminum data. However, one thing to notice is the central peak is still present.

Atomistic Simulations (work in progress)

Atomistic Simulations

DFT-MD simulations for the static structure factor are in progress. However, accurate prediction of the DSF requires large and long simulations that to undertake with Kohn-Sham DFT-MD would require too much computational power.

We are able to run OF-DFT simulations to predict sound speed/transport properties but it appears to not capture the bonds in warm dense methane i.e., the SSF does not appear correct.

Classical molecular dynamic simulations are easily run at the correct length/timescales but the accuracy of the interatomic potential at increased pressure/temperature is questionable.

Finally, both classical and quantum simulations work within the Born-Oppenheimer approximation and neither helps to addresses the question of whether non-adiabatic molecular dynamics is needed.

Fast Non-Adiabatic Dynamics of Many-Body Quantum Systems

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PRL 99, 185003 (2007) PHYSICAL REVIEW LETTERS week ending 2 NOVEMBER 2007

Excited Electron Dynamics Modeling of Warm Dense Matter

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CrossMark click for updates

Cite this: *Phys. Chem. Chem. Phys.*, 2016, **18**, 32466

Scaling relationships for nonadiabatic energy relaxation times in warm dense matter: toward understanding the equation of state

Ekadashi Pradhan,[a] Rudolph J. Magyar[b] and Alexey V. Akimov*[a]

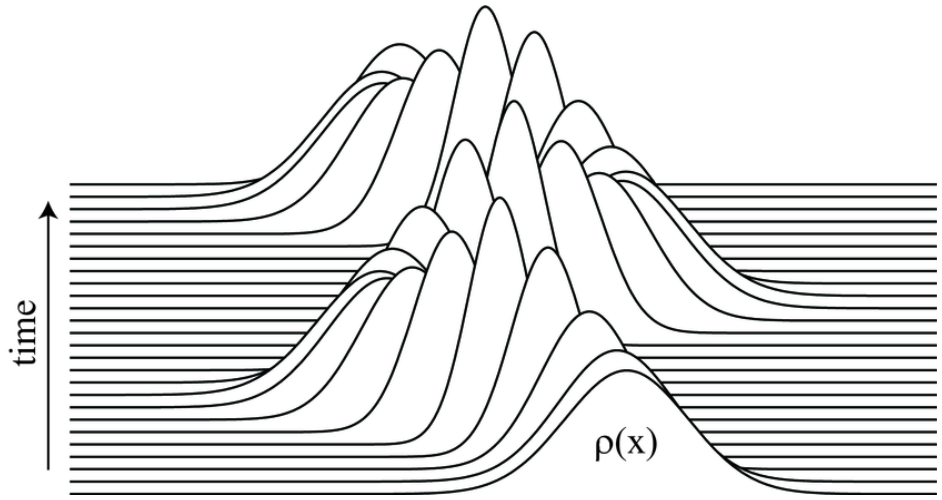
Wavepacket Molecular Dynamics (Electron Force Field)

- No Born Oppenheimer approximation.
- The system evolves according to Hamiltonian Dynamics. Where the electrons and ions are propagated forward in time **simultaneously**.

$$\mathbf{N}\dot{\mathbf{q}} = \frac{\partial H}{\partial \mathbf{q}}$$

$$N_{ab} = \frac{\partial}{\partial q_a^*}\frac{\partial}{\partial q_b}\ln\langle\Psi(\mathbf{q}^*)|\Psi(\mathbf{q})\rangle$$

$$\varphi_{k\alpha}(\mathbf{x}) = \left(\frac{3}{2\pi s_{k\alpha}^2}\right)^{3/4} \mathrm{e}^{-\left(\frac{3}{4s_{k\alpha}^2} - \frac{ip_{s_{k\alpha}}}{2\hbar s_{k\alpha}}\right)(\mathbf{x}-\mathbf{r}_{k\alpha})^2 + \frac{i}{\hbar}\mathbf{p}_{k\alpha}(\mathbf{x}-\mathbf{r}_{k\alpha})}$$



- Electrons described by a floating Gaussian wave packets.
- A Hartree product of the wave packets describes the wave function
- Added Pauli Potential due to the loss of explicit antisymmetry

Extremely Low Electron-ion Temperature Relaxation Rates in Warm Dense Hydrogen: Interplay between Quantum Electrons and Coupled Ions

