High Energy Density Science with X-Ray Free Electron Lasers

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Outline of the Talk

- What we mean by ‘extreme conditions’
- Why we are interested in them
- Why XFELS are good for their creation/diagnosis.
- Underlying Physics –classical and non-ideal plasmas
- Isochoric heating and ionisation depression
- Shock waves and elastic scattering (solids)
- Shock waves and elastic scattering (plasmas)
- Inelastic scattering (plasmas)
- Isentropic compression
- Future prospects

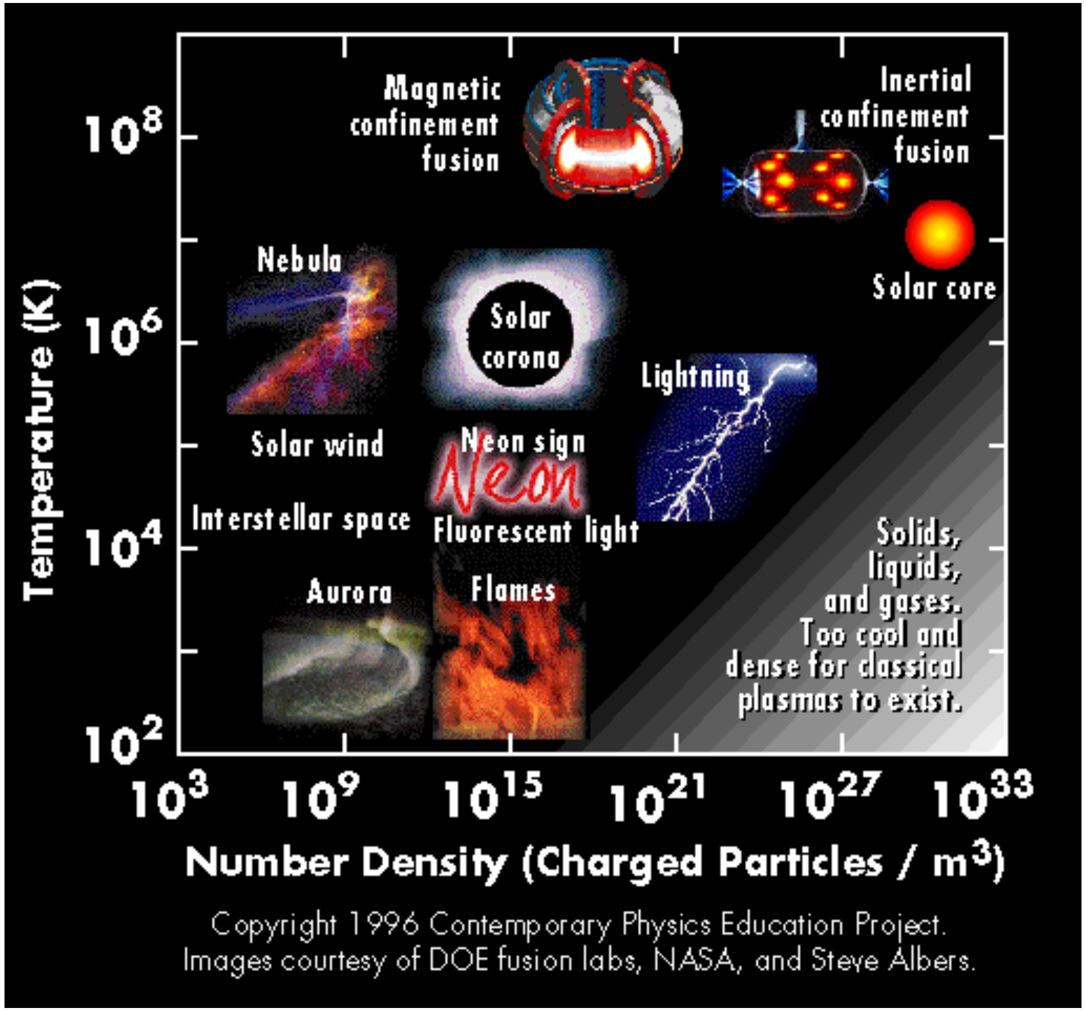
High Energy Density

- Simply by convention, HED science is taken to mean the region where the energy density > 10^{11} Jm^{-3}.
- Note this is about 0.6 eV $Å^{-3}$ – so heating solids to multi-eV temperatures.
- Also compression of solids – to halve the volume of a typical metal takes several Mbar. 1 Mbar is $10^{11}Nm^{-2}$ – so exactly the same energy density.

Level of Talk

- I am aiming this talk at those who do not know *any* plasma physics – so we will cover the basics.
- The goal is to teach at a very simple level the main ideas of ‘ideal’ plasmas, so that you see the difficulties that we have in understanding real ‘non-ideal’ dense plasmas.
- This is deliberately pitched as an undergraduate level talk.

Plasmas are wide-spread throughout the Universe



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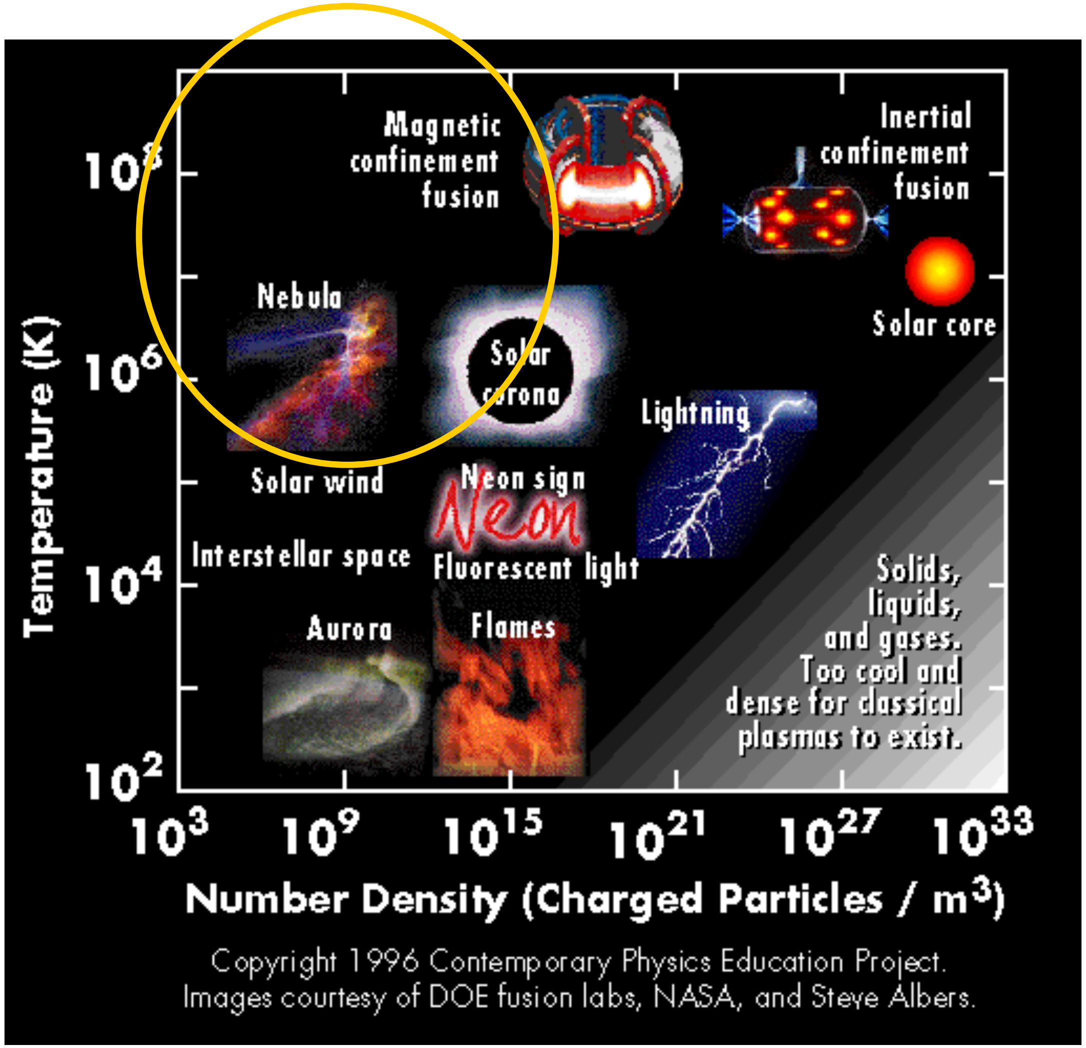
- Interior of large planets and white dwarfs
- Quark-gluon plasmas
- Nuclear matter
- Semiconductor devices and superconductors
- Laser-plasma experiments
- "Dusty plasmas"

“Good” Plasmas



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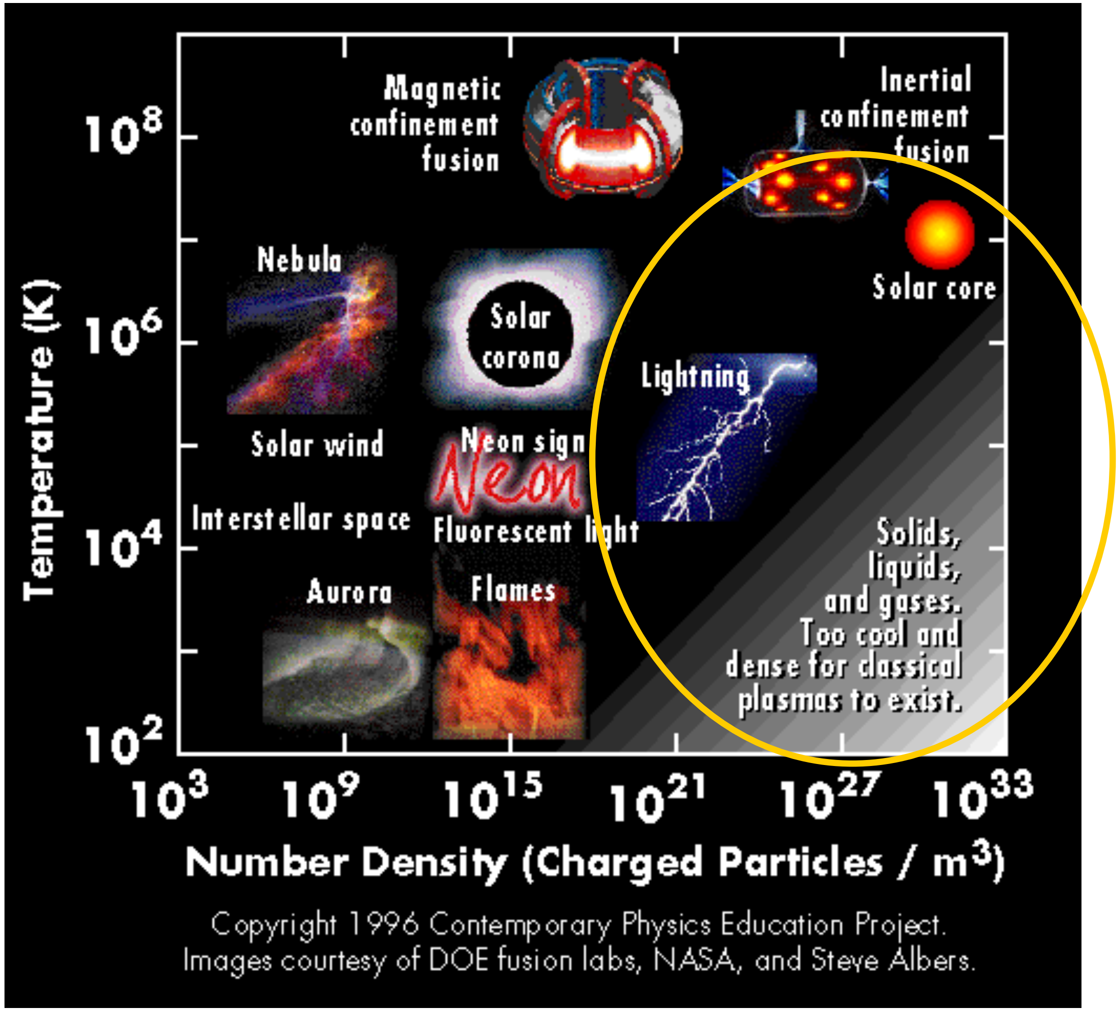
- “Standard” plasma physics covers the top left of the diagram - the plasmas are hot, and not very dense – we will see where this leads us in a moment.

“Bad” (non-ideal) Plasmas



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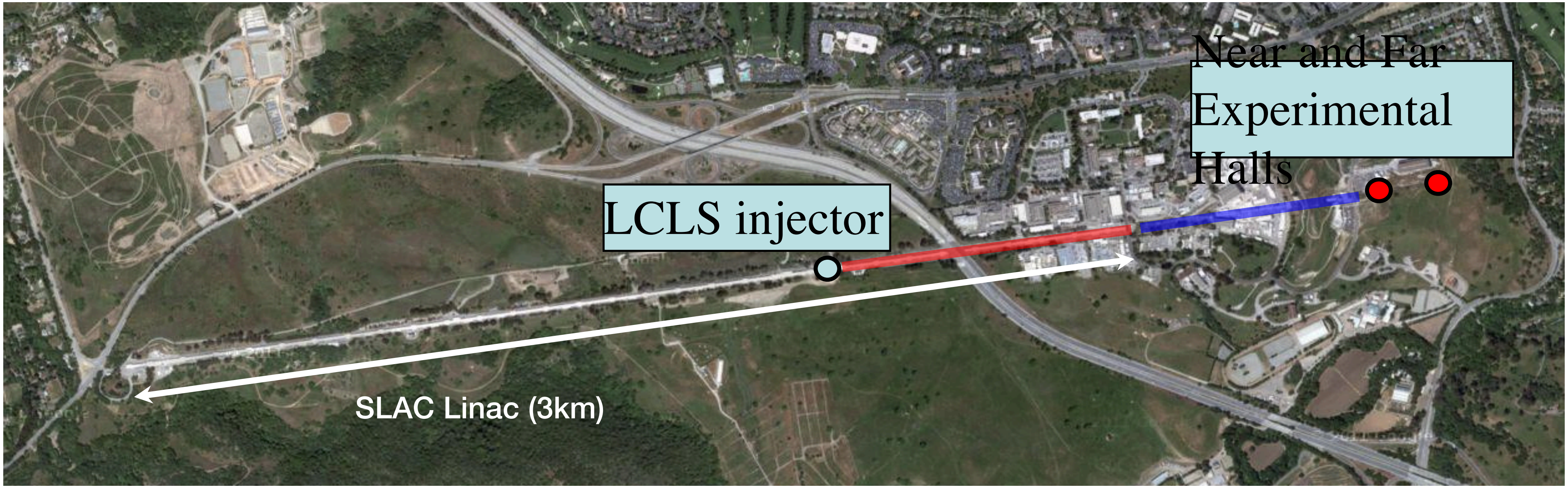
- Warm/cold dense plasma physics covers the bottom right of the diagram - the plasmas are colder and dense – here be dragons.
- This is what we are often interested in for XFEL research – more why later.

Linac Coherent Light Source (LCLS)



Photon energies: 500 eV – 10 keV (fundamental), 0.3 % bandwidth

Pulse lengths: <10 fs – 300 fs

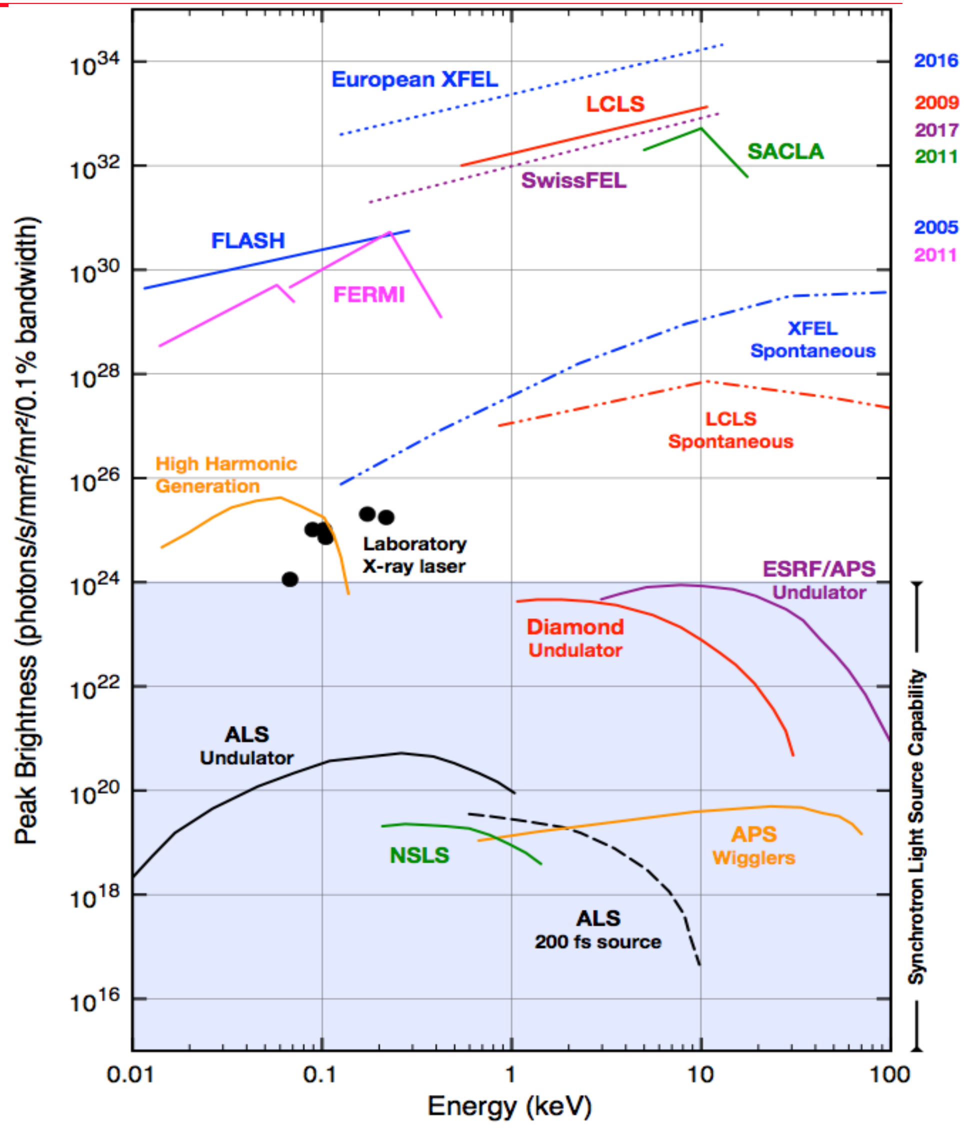
120 Hz, 1-2 mJ per pulse

4th generation light sources
Free-Electron Lasers

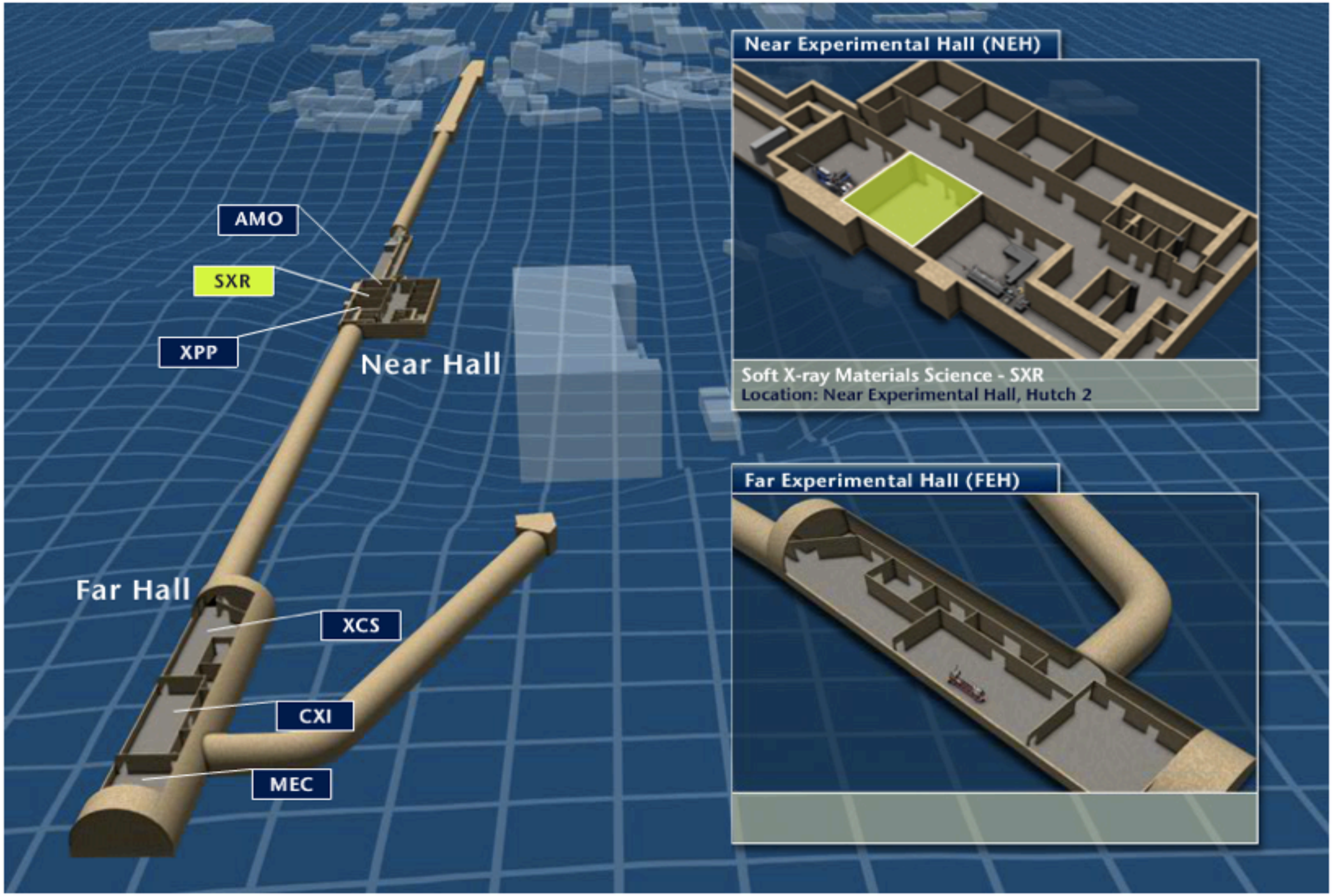
- Peak brightness 10^9 higher than 3rd generation synchrotrons