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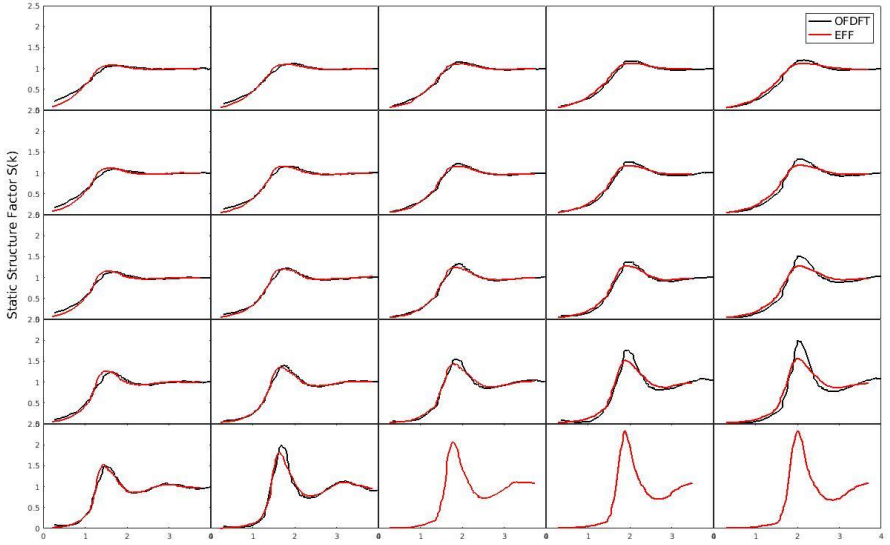
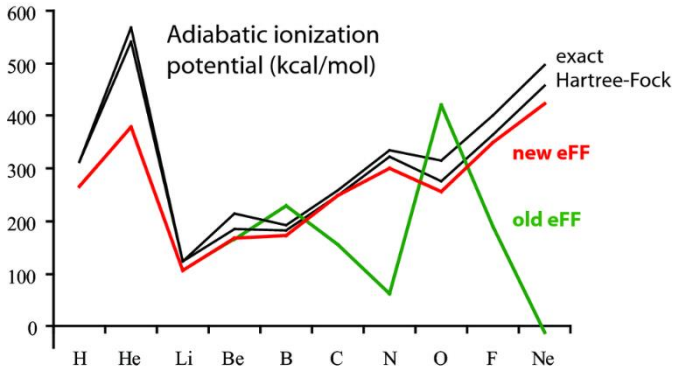
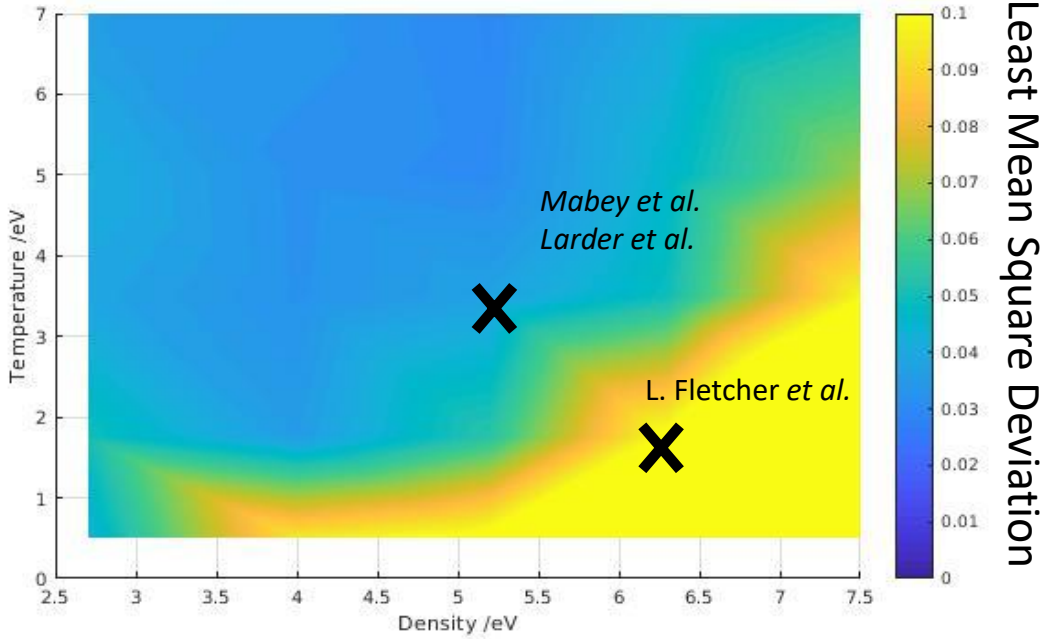
EFF requires benchmarking

Aluminum is well described by OF-DFT so we initially concentrated our simulation efforts here.

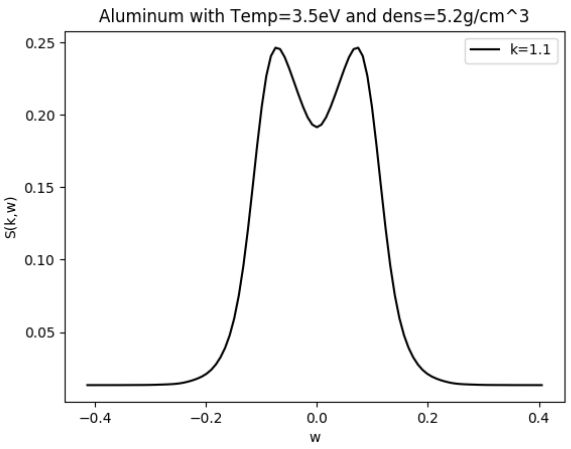
An initial comparison of SSF shows a region where EFF matches OFDFT if the effective core potential is modified over raw EFF.