- short pulse duration (sub 100 fs);
- lots of photons per bunch (≥10^{12});
- tunable wavelength

- Hard X-ray FELs:
  - LCLS at SLAC, USA : 0.12-2.5 nm, **2009**
  - SACLA at SPring-8, Japan : 0.1 - 3.6nm, **2011**
  - European X-FEL at DESY, Germany: 0.1 - 6nm, **2017+**
  - Swiss-FEL at PSI : 0.1 - 7nm, **2016+**



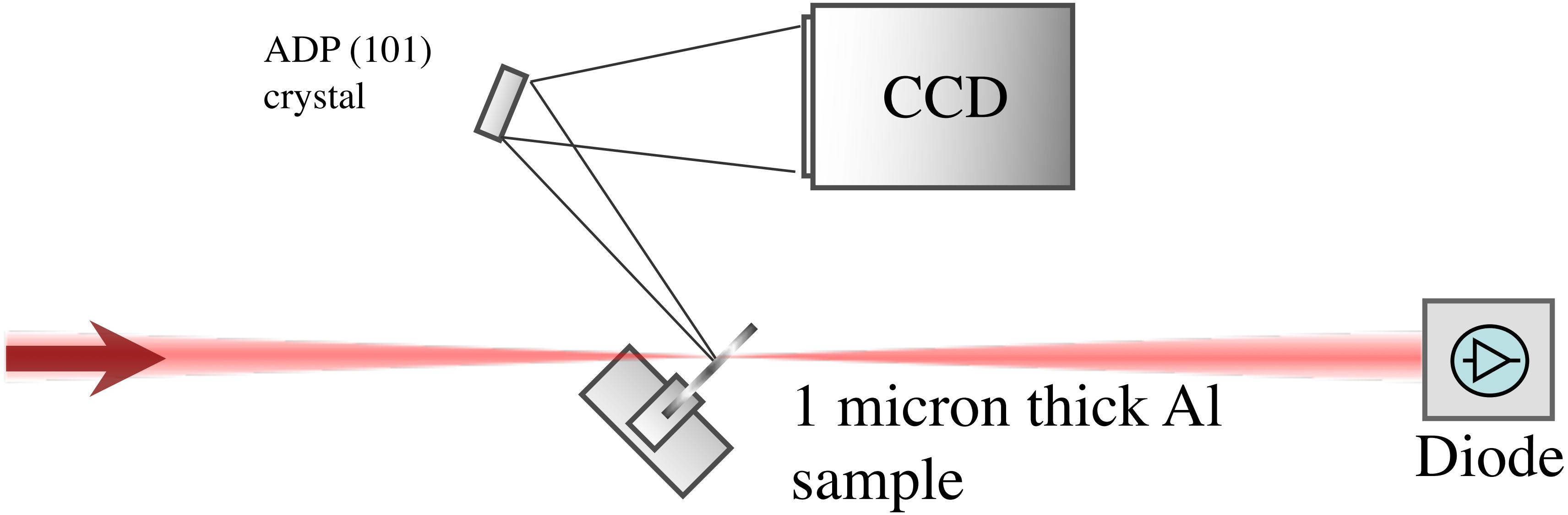
SXR Endstation



LCLS:SXR experimental setup

X-ray spectrometer: Al K-alpha emission

1460–1680 eV



LCLS pulse

Photon energy: 1560–1830 eVPulse length < 80 fs Pulse Energy ~1.5 mJBandwidth ~ 0.4%

Peak Intensity ~10^{17} W cm^{-2}

Typical numbers

- XFEL sources have 10^{12} photons per bunch, each photon with keV energies.
- Absorption lengths in solids ~ 1 micron.
- So in a 1 micron spot these photons are absorbed in about 10^{-18} m^{-3}.
- Density of solid is about 10^{29} m^{-3} – so 10^{11} atoms in the spot.
- So each atom can absorb a good fraction of 10 photons!
- Very hot plasma results (remember room temperature is 1/40th eV, 1 eV ~ 11,000 K).
- Energy goes into ionisation and heating – we have created 200 eV solid aluminum plasmas

What do we want to know?

- Our goal is to understand the thermodynamics, electrical, optical and transport properties.
  - Equation of state, heat capacity, conductivity (electrical and thermal): collision time, optical absorption/emisssion etc. etc. etc.
- Just the same as for any state of matter!

“Good” Plasmas

- In order to understand why warm dense plasmas are hard to model and understand, we first need to note that all classical plasma theory is based on the notion of a “Good” Plasma (WDM turns out not to be ‘good’).

- A “good” plasma is one where the thermal energy of the particles is large compared with their coulomb energy: it is weakly coupled (coupling is defined as the ratio of these energies - coulomb to kinetic).

- Obviously, we would not be far off if we said that the perfect gas equation of state was applicable (PV=RT), as for a perfect gas, we ignore interaction energy (and finite volume of the particles).

Some Basics

- We are going to look at 3 main parameters of a plasma (1) the basic ‘time-scale’, (2) the basic ‘length-scale’, and (3) a dimensionless number linking the two, and containing a huge amount of physics.
- (1) The basic ‘time-scale’ is the plasma frequency.
- (2) The basic ‘length-scale’ is the Debye Length.
- (3) The dimensionless number – the plasma parameter – represents the number of particles in a sphere of radius the Debye-length – but it contains much more physics than that!

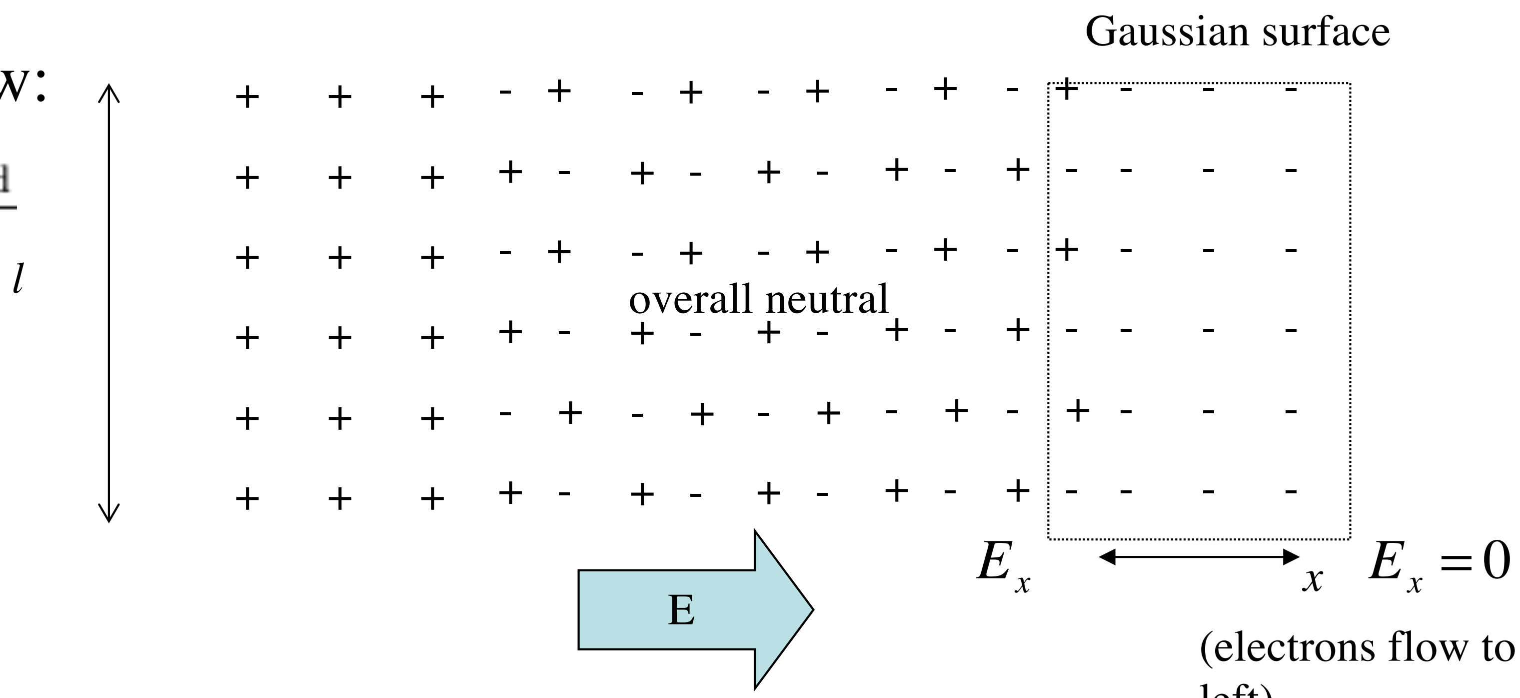
Plasma Oscillations

- Ions are heavy compared with electrons - assume they are stationary
- Displace the electrons from the ions and 'let go' - the resulting oscillation occurs at what is known as the plasma frequency
- Note we assume no collisions take place
- We also assume that the electrons are cold - their motion is due to the electrostatic restoring force - we ignore thermal motion

Plasma Frequency

Apply Gauss’ law:

$$\oint_S \mathbf{E} \cdot \mathrm{d}\mathbf{A} = \frac{Q_{\mathrm{enclosed}}}{\varepsilon_0}$$



(electrons flow to left)

$$El^2 = \frac{nel^2 x}{\varepsilon_0}$$

$$E = \frac{nex}{\varepsilon_0}$$

$$\therefore F = m\frac{\mathrm{d}^2 x}{\mathrm{d}t^2} = -eE = -\frac{ne^2 x}{\varepsilon_0}$$ Equation of motion

Oscillation at frequency $\omega_p^2 = \dfrac{ne^2}{\varepsilon_0 m}$

Typical plasma frequencies

Useful aide memoire : $f_p \sim 9000\ n^{1/2}$ (n in cm^{-3})

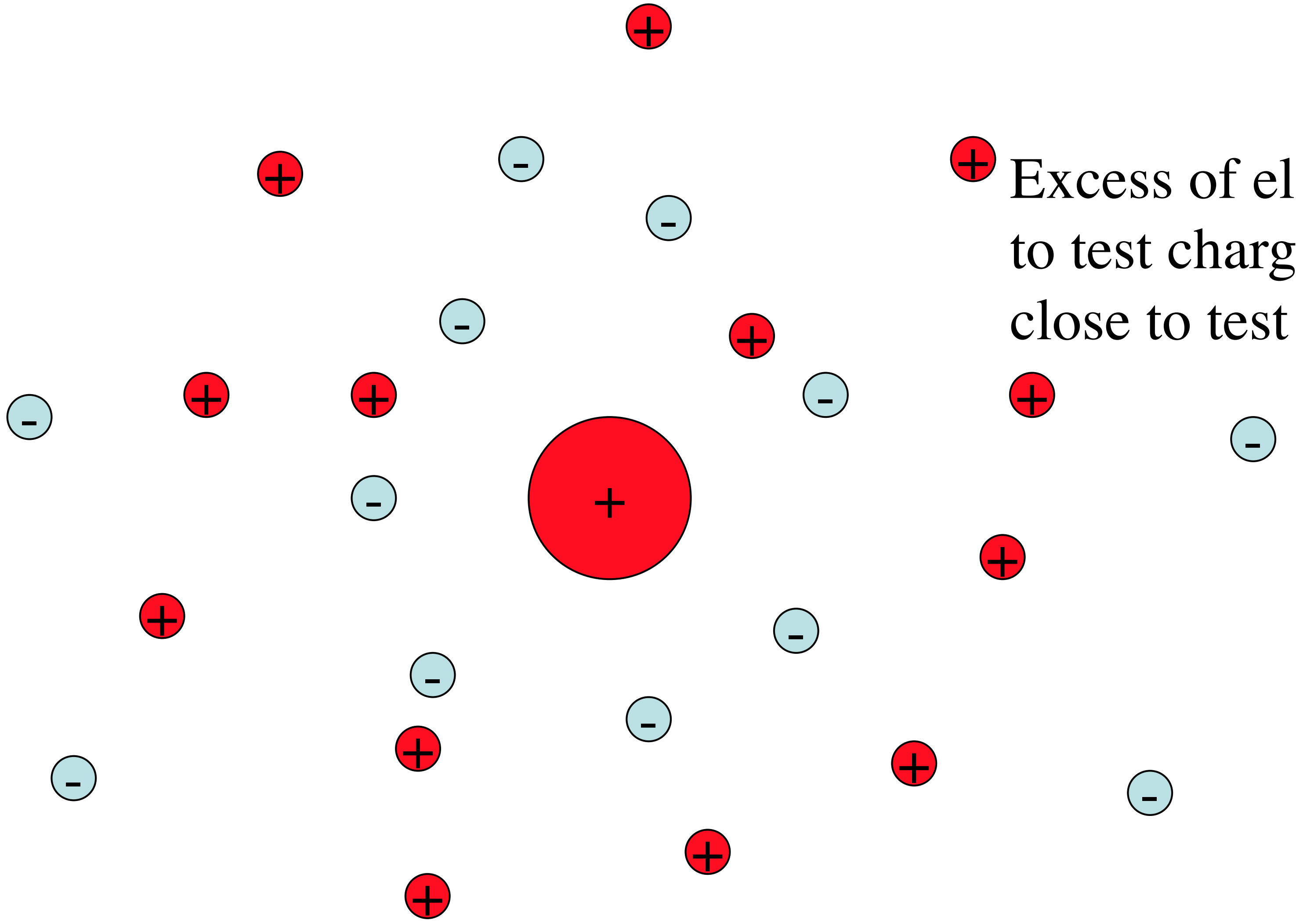
Ionosphere , $n \sim 10^4$ cm^{-3} $f_p \sim 1$ MHz

Tokamak $n \sim 10^{12}$ cm^{-3} $f_p \sim 10$ GHz

Laser plasma $n \sim 10^{21}$ cm^{-3} $f_p \sim 3 \times 10^{14}$ Hz (visible – few eV)

Solid metal $n \sim 10^{23}$ cm^{-3} $f_p \sim 3 \times 10^{15}$ Hz (VUV – few - tens eV)

Length scales - Debye Shielding(1)



Excess of electrons close to test charge, deficit of ions close to test charge.

Debye Shielding (2)

Assume potential at a distance *r* from the test charge is $\phi(r)$

$$n_e(r) = n_0 \exp\left(\frac{e\phi(r)}{kT}\right) \approx n_0\left(1 + \frac{e\phi(r)}{kT}\right)$$

Therefore there is an excess of electrons of order

$$n_e^+(r) \approx n_0 \frac{e\phi(r)}{kT}$$

By similar reasoning there is a deficit of ions at the same position

$$n_i^-(r) \approx -n_0 \frac{e\phi(r)}{kT}$$

Debye Shielding (3)

Therefore the excess charge density at the point *r* is given by

$$\rho(r) = e(n_i^-(r) - n_e^+(r)) \approx -2n_0 \frac{e^2\phi(r)}{kT}$$

For self consistency the potential itself is related to the charge density by Poisson’s equation

$$\nabla^2\phi(r) = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\phi}{\partial r}\right) = -\frac{\rho(r)}{\varepsilon_0} = \left(\frac{2n_0e^2}{\varepsilon_0 kT}\right)\phi(r)$$

Debye Shielding: solution

$$\phi(r) = \frac{A}{r}\exp\left(-\frac{\sqrt{2}r}{\lambda_D}\right)$$

where the Debye length is given by

$$\lambda_D = \sqrt{\frac{\varepsilon_0 kT}{ne^2}} \qquad k_D = 1/\lambda_D$$

$$\phi(r) = \frac{Q}{4\pi\varepsilon_0 r}\exp\left(-\frac{\sqrt{2}r}{\lambda_D}\right)$$

Plasma Parameter

A 'good' plasma is one that has a large number of particles within a Debye Sphere:

$$N_D = n_0 \frac{4}{3} \pi \lambda_D^3$$

Also note that the Debye length is the typical distance that a thermal electron travels during a plasma period…

$$\lambda_D = \sqrt{\frac{\varepsilon_0 kT}{ne^2}} = \sqrt{\frac{kT}{m}} \sqrt{\frac{\varepsilon_0 m}{ne^2}} = \frac{v_{th}}{\omega_p}$$

Large Plasma Parameter – Weak Coupling

A ‘good’ plasma is one that has a large number of particles within a Debye Sphere:

$$N_D = n_e \frac{4}{3}\pi\lambda_D^3 = n_e \frac{4}{3}\pi\left(\frac{\varepsilon_0 k_B T}{n_e e^2}\right)^{3/2}$$

$$= \frac{4}{3}\pi\left(\frac{n_e^{2/3}\varepsilon_0 k_B T}{n_e e^2}\right)^{3/2} = \frac{4}{3}\pi\left(\frac{\varepsilon_0 k_B T}{n_e^{1/3} e^2}\right)^{3/2}$$

$$\approx \left(\frac{4\pi\varepsilon_0 r k_B T}{e^2}\right)^{3/2}$$

r is the mean distance between particles.

So….The plasma parameter is also a measure of the thermal energy to the Coulomb energy! We said this had to be large for our analysis to work in the first place!

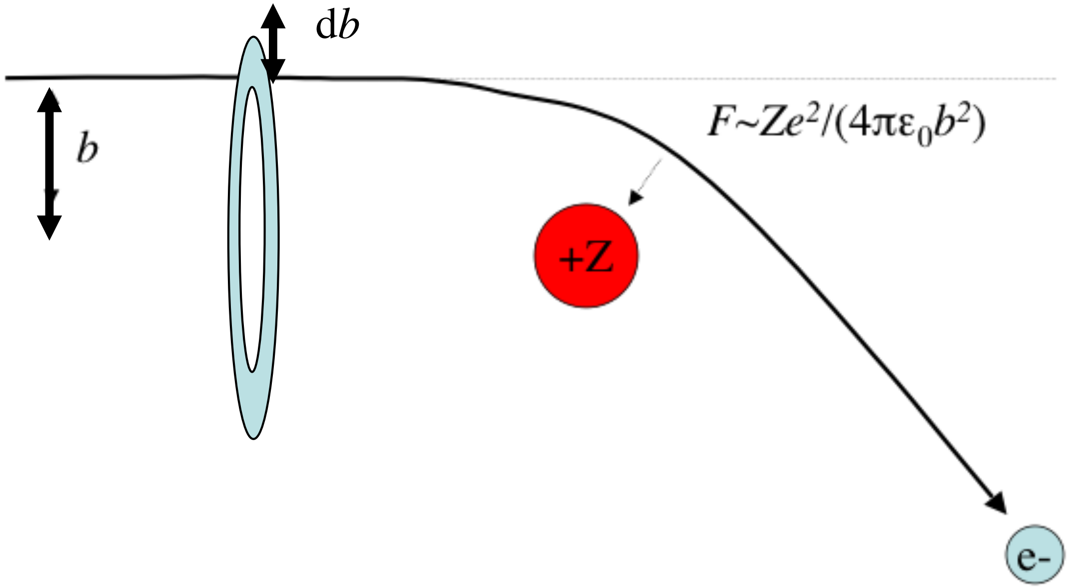
Collisions in a Plasma

I'm going to skip the next few slides in the lecture itself, but include them here for you to read at your leisure.

The important point is that the plasma parameter (the number of particles in a Debye sphere) also tells us the collision time - the electrons undergo lots of small angle scattering events, and get deflected by about $\pi/2$ radians after about the plasma period multiplied by the plasma parameter.

'Good' plasmas are often called 'collisionless'.

Electron-Ion Collisions



Electron feels a force of ~

$$\frac{Ze^2}{4\pi\varepsilon_0 b^2}$$

for a time of order ~

$$\Delta t \approx \frac{2b}{v}$$

Thus

$$\boxed{\Delta v \approx \frac{Ze^2}{2\pi\varepsilon_0 bvm}}$$

$$F = \frac{Ze^2}{4\pi\varepsilon_0 b^2} = m\frac{\Delta v}{\Delta t} = m\Delta v\frac{v}{2b}$$

Collision Time (1)

- Typically the deflection angle in a *good plasma* is small - this leads us to ask what is a collision?
- A collision corresponds to scattering the particle through 90 degrees, by lots of random small-angle collisions.
- To calculated this, we work out how quickly $(\Delta v)^2$ changes - and say a collision has taken place when the rms value of Δv has changed by v.

Collision Time (2)

For a given collision with impact parameter b $\quad (\Delta v)^2 = \dfrac{Z^2 e^4}{4\pi^2 \varepsilon_0^2 m^2 b^2 v^2}$

How quickly does this change? Well the rate of encounters with ions is $n\sigma v$, where σ is the cross section: $2\pi b \mathrm{d}b$

$$\frac{\mathrm{d}\left\langle (\Delta v)^2 \right\rangle}{\mathrm{d}t} = \int 2\pi b \,\mathrm{d}b n_i v (\Delta v)^2$$

$$= \int \frac{n_i Z^2 e^4 \mathrm{d}b}{2\pi \varepsilon_0^2 m^2 v b}$$

What are the limits on the integral?

Limits on the Integral

- An electron cannot have an infinitely large impact parameter - eventually it does not ‘feel’ the ion due to Debye shielding - the upper limit of the integral is the Debye length.
- Our simple approach gives infinite change in velocity for a head-on collision - this is not correct - we know that in this case the change in velocity is of order the velocity. Thus

$$b_{\max} = \lambda_D = \sqrt{\frac{\varepsilon_0 k_B T}{n_e e^2}} \qquad\qquad b_{\min} \approx \frac{Ze^2}{4\pi\varepsilon_0 m v^2}$$

The Coulomb Logarithm

$$\frac{\mathrm{d}\left\langle (\Delta v)^2 \right\rangle}{\mathrm{d}t} = \int_{b_{\min}}^{\lambda_D} \frac{n_i Z^2 e^4 \mathrm{d}b}{2\pi\varepsilon_0^2 m^2 v b} = \frac{n_i Z^2 e^4}{2\pi\varepsilon_0^2 m^2 v} \ln\Lambda$$

Where lnΛ is known as the Coulomb logarithm:

$$\Lambda = \frac{\lambda_D}{b_{\min}} = \sqrt{\frac{\varepsilon_0 k_B T}{n_e e^2}}\,\frac{4\pi\varepsilon_0 m v^2}{Z e^2}$$

which, assuming the velocity is of order the thermal velocity, can be written

$$\boxed{\Lambda \approx \frac{12\pi n_e \lambda_D^3}{Z} \approx \frac{9 N_D}{Z}}$$

Typical numbers for the Coulomb logarithm range from 1 – 30 but it becomes meaningless for ‘bad’ plasmas as it can go negative!.

Collision Time (3)

During a collision time, τ, the change in the velocity is of order the velocity, i.e.

$$\frac{1}{\tau}v^2 = \frac{n_i Z^2 e^4}{2\pi\varepsilon_0^2 m^2 v}\ln\Lambda$$

$$\therefore \tau = \frac{2\pi\varepsilon_0^2 m^2 v^3}{n_i Z^2 e^4 \ln\Lambda}$$

When we average over the velocities present in a Maxwellian -

$$\tau \approx 6.4\frac{2\pi\varepsilon_0^2 m^2 (k_B T/m)^{3/2}}{n_i Z^2 e^4 \ln\Lambda}$$

Collisionless Plasmas (1)

- Earlier on we said that we could ignore collisions when deriving the plasma frequency - we said that there were many oscillations of the electrons on the timescale of a collision - we are now in a position to prove this for a good plasma.
- We need to show that $\omega_{pe}\tau >> 1$

Collisionless Plasmas (2)

$$\omega_{pe}\tau = \sqrt{\frac{n_e e^2}{\varepsilon_0 m}}6.4\frac{2\pi\varepsilon_0^2 m^2 (k_B T/m)^{3/2}}{n_i Z^2 e^4 \ln\Lambda}$$

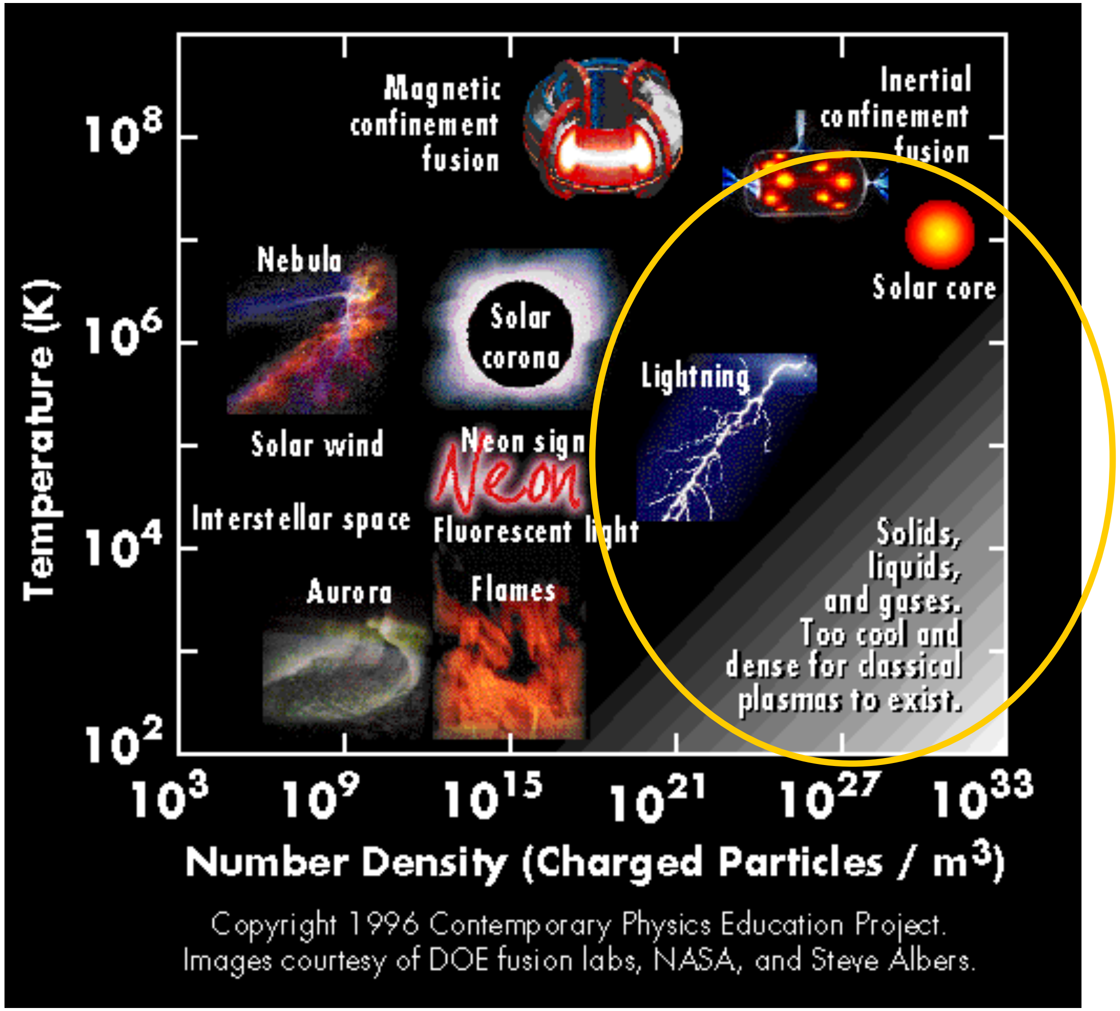
$$= 6.4\sqrt{\frac{n_e e^2}{\varepsilon_0 k_B T}}\frac{2\pi\varepsilon_0^2 (k_B T)^2 2}{n_e Z e^4 \ln\Lambda}$$

$$= 6.4\frac{1}{\lambda_D}2\pi n_e\frac{\lambda_D^4}{Z\ln\Lambda}$$

$$\approx \frac{10N_D}{Z\ln\Lambda}$$

N.B.: - A 'good' plasma is collisionless - but a 'bad' plasma is not - For them, large angle binary encounters are important.

“Bad” Plasmas



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- The plasmas we are interested in are dense – so have coulomb energies of eV – 10’s 100’s eV, but similar temperatures.
- This means there are few (often <1) particles in a debye sphere.
- All our normal analysis no longer applies!.

Problems for Warm Dense Matter

- As the plasmas are not ‘good’ there are few particles in a Debye Sphere, so the whole theory breaks down.
- The coulomb logarithm even goes negative!
- Collisions and collisionless phenomena compete.
- So don’t know the collision time….
- So we don’t know the transport coefficients.
- As the coulomb energy and thermal energies compete, it is not clear what the pressure is, or the internal energy – so we don’t know the equation of state.
- Also the ions are packed so close together that their ionisation potentials are altered (as in metal), so we have problems with both optical properties and with heat capacities.
- All of these problems are *starting* to be addressed with X-Ray FELs.

Ionisation in ‘Bad’ Plasmas

- In low density plasmas the Ionisation potential is similar to that of a free ion – but electrons do not feel the full coulomb field beyond a Debye length.
- However, at high densities the orbit of an electron in one ion starts to overlap with that of its neighbour - just like in a metal.
- Ionisation thus depends in a complicated way on both temperature and density, in a way that until recently had never been tested accurately.
- In this sense there is overlap between solid state physics and dense plasmas, and concepts of band theory.
- Current plasma codes use a classical ‘fix’ between the low and high density limits - the Stewart Pyatt model (50 years old).

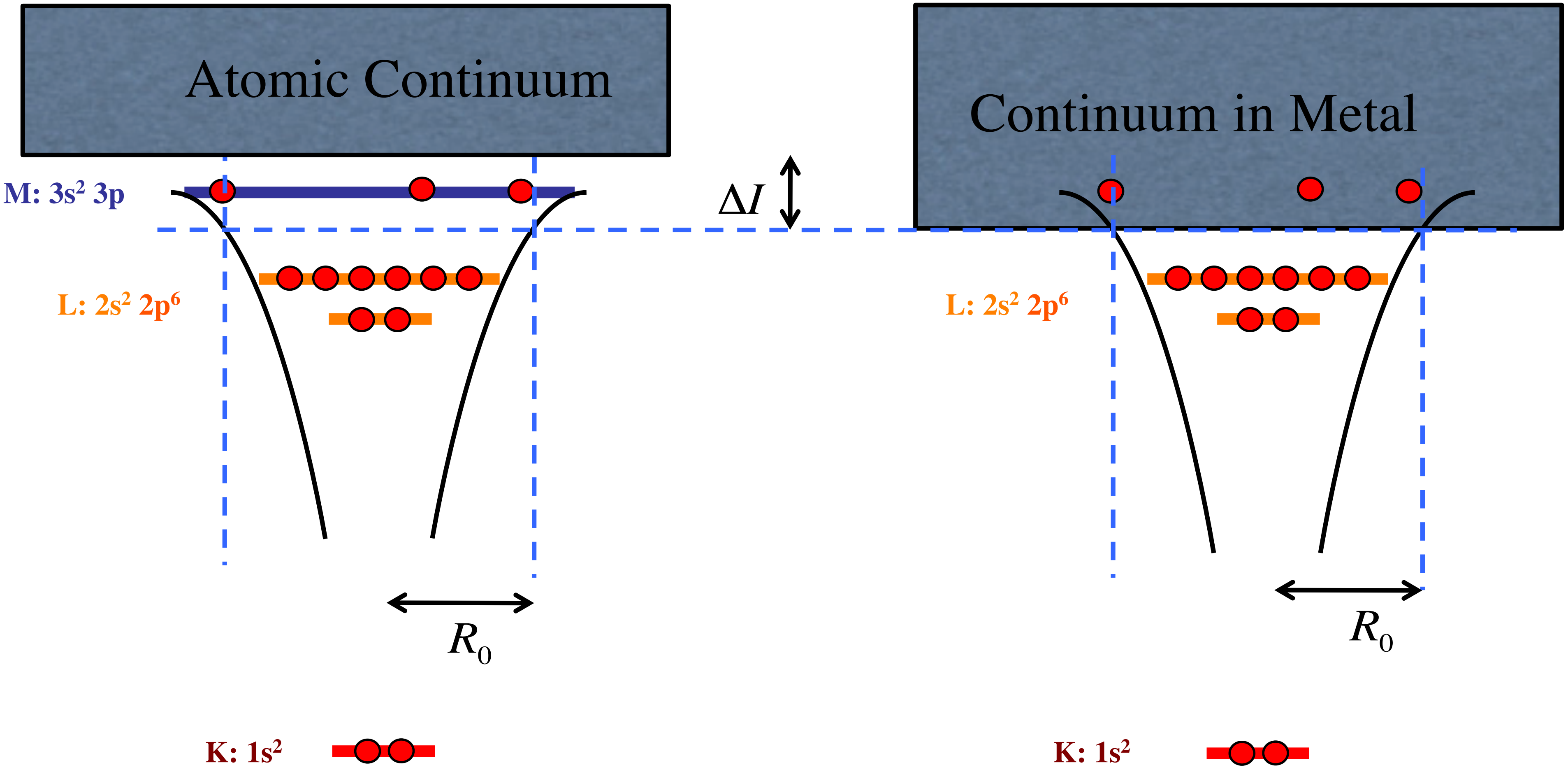
The Ion Sphere Model
(thought to work at strong coupling)

$$\frac{4\pi R_0^3}{3} = \frac{z^*}{n_e} = \frac{1}{n_i}$$

Aluminum (Z=13 Atoms)

$$\Delta I = C\frac{z^* e^2}{4\pi\varepsilon_0 R_0}$$

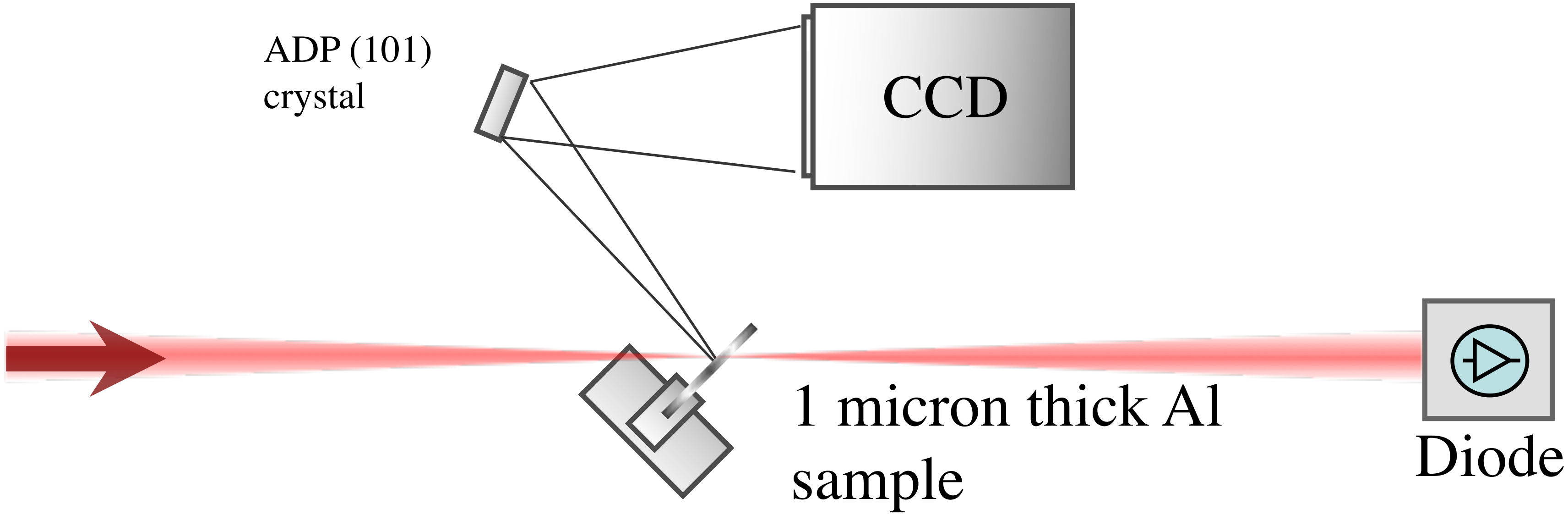
Aluminum Metal



LCLS:SXR experimental setup

X-ray spectrometer: Al K-alpha emission

1460–1680 eV



LCLS pulse

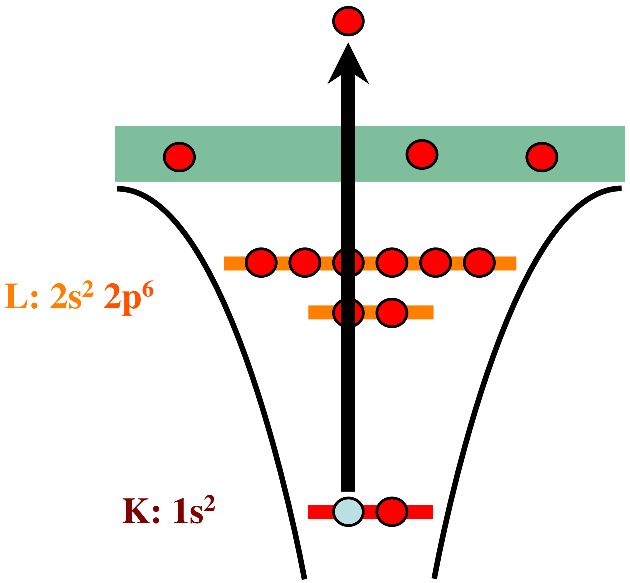
Photon energy: 1560–1830 eVPulse length < 80 fs Pulse Energy ~1.5 mJBandwidth ~ 0.4%