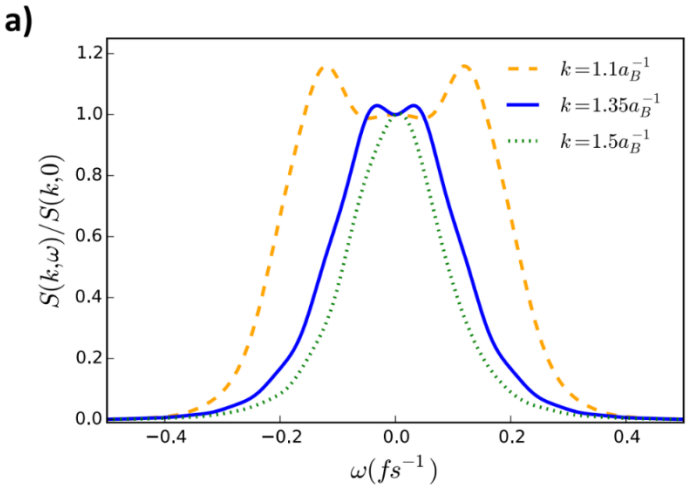




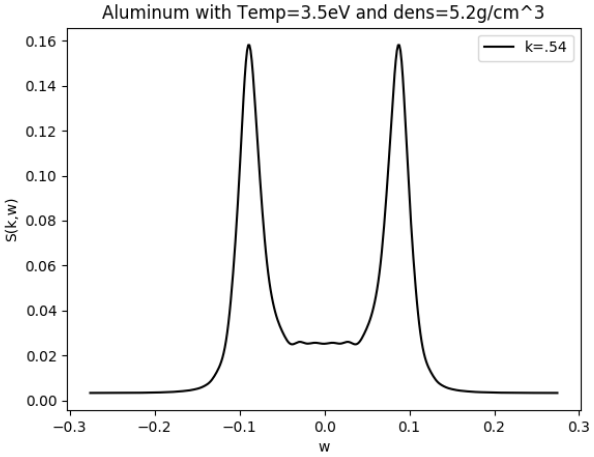


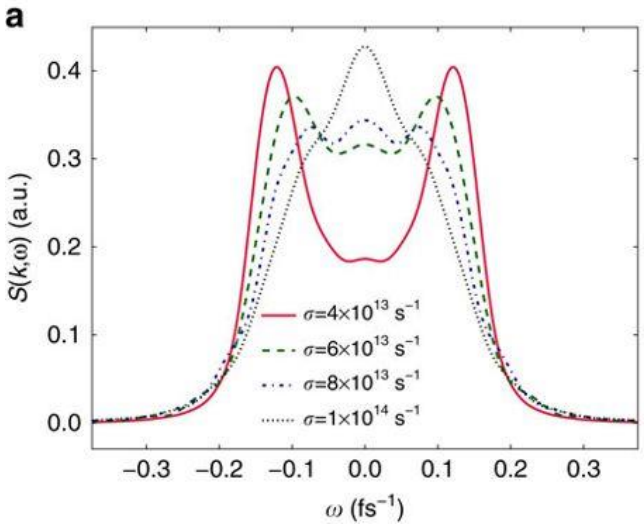
Dynamic Structure Factors produced from EFF simulations





The dynamic structure factor agrees with the Bohmian dynamics





The dynamic structure factor disagrees with Mabey et al. and the Langevin thermostat

It appears that neither non-adiabatic simulations exhibit a diffusive mode around ω = 0.

Conclusions

- Preliminary results of an experiment designed to measure the sound speed in warm dense methane.
- Created a WDM state through the laser ablation of a cryogenic liquid methane jet
- 7.49 keV, 32 meV bandwidth X-ray probe beam created using a four-pass silicon monochromator. The scattered photons were collected by a high-resolution X-ray spectrometer with an energy resolution of ~55 meV.
- Through the use of single photon counting codes, and by integrating over several thousand shots, we were able to clearly observe ion acoustic peaks in the dynamic structure factor and thus calculate a sound speed.
- Further improvements are needed before we can truly ascertain the transport properties.
- The simulations needed to described the experiment are complex.
- The applicability of the Born-Oppenheimer approximation is unknown.

Future work

- Simulate methane SSF/DSF using the EFF method and Kohn-Sham DFT.
- Complete the work on benchmarking EFF in aluminum.
- Apply for more experiments at LCLS 2 and European XFEL to get even better statistics.

Questions?

Thank you

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