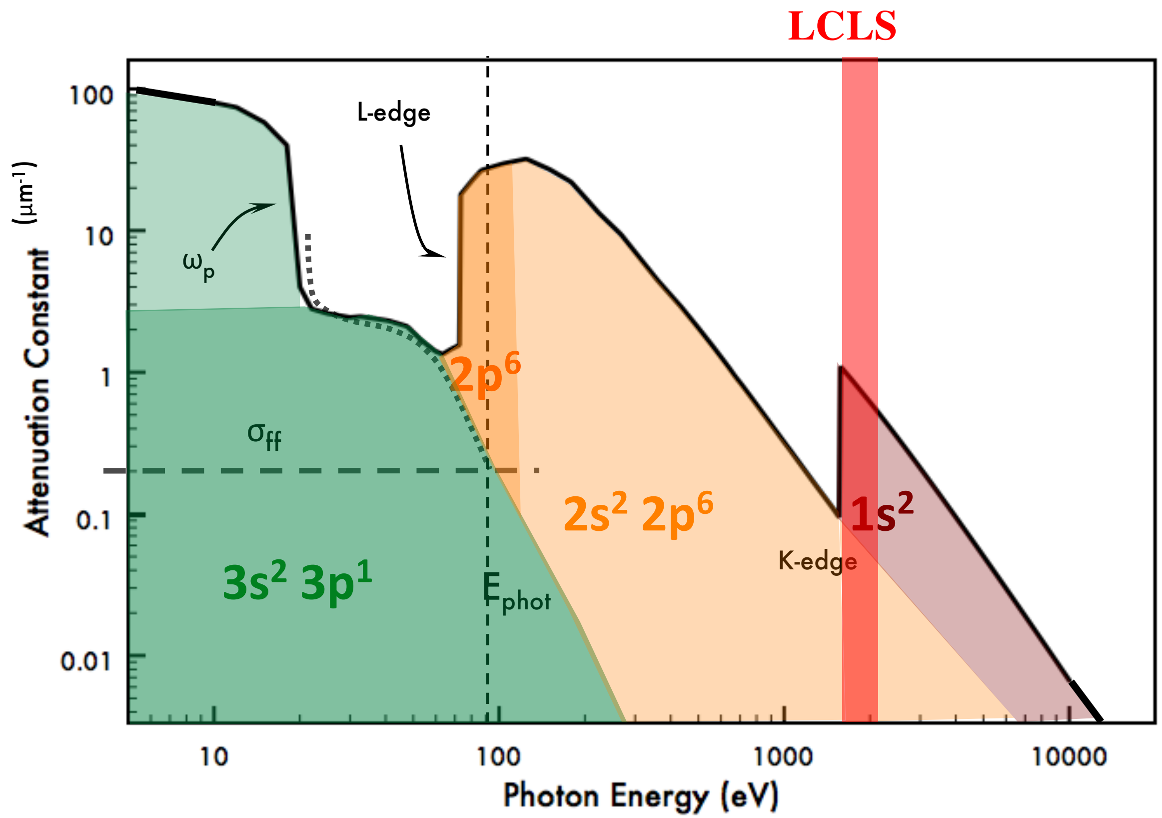
Peak Intensity ~10^{17} W cm^{-2}

Electronic structure of Aluminium

Neutral Al

Photo-excitation

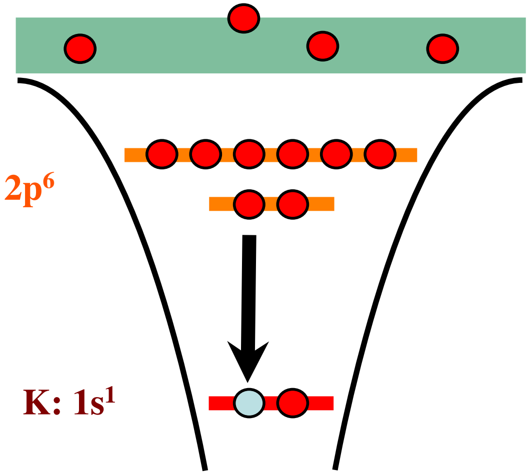


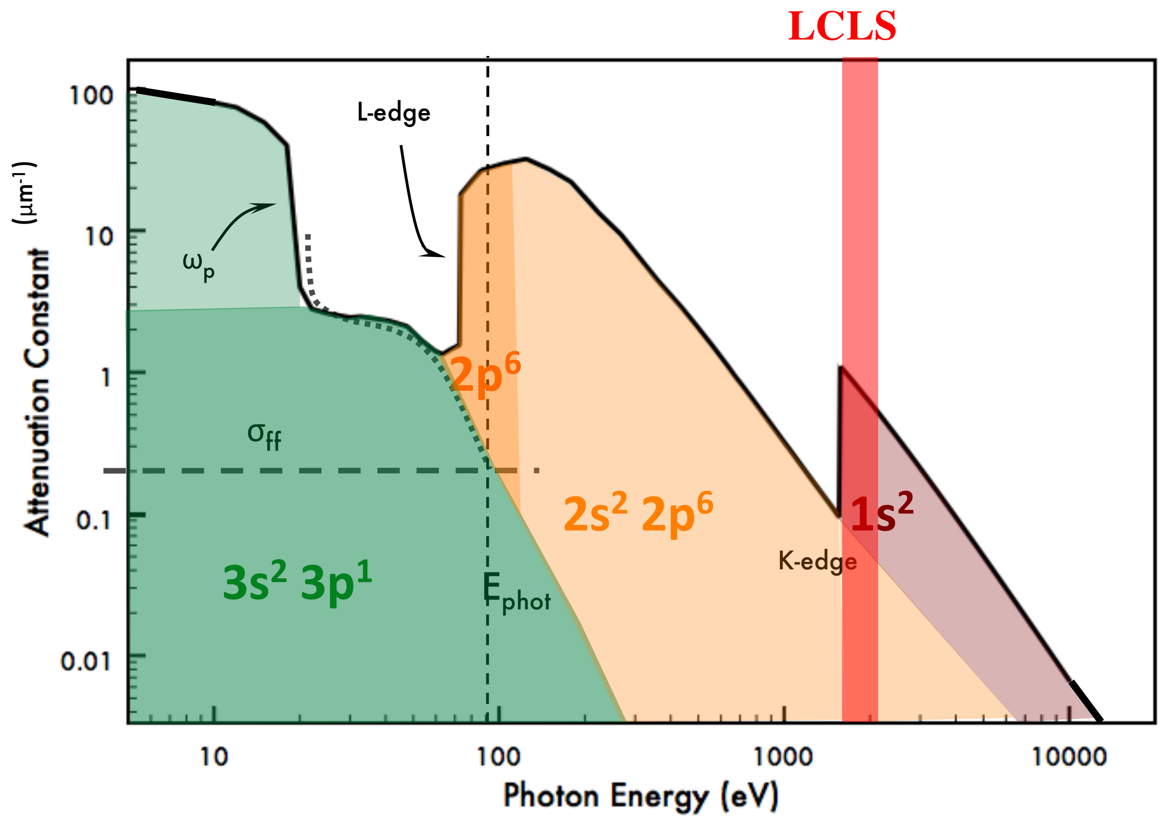


Electronic structure of Aluminium

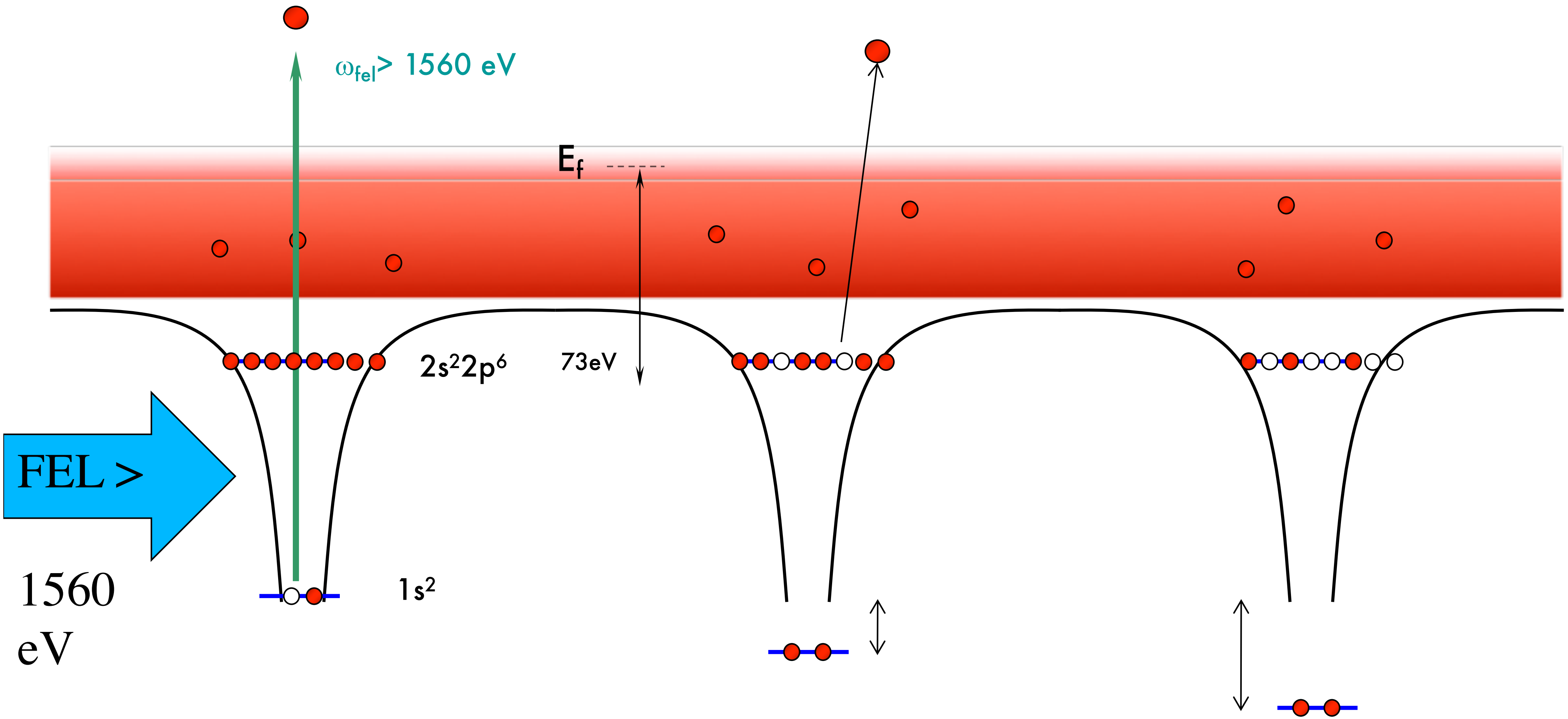
Neutral Al

K-alpha emission



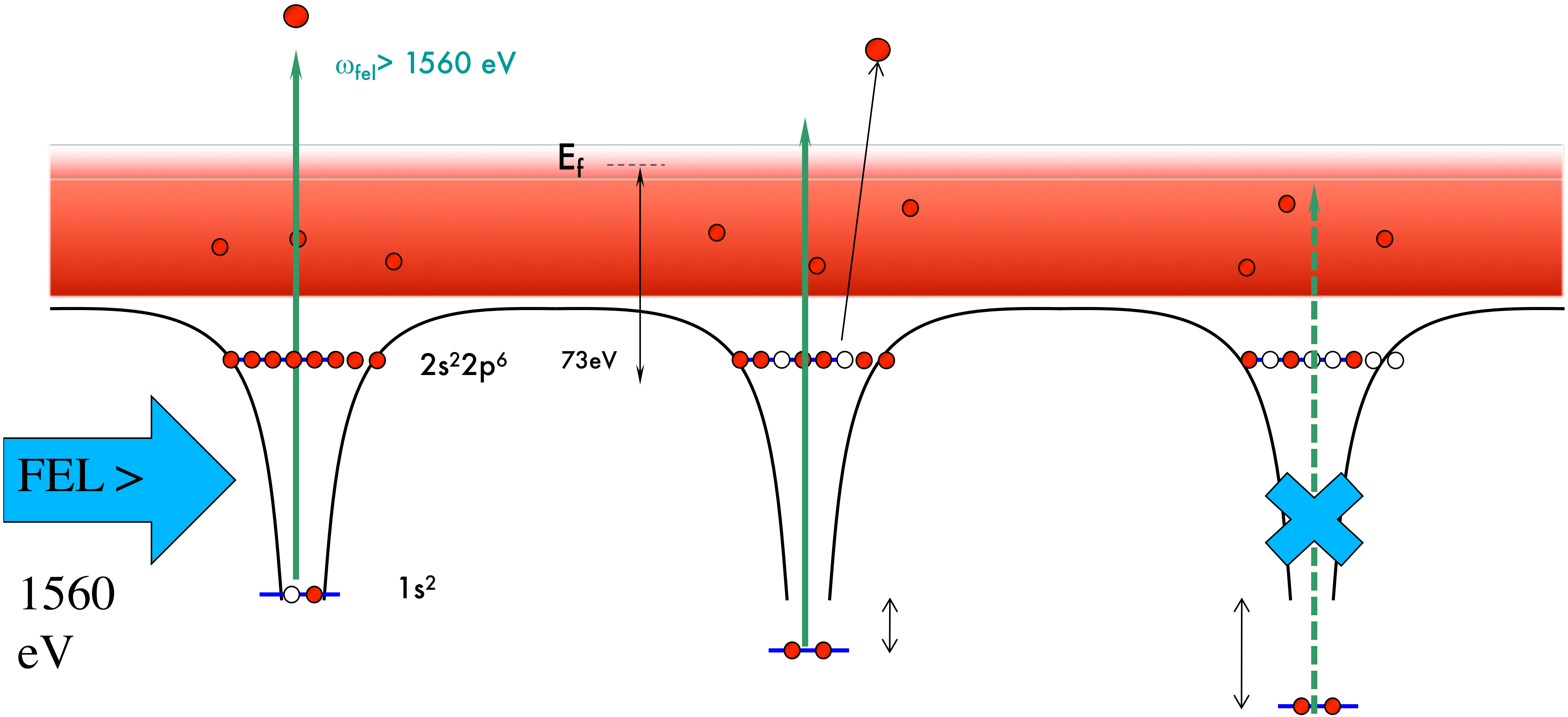


K-edge/alpha shift



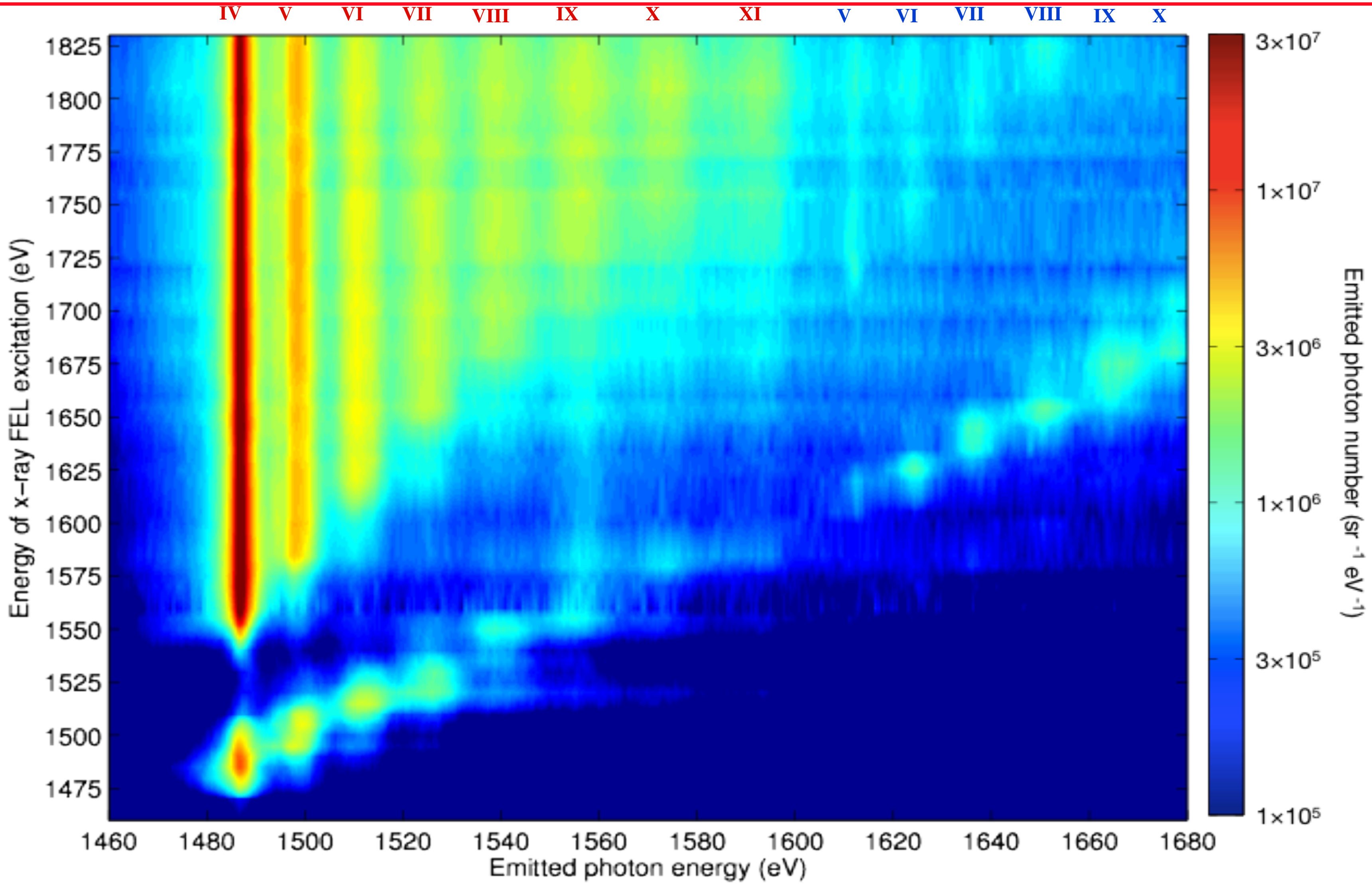
As the L-shell is ionized the K-electrons become more tightly bound. Both the K-alpha and K-edge shift to higher energies for higher charge states.

K-edge/alpha shift

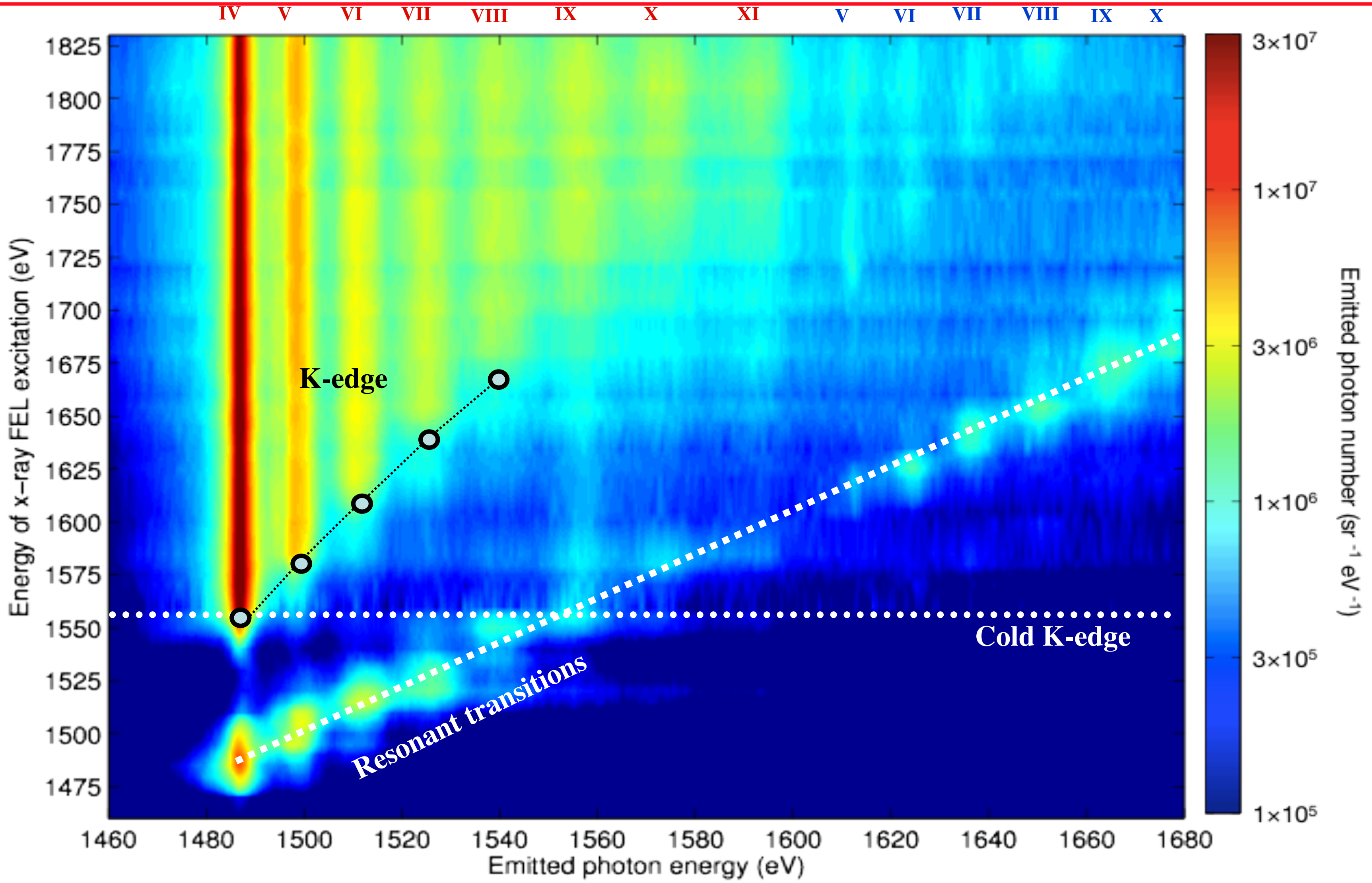


As the L-shell is ionized the K-electrons become more tightly bound. Eventually, if the FEL photon energy was initially only just greater than the original K-edge, it can no longer excite core holes in the highly ionized states. LCLS is a PUMP and a PROBE.

K-shell spectroscopy of Hot Dense Aluminium



K-shell spectroscopy of Hot Dense Aluminium

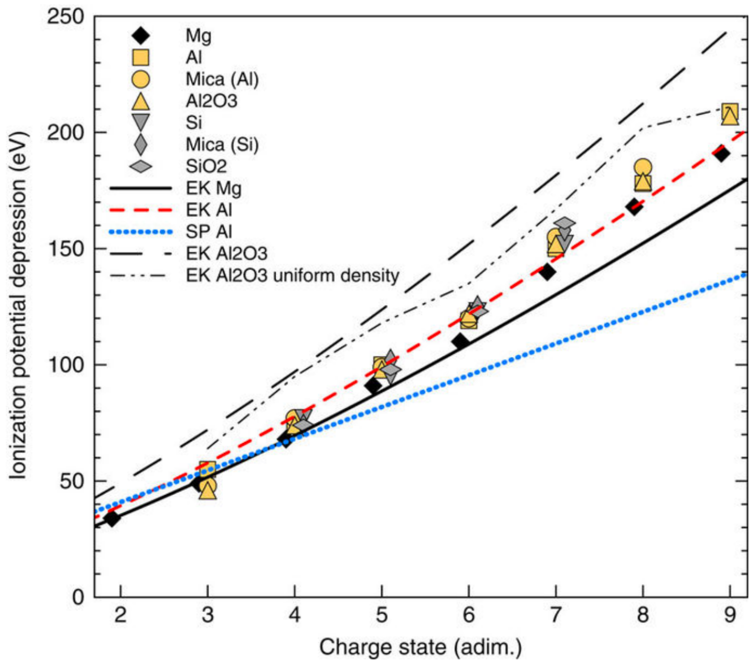


Ionisation Potential Depression

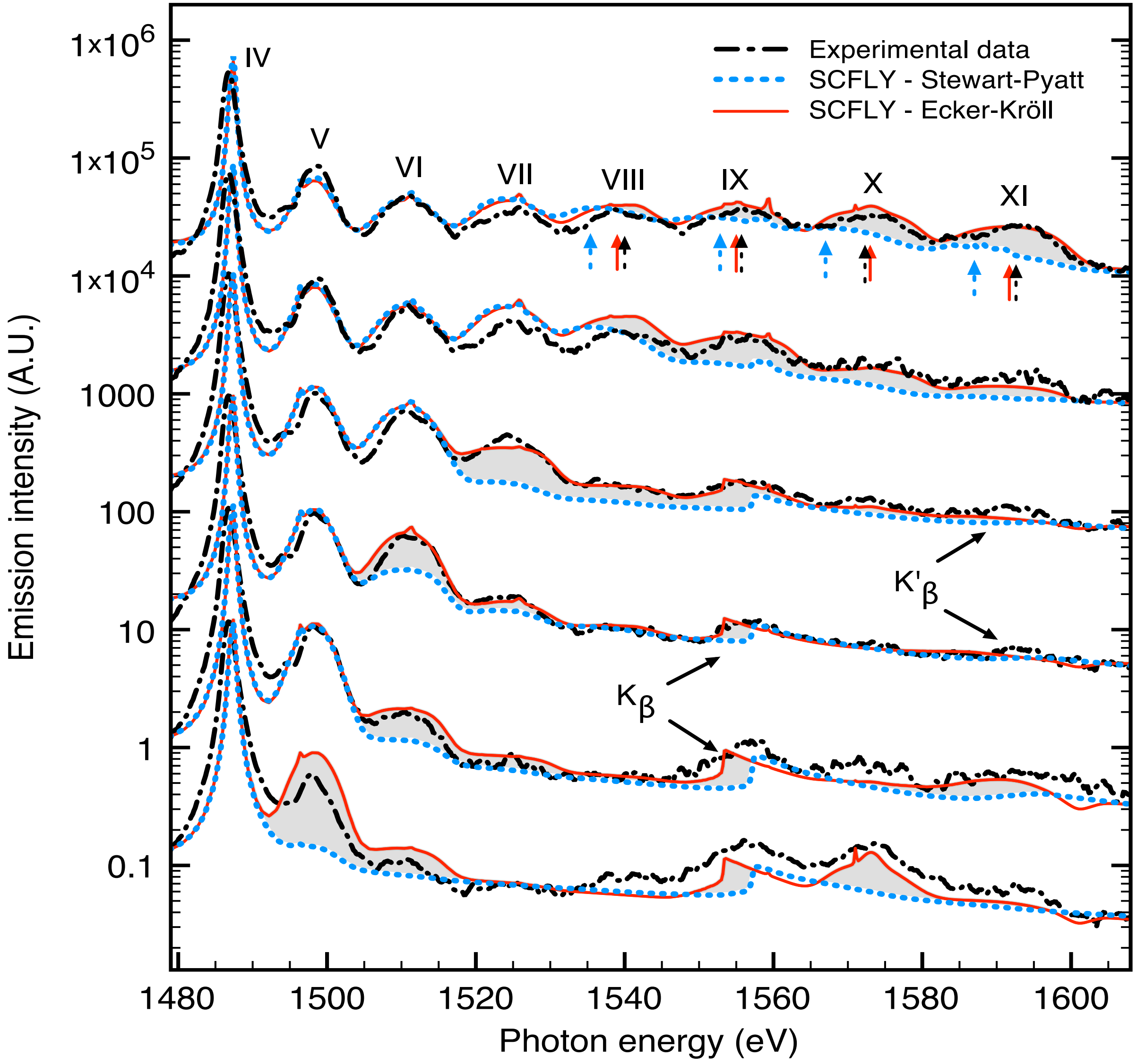
- The LCLS experiments found that the ionisation potential was lowered much more than previously thought, at least for these very strongly coupled plasmas

- Key to this finding is the FELs ability to heat matter before it has time to expand, so the density is well known

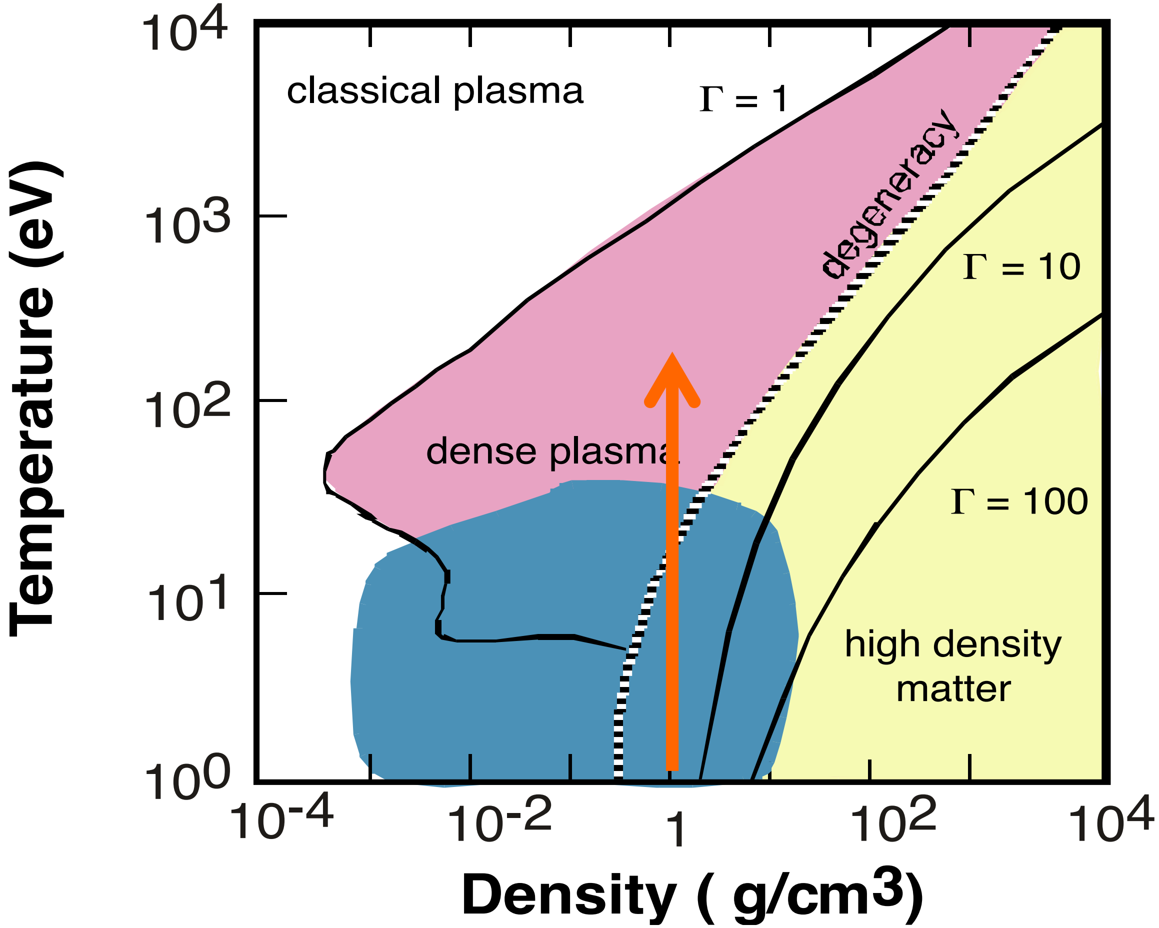
- Other materials have now been studied, and the results fit well with ab initio quantum calculations (density functional theory).



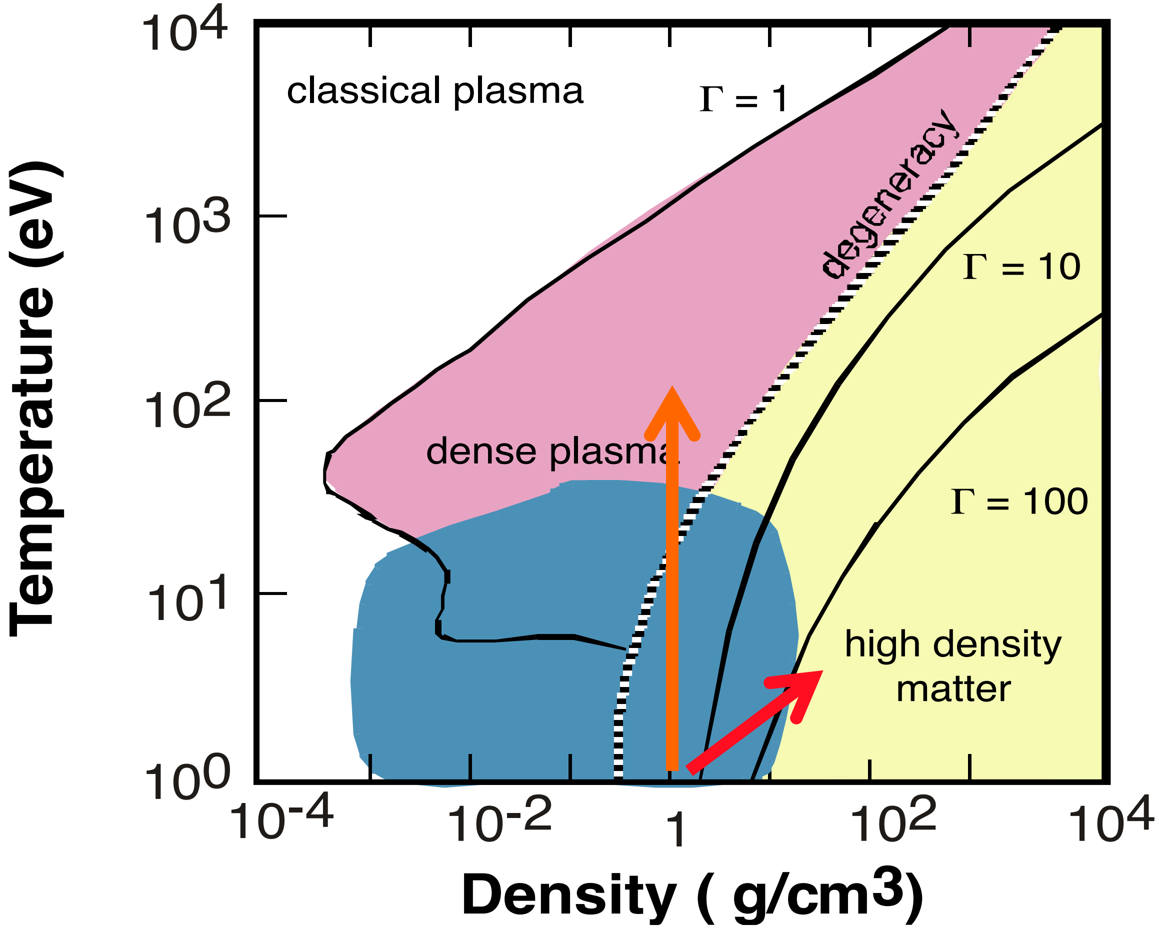
Measured and Calculated Spectra



Up to now we have been studying isochoric heating



Higher densities require us to compress…

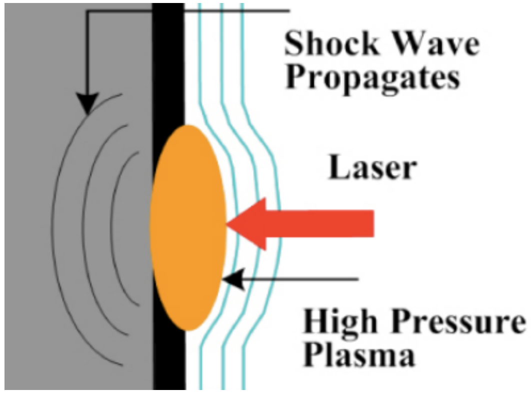


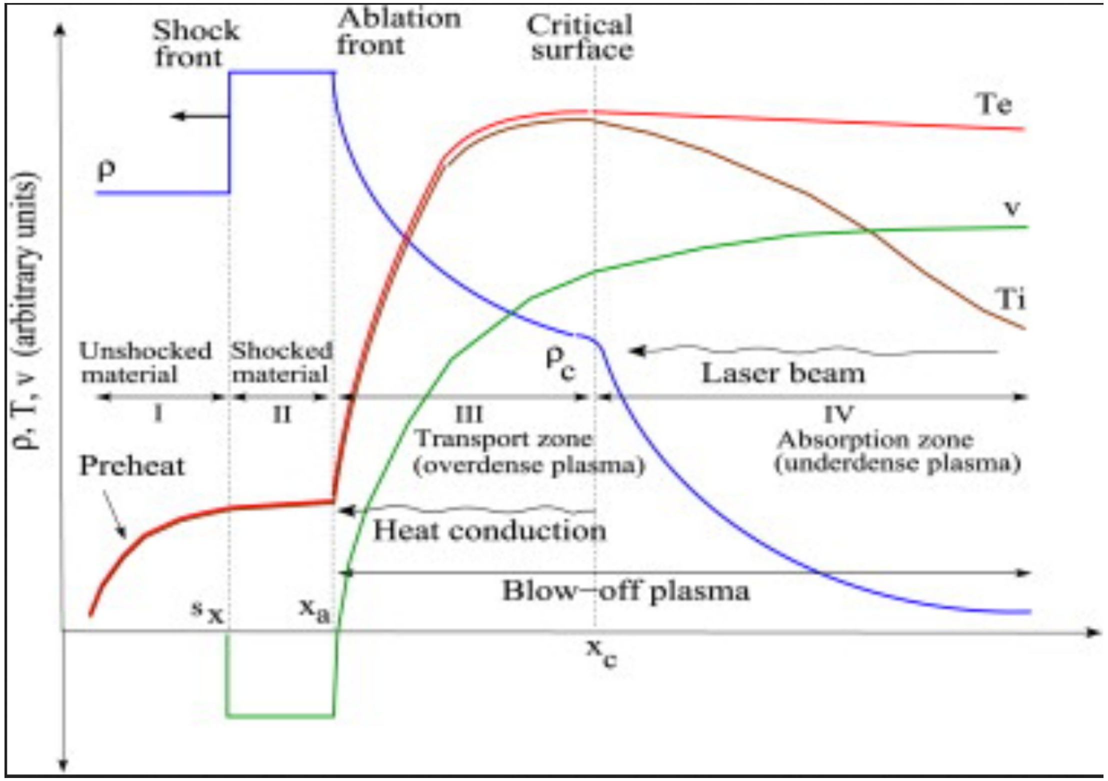
Ablative compression with optical lasers

High pressure (few Mbar) matter can be made by compressing crystals between two diamond anvils, but eventually the diamonds break.

In principle much higher pressures can be created by laser ablation with nanosecond lasers…

Ablative compression with optical lasers





Refractive index of plasma is

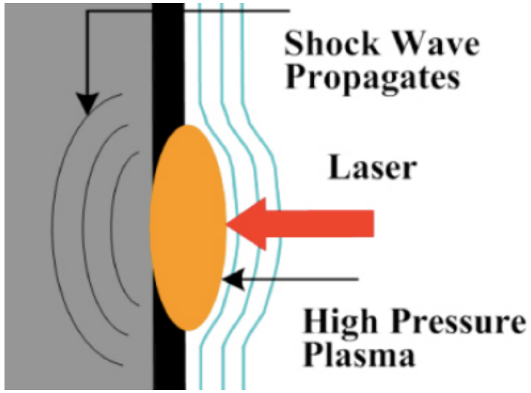
$$\mu = \sqrt{1 - \frac{\omega_{\mathrm{p}}^2}{\omega^2}}$$

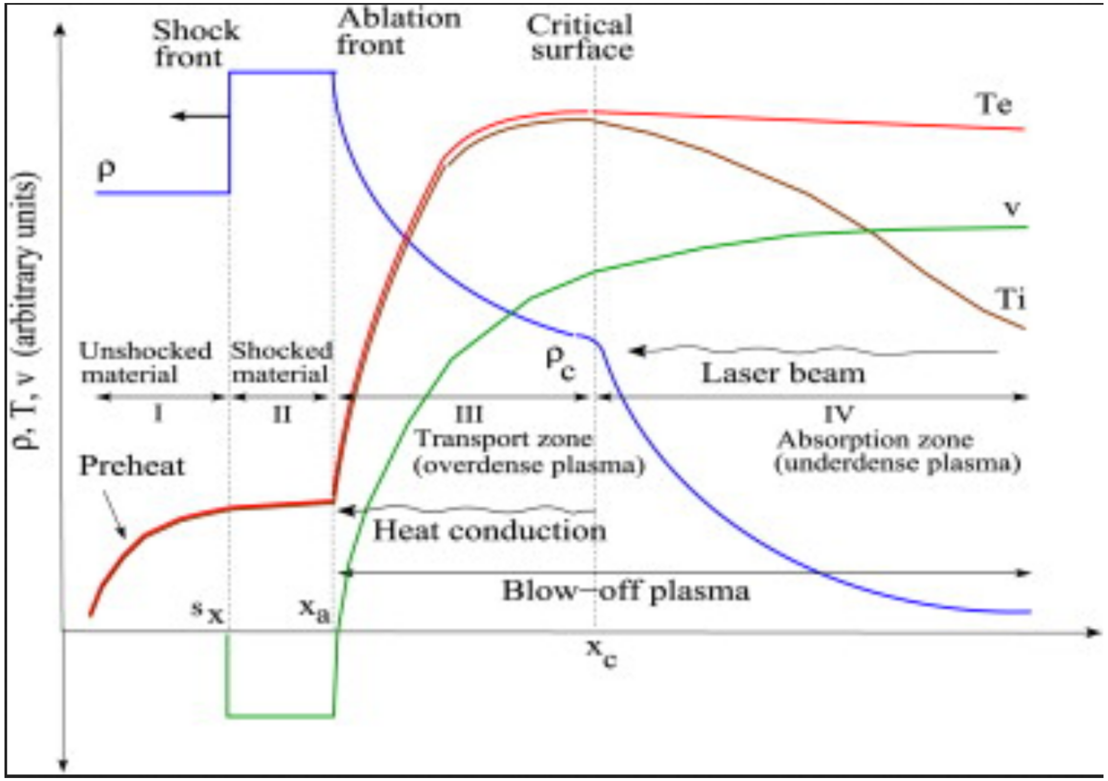
So light cannot penetrate past the critical surface, where the electron density is

$$n_c = \frac{\varepsilon_0 m_e \omega^2}{e^2}$$

From this point heat conduction takes over

Ablative compression with optical lasers





We equate a fraction of the laser intensity to the kinetic energy flow at the critical surface ...

$$\alpha I = \frac{1}{2}\rho_c v_c^3$$

and say the flow is regulated by the speed being mach 1 at the critical surface

$$v_c = \sqrt{P_c/\rho_c}$$

$$\rho_c \approx 2m_p n_c \approx \frac{2m_p m_e \varepsilon_0 \omega^2}{e^2}$$

Which gives a pressure

$$P \approx \left(\frac{8\pi^2 m_p m_e \varepsilon_0 c^2}{e^2}\right)^{1/3} (\alpha I/\lambda)^{2/3}$$

Laser Ablation Pressure

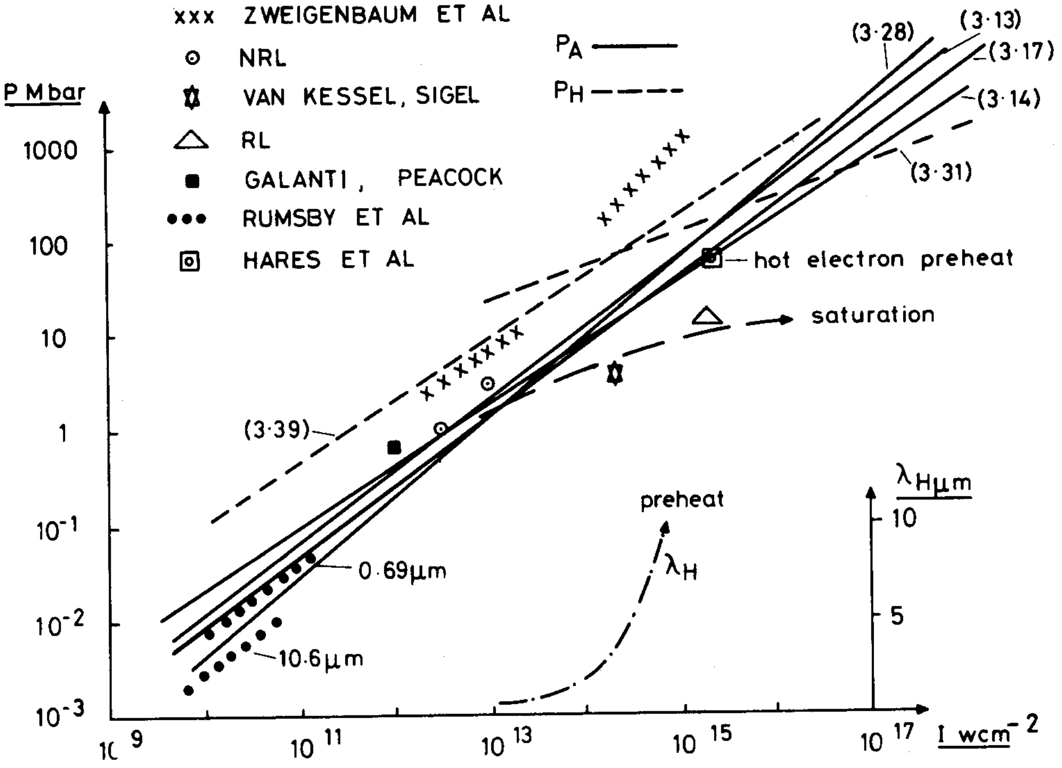
A VERY simple model would say a good fraction of the irradiance, I, flows down the temperature gradient to produce more plasma at the ablation surface. In steady state this produces a kinetic energy flow at the critical surface. Assume Mach 1 at critical surface

$$I \approx \frac{1}{2}\rho_c u_c^3 \approx \frac{\rho_c}{2}\left(\frac{P_c}{\rho_c}\right)^{3/2}$$

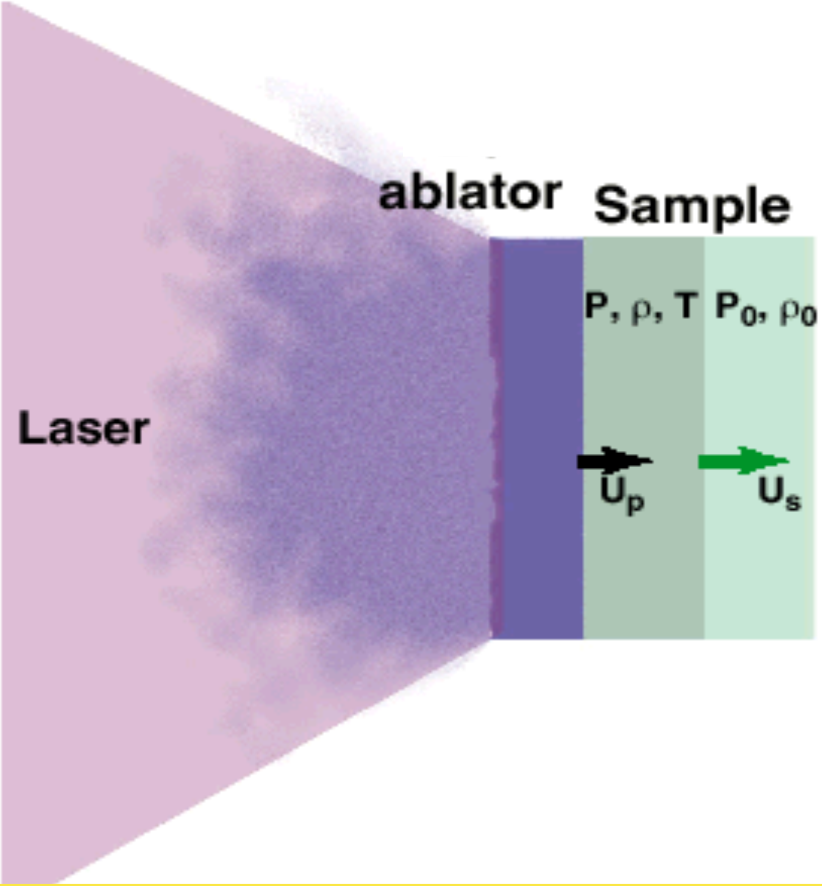
(note this equation must be wrong by about a factor of two, as there is thermal energy at the critical surface - we should really talk in terms of enthalpy flow)

$$P_c \approx \rho_c^{1/3} I^{2/3}$$

$$P_a = 3\times 10^{-9}\lambda_{\mu m}{}^{-2/3} I_{Wcm^{-2}}{}^{2/3} \text{ Mbar}$$



High Pressures can be generated by laser ablation



$$\rho_0 \left(Us \right) = \rho \left(Us - Up \right)$$

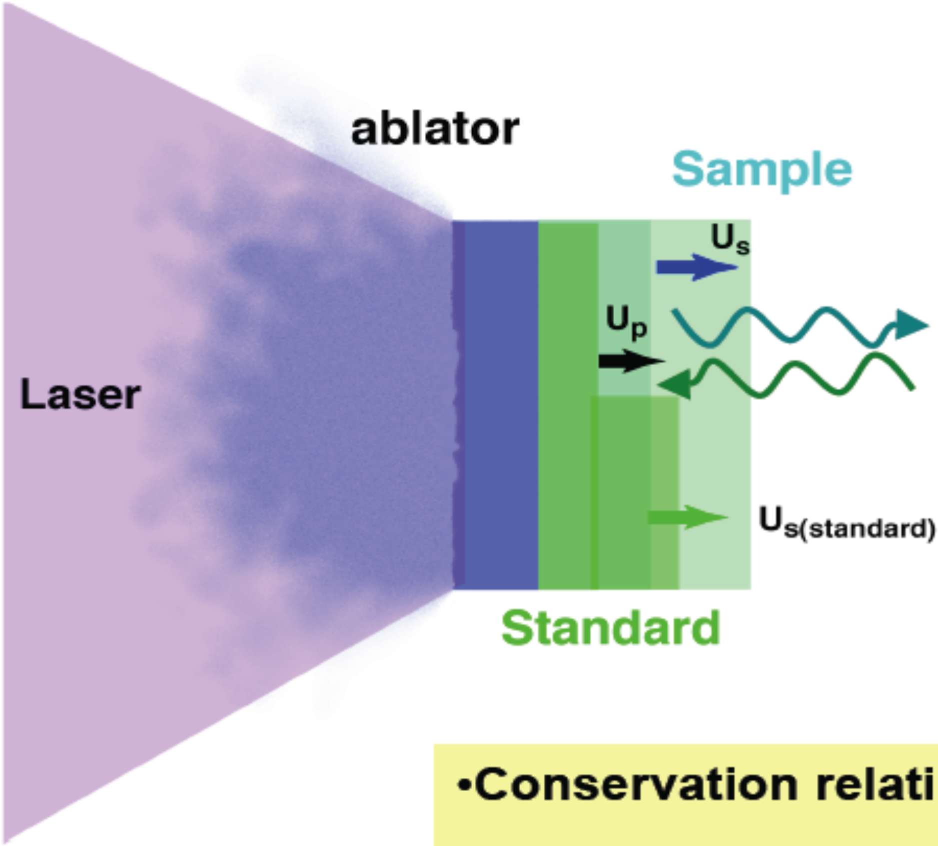
$$P = \rho_0 Us Up$$

$$E = \tfrac{1}{2} P \left(V_0 - V \right)$$

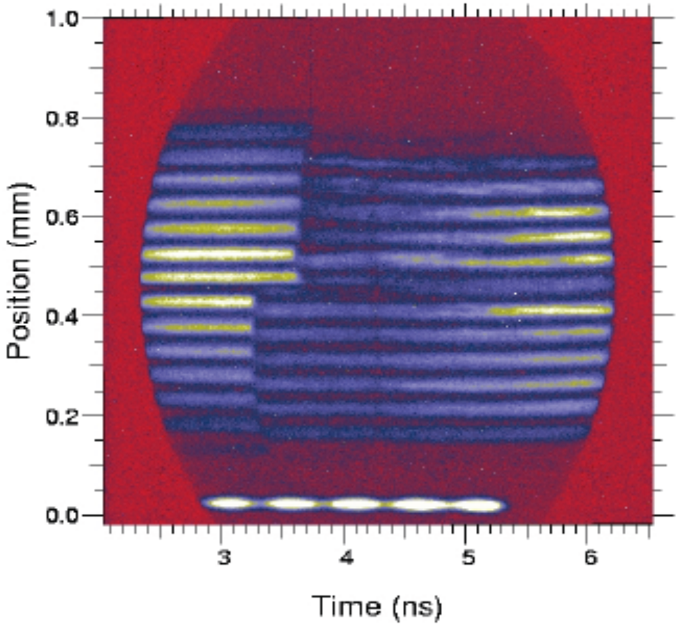
For SHOCKS, 3 equations, 5 unknowns (*Us, Up, P, ρ, E*).

Measure two of these, the others can be calculated.

How do we measure the equation of state and transport properties



VISAR measures velocity and reflectance



- Conservation relations => $P = \rho_0 U_s U_p$

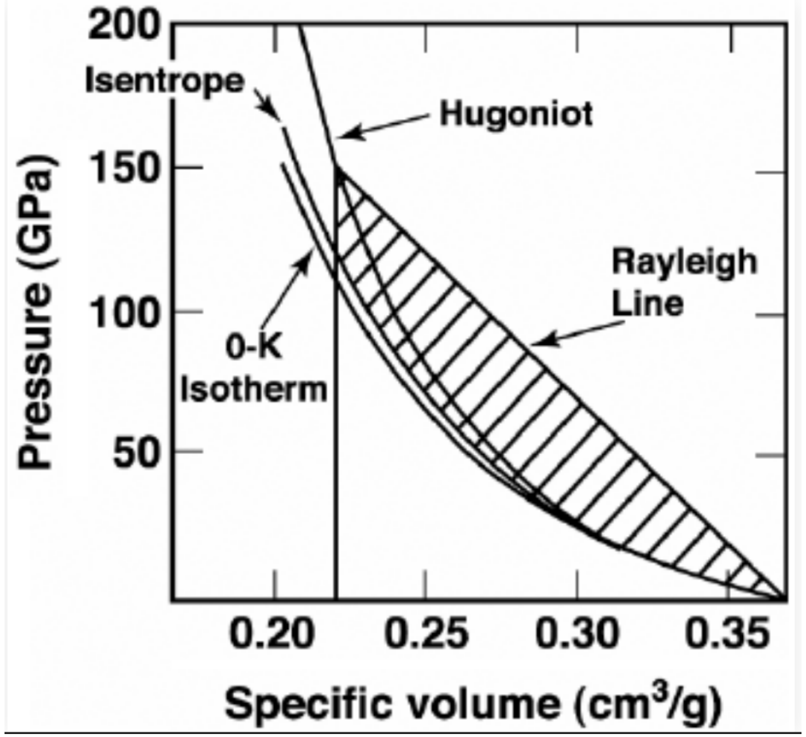
  $\rho/\rho_o = 1/(1-U_p/U_s)$

- Temperature needs to be measured separately

Slide from Rip Collins

The Hugoniot

The Hugoniot is the locus of all final states that can be reached by shock compression. It lies above the cold compression curve, as the shock produces lots of entropy and heat (thermal pressure).



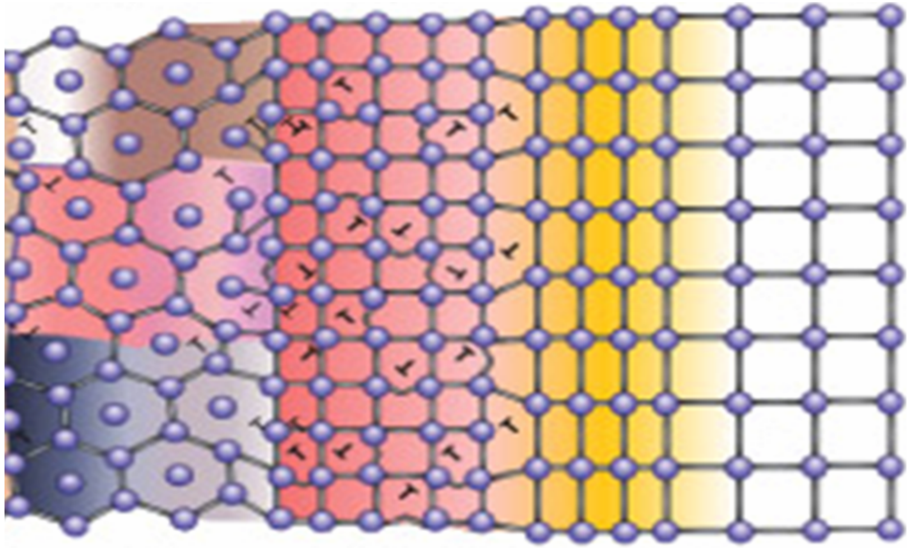
Questions in shock physics

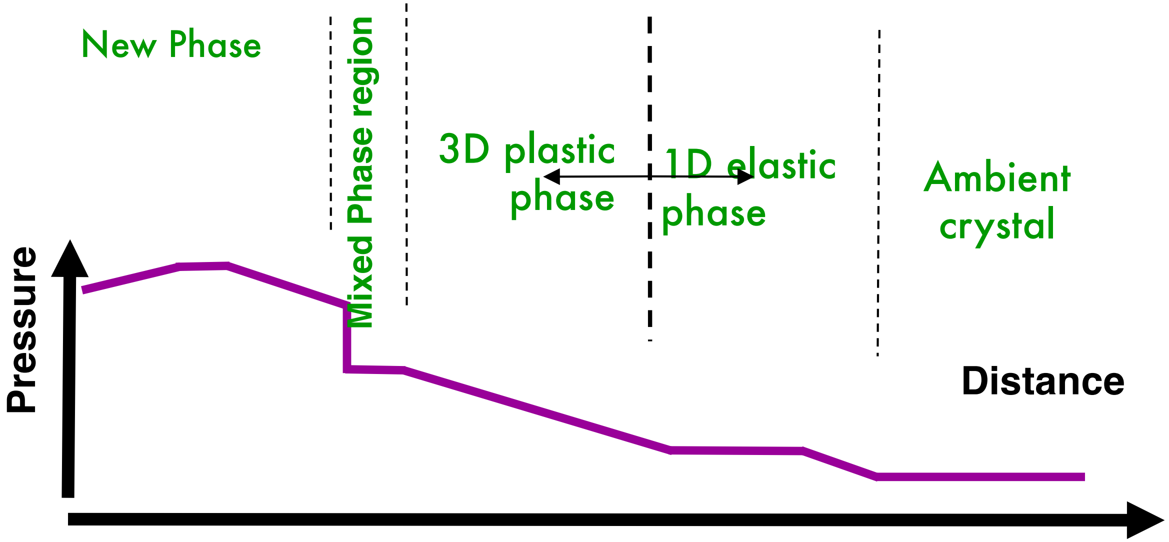
When we compress with laser ablation, we do so under conditions of uniaxial strain - how quickly does that matter ‘flow’ in the solid state to relieve the shear stresses?

How quickly do complex phase transitions take if we suddenly apply high pressures?

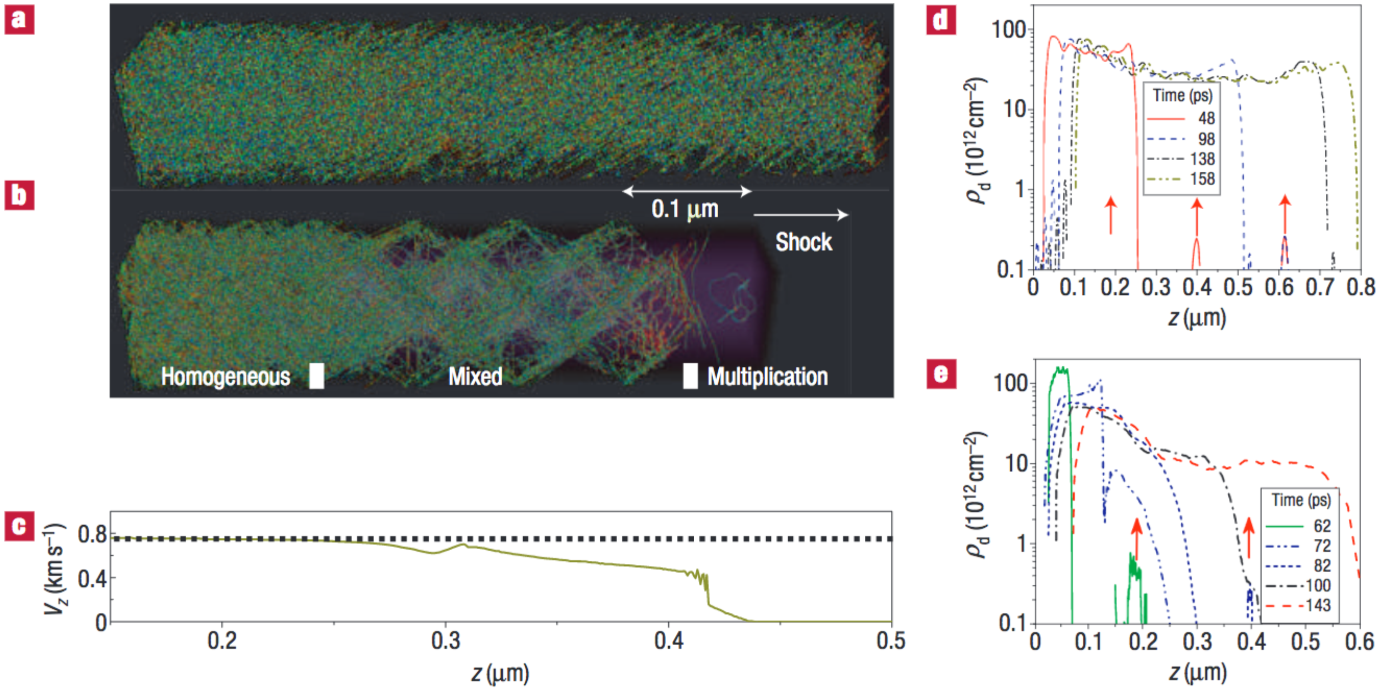
We can answer these questions by using laser ablation to create shocks in materials, and the x-ray laser to probe that material via x-ray diffraction.

How do crystals deform under uniaxial compression?





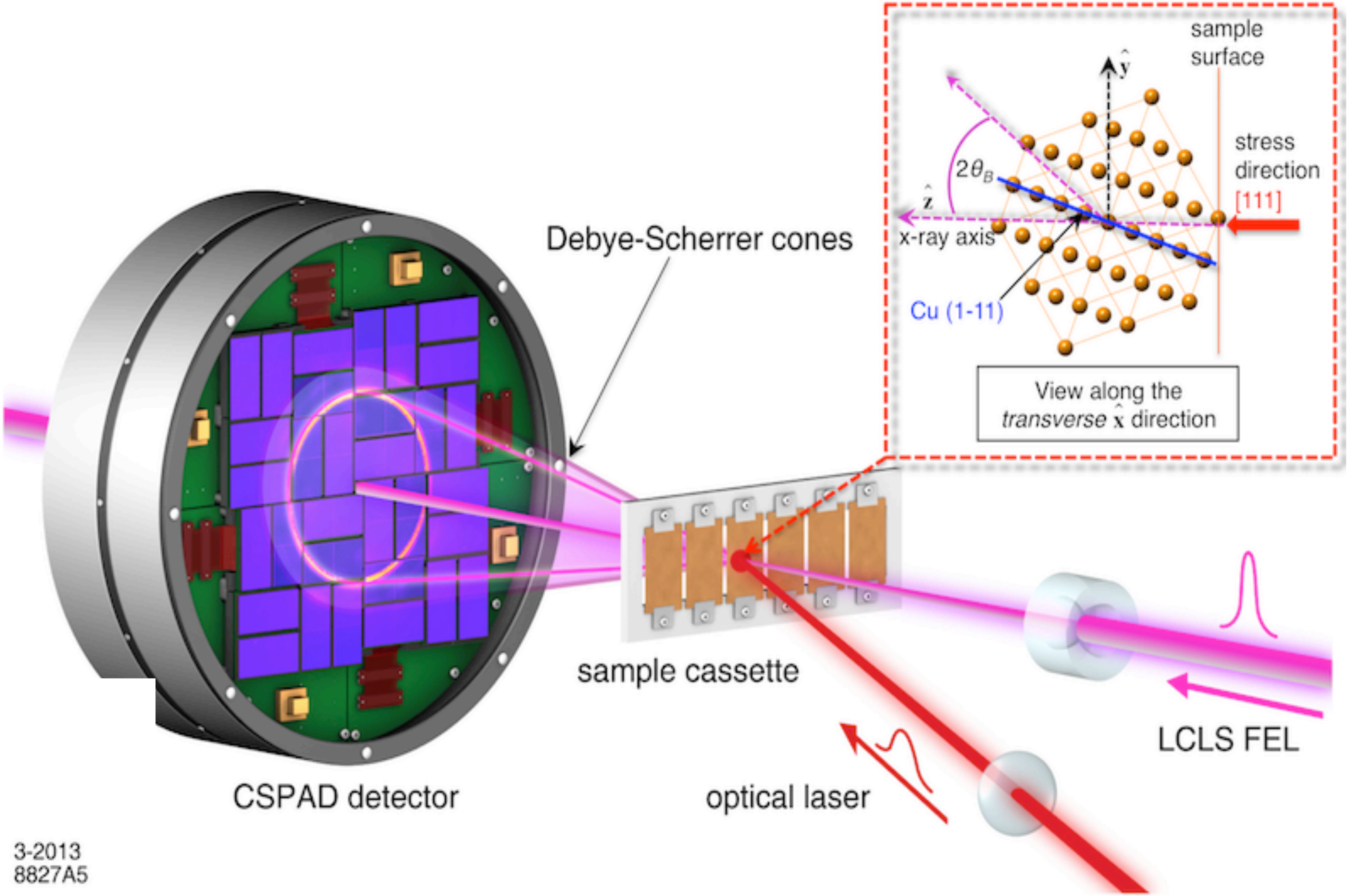
The Prediction : 2006



On the basis of our simulations, we make two predictions for experiments with shock rise times of only a few picoseconds. (1) We predict that prompt 3D relaxation would not be observed in copper, provided that diffraction measurements could be made at time intervals of a few picoseconds behind the shock front, which is now feasible experimentally[27–29]. (2) For strong shocks, above the limit

E. M. BRINGA, K.ROSOLANKOVA, R.E.RUDD, B.A.REMINGTON, J.S.WARK, M. DUCHAINEAU, D. H. KALANTAR, J.HAWRELIAK AND J. BELAK, Nature Materials, 5, 805 (2006)

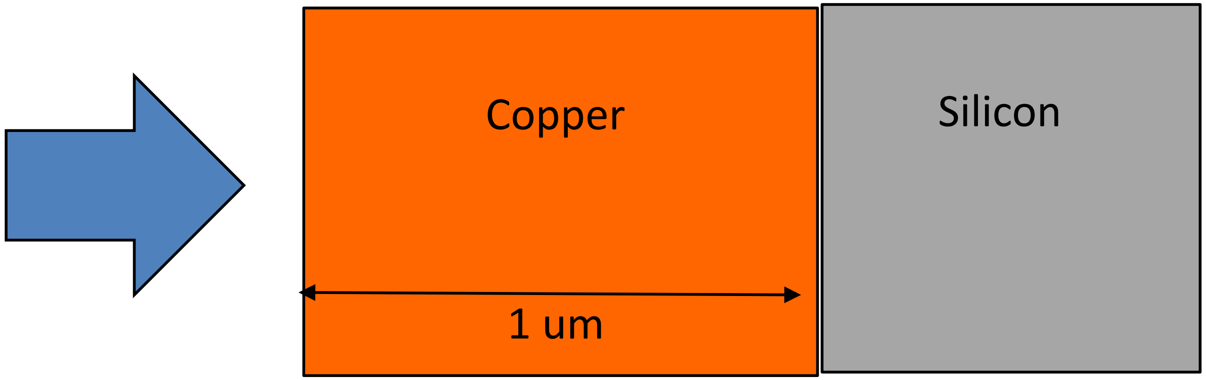
Experimental Set-Up - CXI



3-2013
8827A5

Applied Stress Pulse

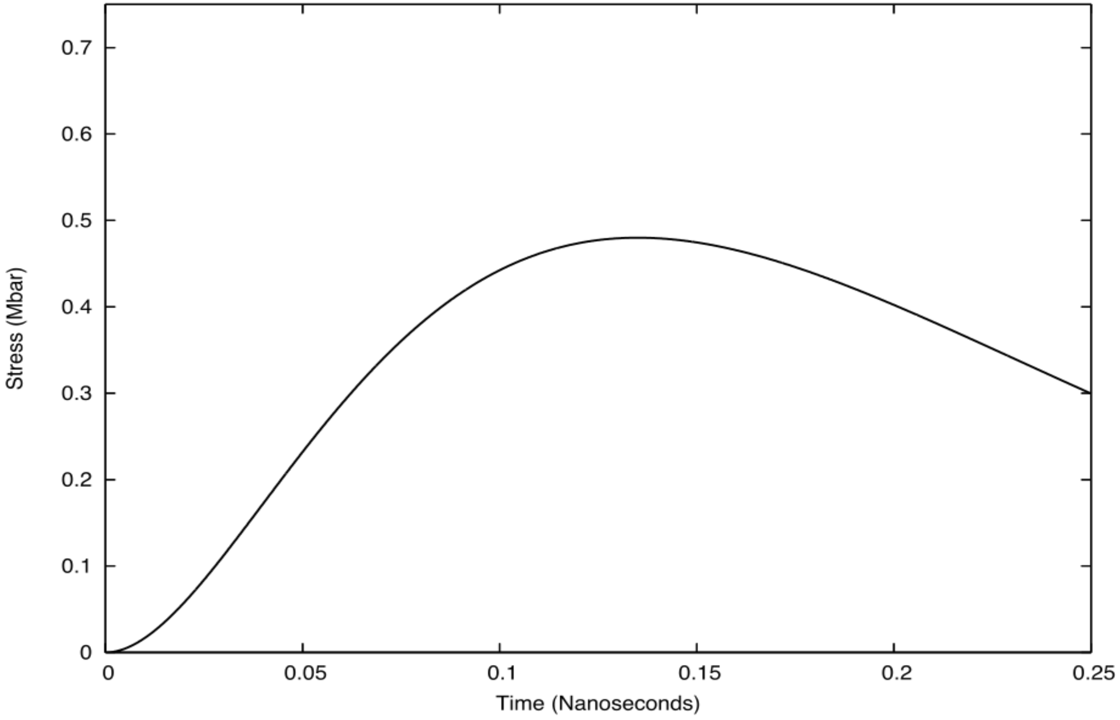
$$\sigma_n = \sigma_0 t^2 \exp\left(-\frac{t}{T_0}\right)$$



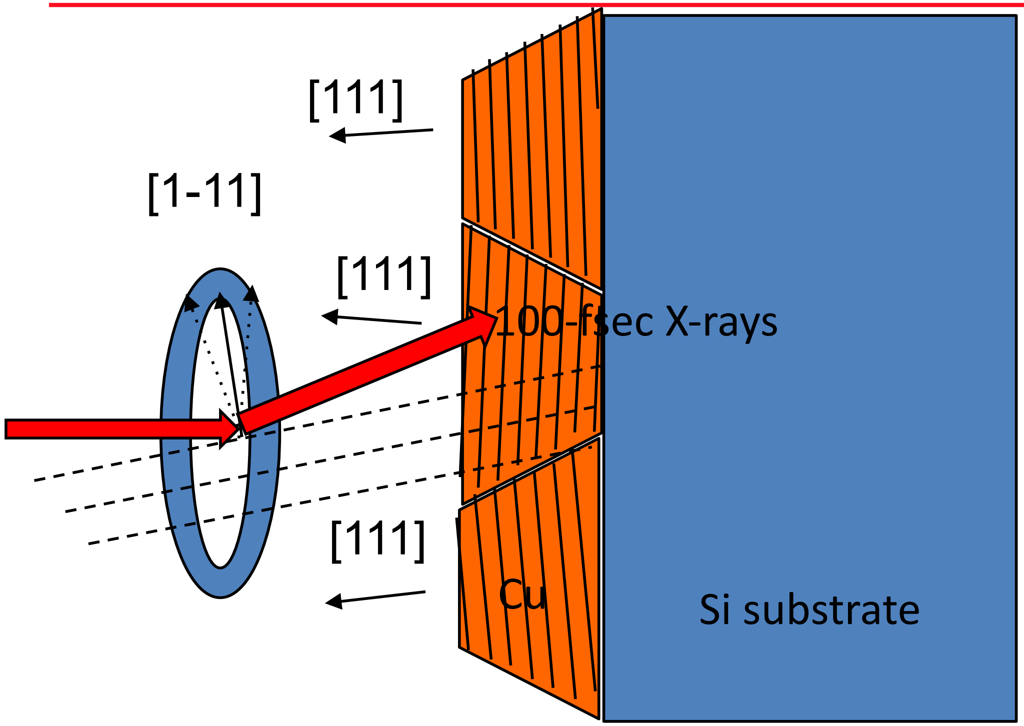
Copper

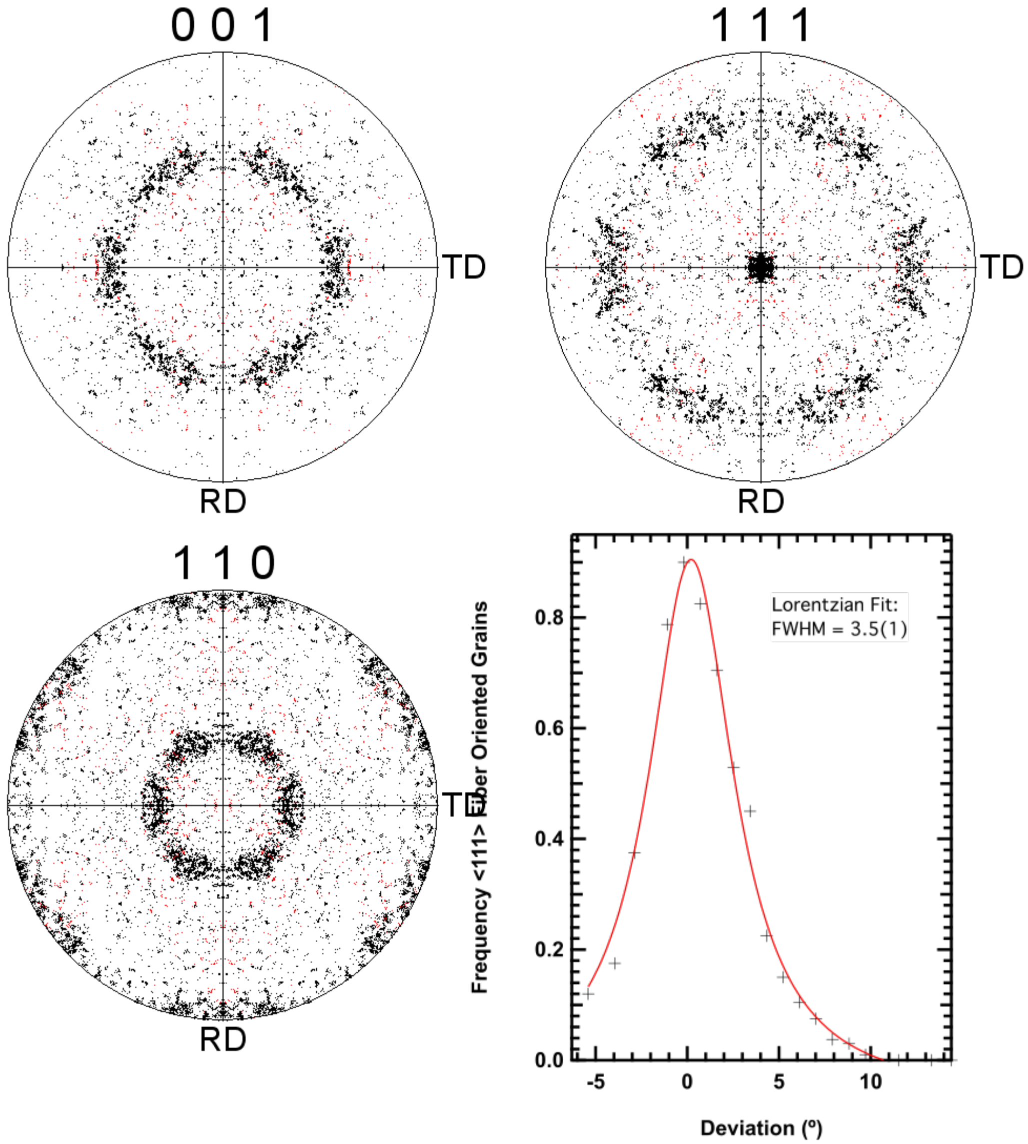
Silicon

1 um



Target Geometry and Texture



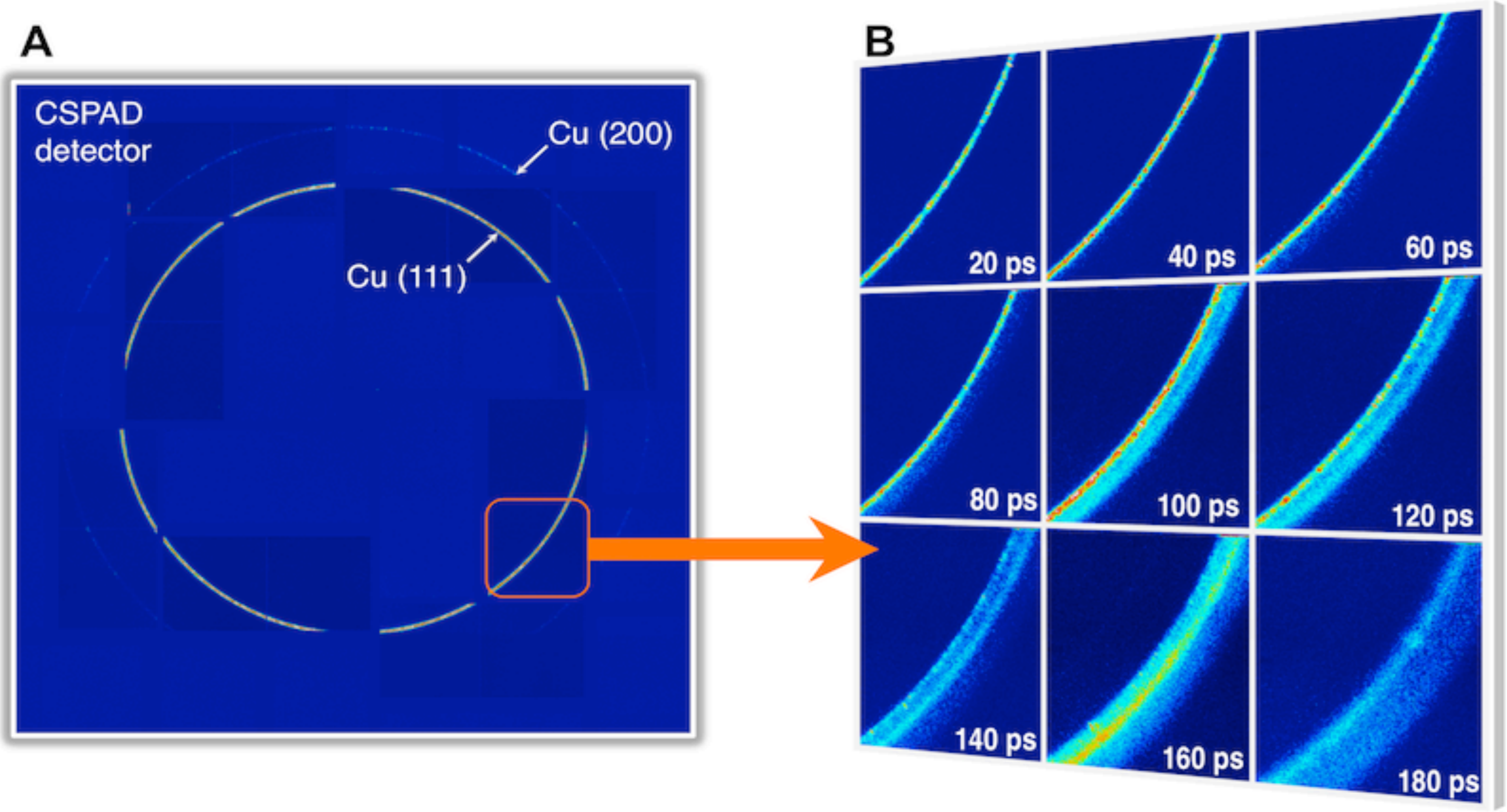


Note - grain size is of order target thickness: 1μm.
Highly oriented along 111.

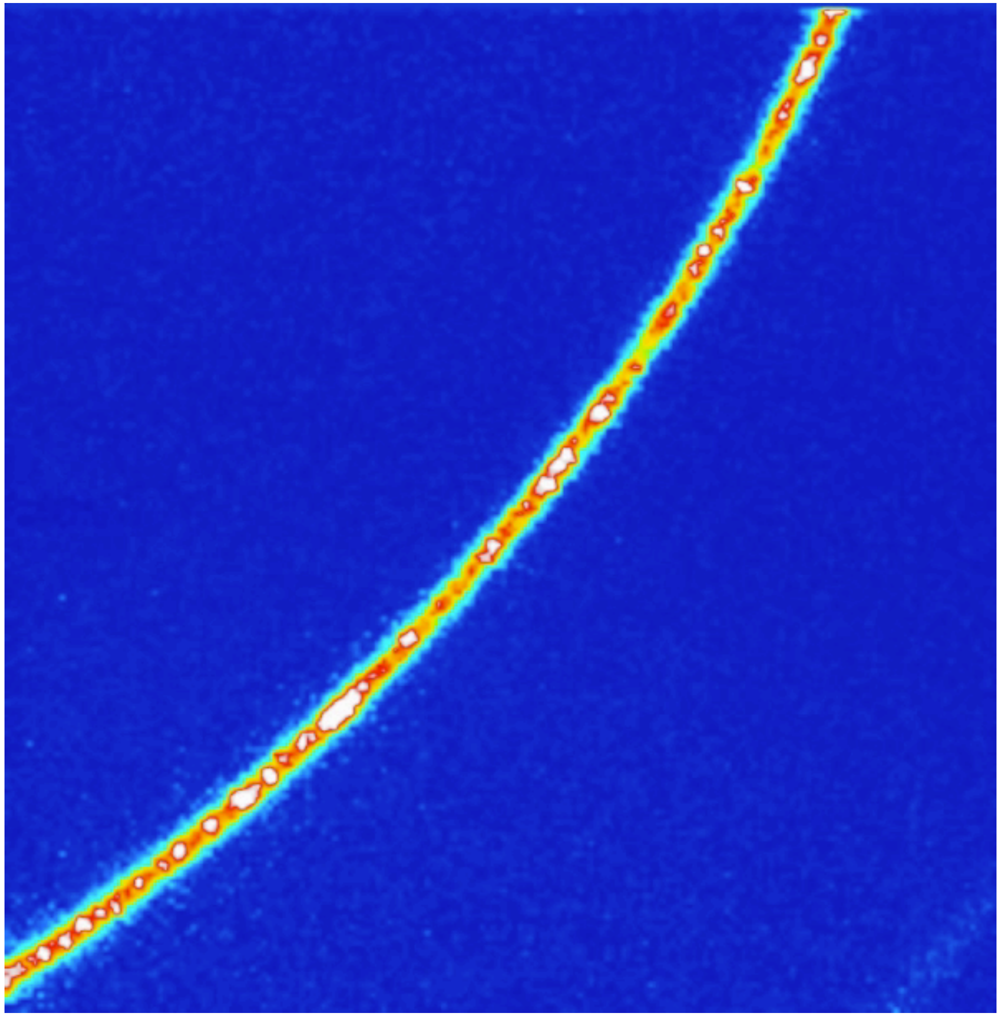
Just enough random distribution to allow us to interrogate full range of strain states as lattice planes rotate under compression.

BUT…Can still simulate as 'single crystal' in terms of hydrodynamic response.

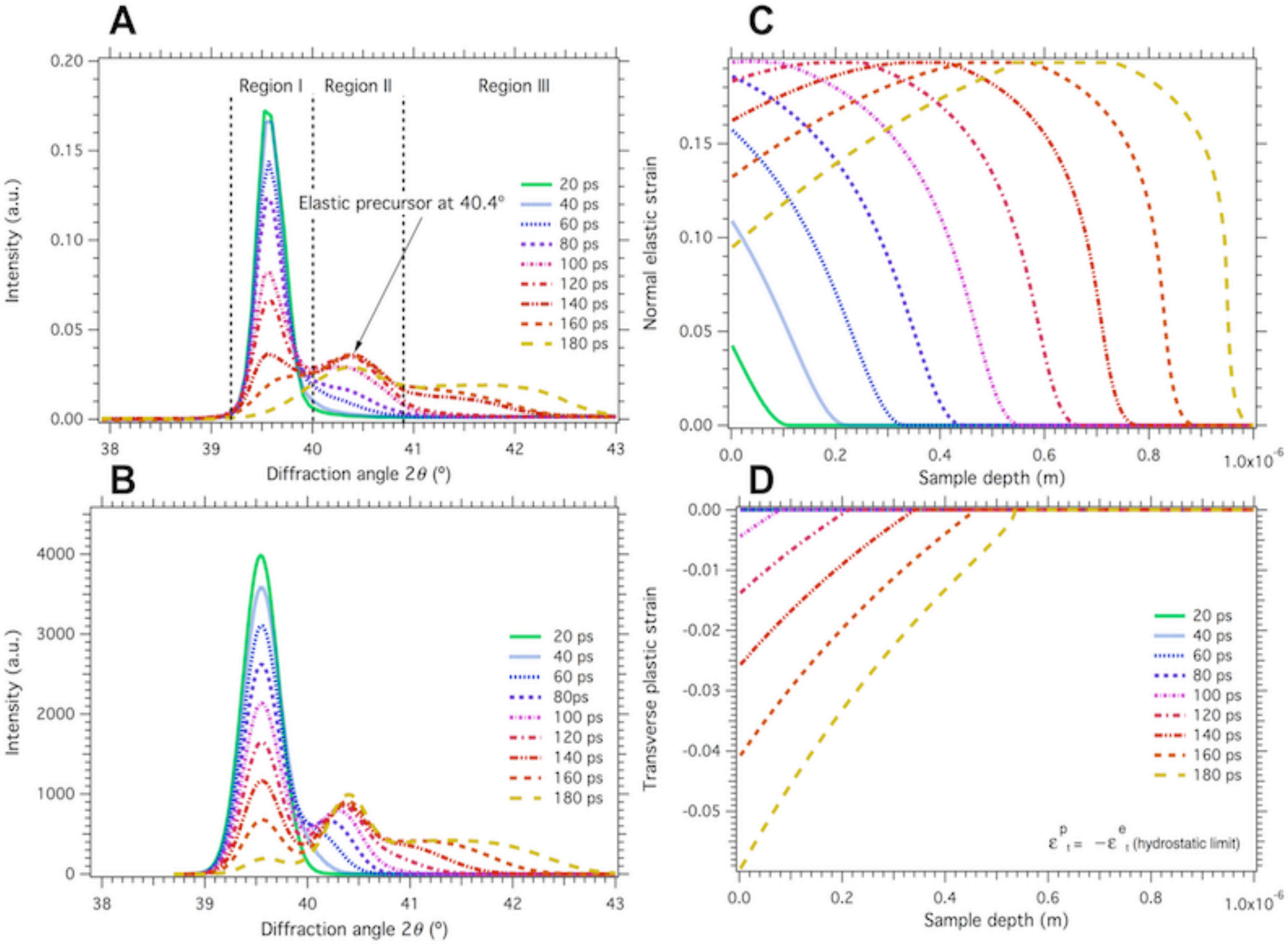
Experimental Results



Experimental Results - Movie

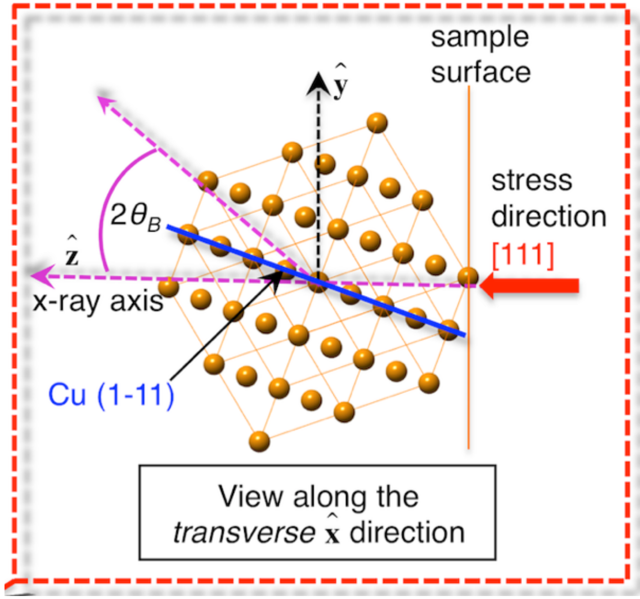


Simulations of Experimental Results



D. Milathianaki et al, Science 342, 220, (2013)

Diffraction Model in Hull Geometry



View along the *transverse* x̂ direction

$$\vec{G}=\frac{2\pi}{\lambda}(\sin 2\theta_B, 0, \cos 2\theta_B-1)$$

$$F=\begin{pmatrix} 1+\varepsilon_t^e & 0 & 0 \\ 0 & 1+\varepsilon_t^e & 0 \\ 0 & 0 & 1+\varepsilon_n^e \end{pmatrix}$$

$$\vec{G}_0\bullet\vec{G}_0=(F\vec{G})\bullet(F\vec{G})$$

$$\sin^4\theta_B\times\left(\left[1+\varepsilon_n^e\right]^2-\left[1+\varepsilon_t^e\right]^2\right)+\sin^2\theta_B\times\left[1+\varepsilon_t^e\right]^2=\sin^2\theta_0$$

Note in the hydrostatic limit we find $\sin^2\theta_B=\sin^2\theta_0/\left[1+\varepsilon_t^e\right]^2,\qquad \Delta\theta=-\tan\theta_0\varepsilon_n^e$

Note in the elastic limit we find $\Delta\theta=-\sin^2\theta_0\ \tan\theta_0\varepsilon_n^e$

For our Bragg angle of 19.8 degrees we are 9 times more sensitive to plastic strain!

N.B. For general formula for arbitrary strain see Higginbotham: http://arxiv.org/abs/1308.4958

Questions in shock physics

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How quickly do complex phase transitions take if we suddenly apply high pressures?

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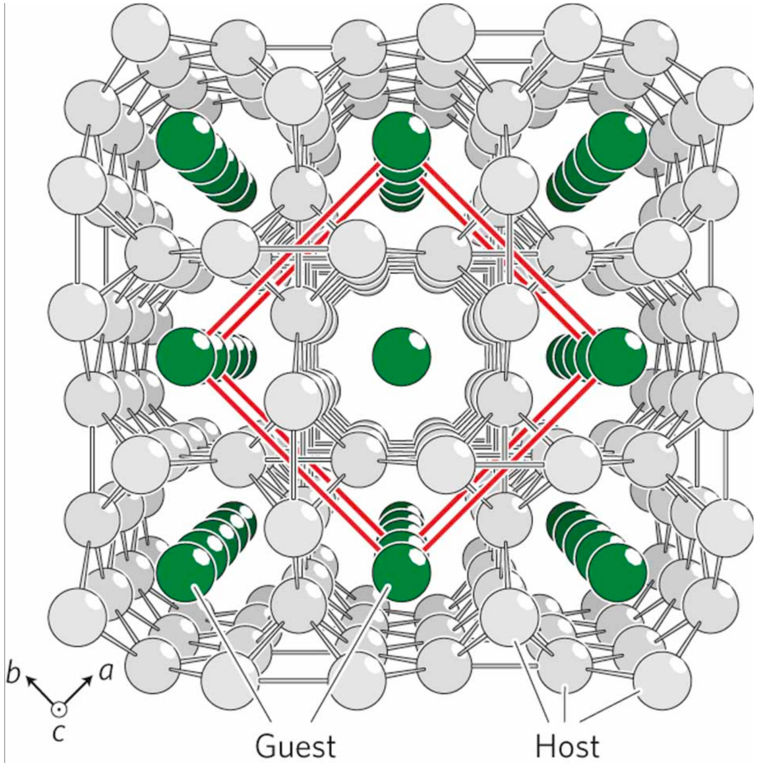
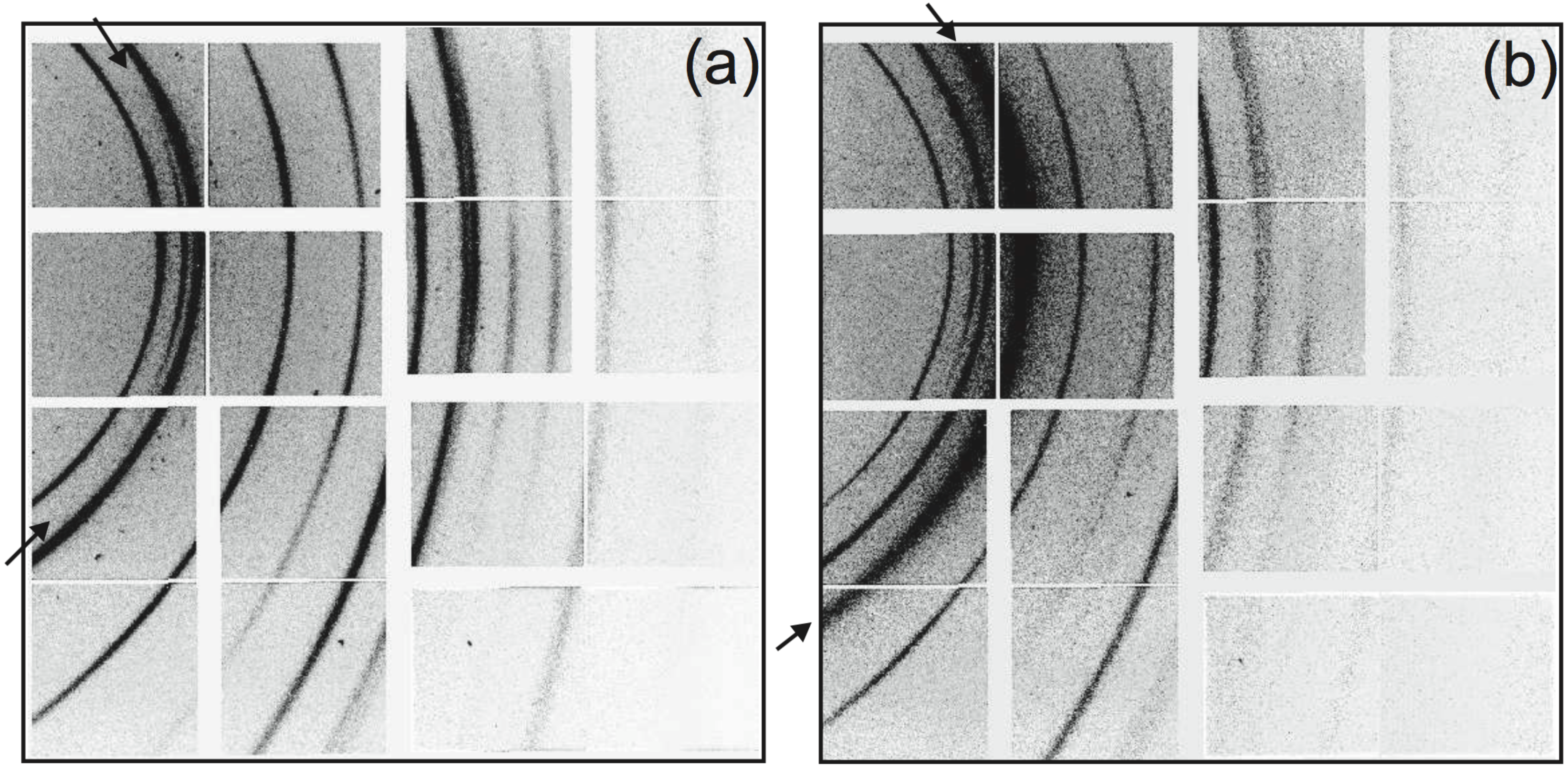
Host-Guest Structures

These are remarkable crystal phases, of single elements, where a string of atoms, the guest, resides down holes in the crystal (the host). The spacing of atoms in the string is incommensurate with that of the host spacing! Can these form within a nanosecond…?



Scandium Diffraction

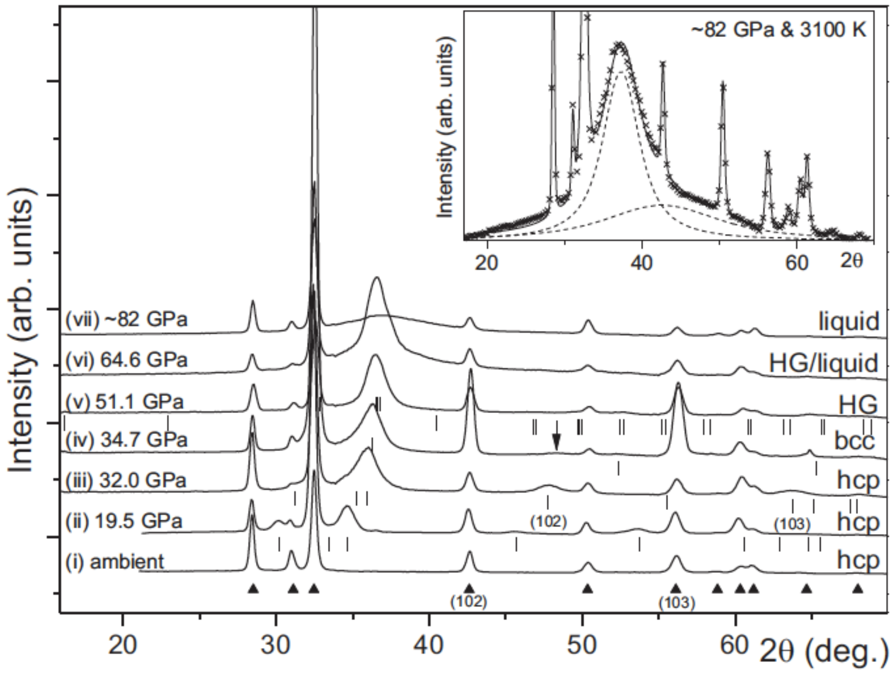


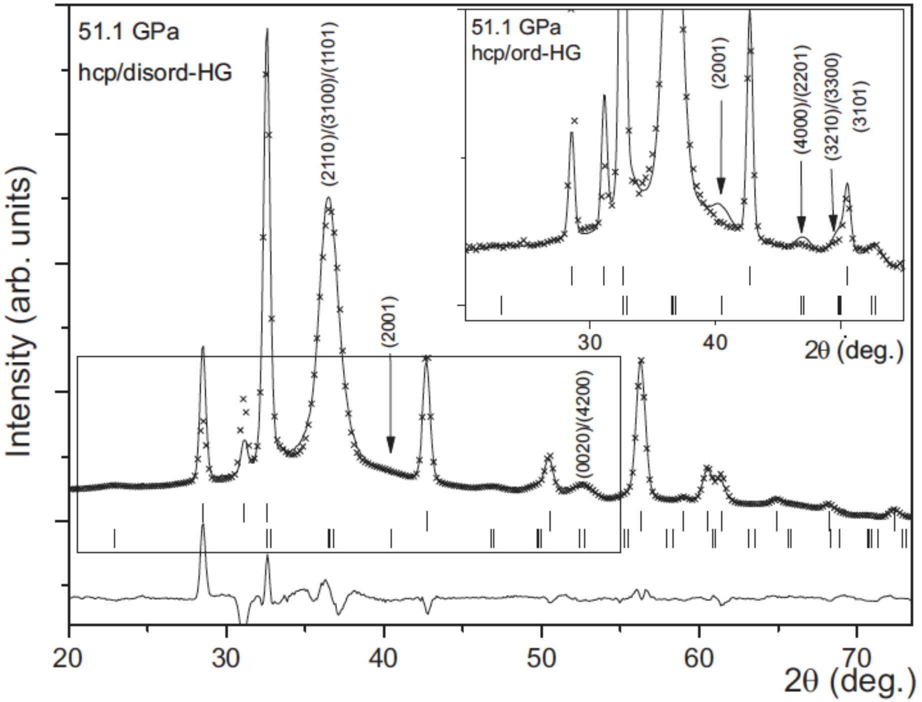
Uncompressed hcp

Compressed to 51.8 GPa

R. Briggs et al, Phys. Rev. Lett., 025501 (2017)

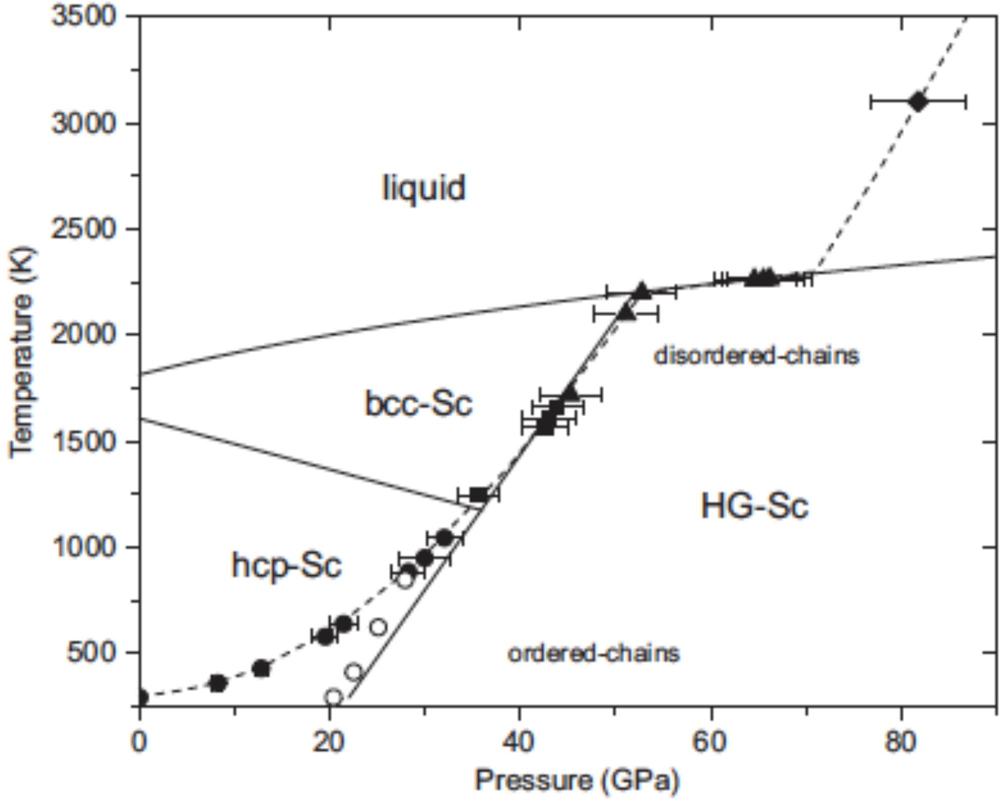
Scandium Diffraction





A disordered host-guest structure forms on nanosecond timescales

Scandium Diffraction



Proposed phase diagram including the Hugoniot. Note the temperatures are inferred, with some data from synchrotron measurements. Development of in situ temperature measurements is an on-going challenge.

R. Briggs *et al*., Phys. Rev. Lett., 025501 (2017)

‘Diffraction’ from a plasma

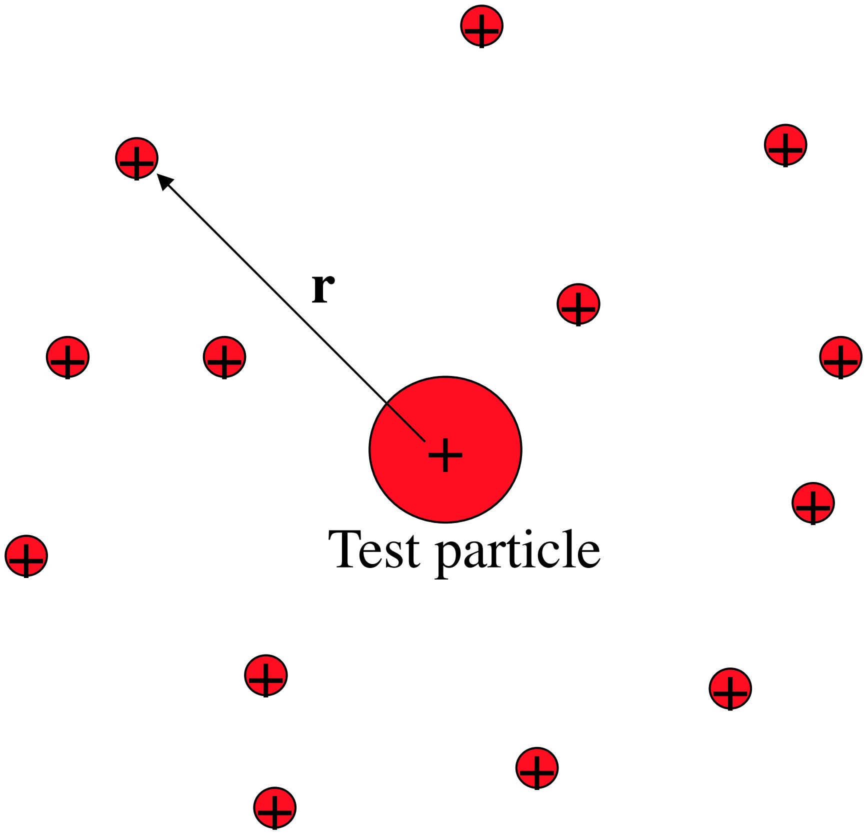
At higher shock strengths the material melts, and eventually we can shock into the plasma phase.

We can perform ‘diffraction’ (elastic scattering) from this state, and will see something that looks like a liquid diffraction pattern.

Why is this important?

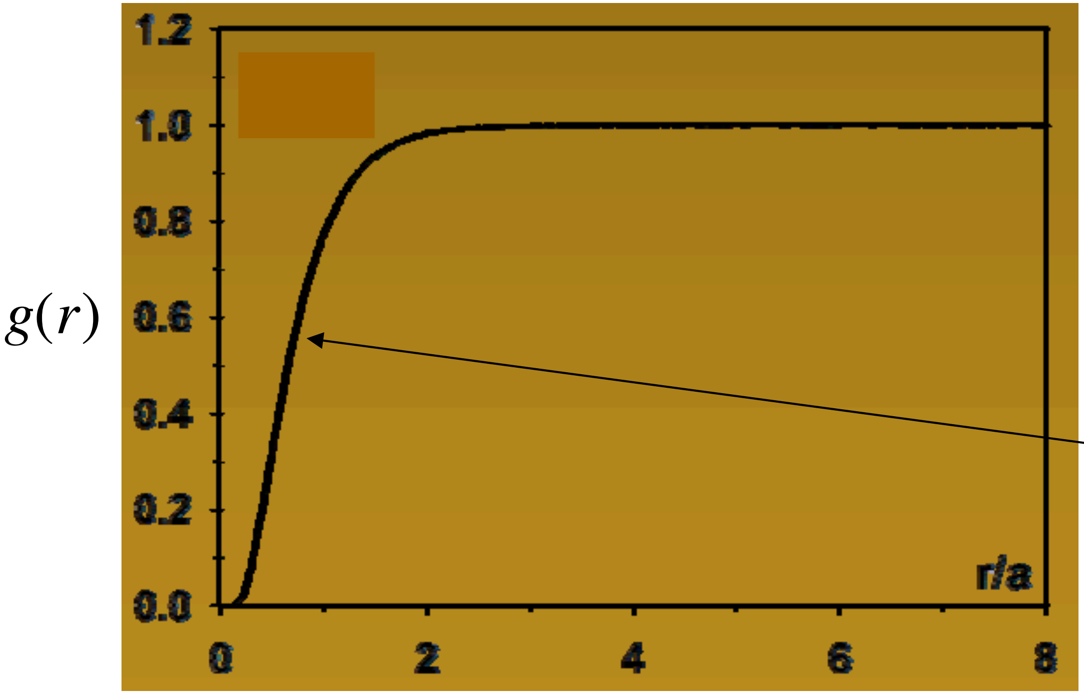
Pair distribution function

- The pair distribution function, *g(r)*, describes the probability of finding a particle at a distance **r**, given that there is one at **r**=0



Pair distribution function: simple examples (1)

- In an **ideal plasma** (gas), the ions are distributed around the test ion following the Boltzmann's relation (barometric law)
  - Look at the derivation of the Debye length

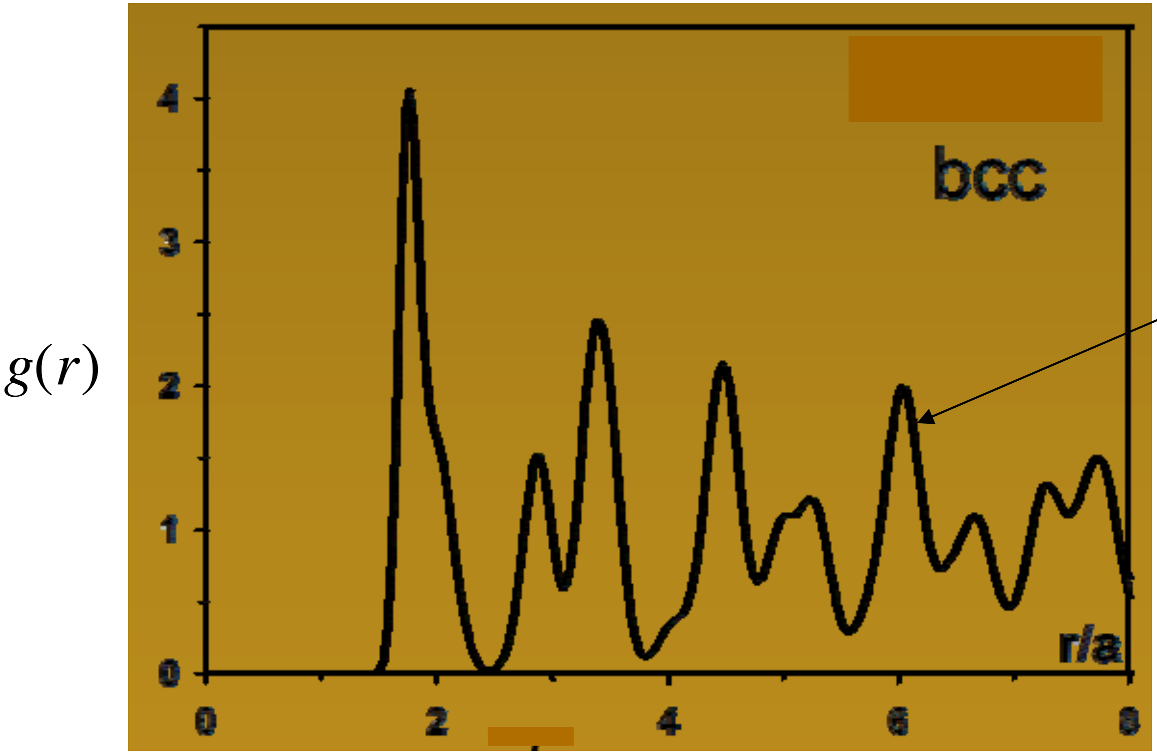


The probability of finding another ion at large distances is 1

$$g(r) \sim \exp\left(-\frac{e\phi(r)}{k_B T}\right)$$

Pair distribution function: simple examples (2)

- In an **ideal solid** (crystal), the ions are distributed in a lattice. You will find another ion at a distance **r** if the Bragg condition is satisfied

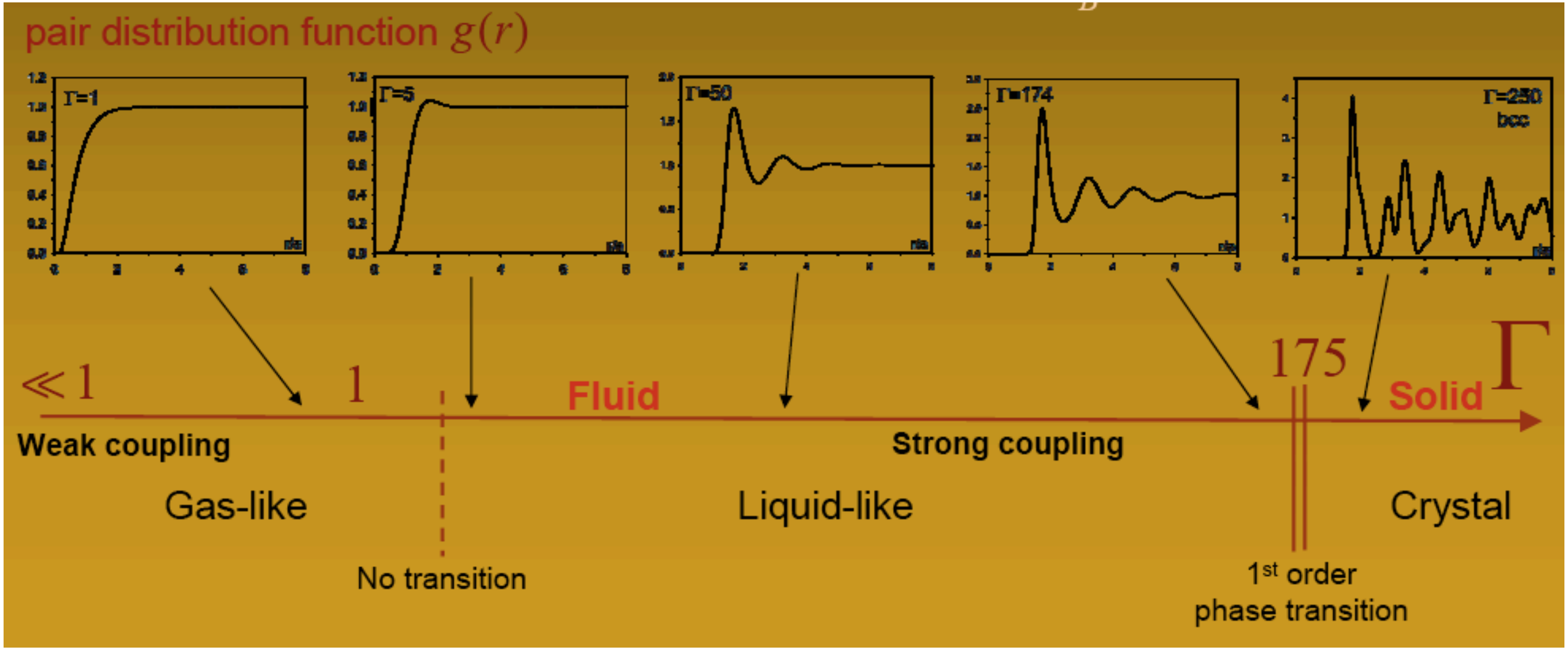


$$\vec{G} \cdot \vec{r} = 2\pi n$$

g(r) is very "spiky" and the specific details depends on the arrangements of the ions in the lattice

Pair distribution function: strongly coupled plasmas

- The strongly coupled plasma is not an ideal gas nor a solid: it exhibits intermediate properties and the corresponding pair correlation function share features common to both the solid and the gas-like behavior



Static Structure Factor (1)

- In analogy with the condensed matter theory, we describe the plasma in the reciprocal space
- The static structure factor *S(k)* is defined as

$$S(\mathbf{k}) = \frac{1}{N}\langle \rho_{\mathbf{k}}(t)\rho_{-\mathbf{k}}(t)\rangle = \frac{1}{N}\langle \rho_{\mathbf{k}}(0)\rho_{-\mathbf{k}}(0)\rangle$$

Time invariance!

$$S(\mathbf{k}) = 1 + \frac{1}{N}\langle \sum_{i\neq j=1}^{N} \mathrm{e}^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\rangle$$

Using the definition of the point-particle distribution:

$$\rho_{\mathbf{q}}(t) = \sum_{i=1}^{N} \mathrm{e}^{-i\mathbf{q}\cdot\mathbf{r}_i(t)}$$

Static Structure Factor: Internal Energy (2)

- The internal energy per particle is then:

$$\frac{E_{int}}{N} = \frac{E_{int}^{ideal}}{N} + \frac{1}{N}\langle \mathcal{H}_{Cou} \rangle = \frac{3}{2}k_B T + \underbrace{\frac{1}{2V}\sum_{\mathbf{k}\neq 0} V_k \left[S(\mathbf{k}) - 1 \right]}_{}$$

In an ideal plasma, $S(k)=1$, thus the internal energy is just $(3/2)k_B T$

- If we know the structure factor, we can calculate the internal energy and from that all the thermodynamic properties (EOS = equation of state)
- The EOS of a strongly coupled plasma is different from that of an ideal plasma!

The Debye-Hückel Theory

- If we now substitute the Maxwell-Boltzmann distribution in the fluctuation-dissipation theorem and solve the integral, we obtain the Debye-Hückel approximation for the static structure factor

$$S(k) \equiv S_{DH}(k) = \int d\omega S(k,\omega) = \frac{k^2}{k^2 + k_D^2}$$

- The corresponding pair distribution function is then obtained by Fourier transform (as we have shown earlier)

$$\begin{aligned} g_{DH}(r) &= 1 + \frac{1}{(2\pi)^3 n_i} \int_0^\infty \int_0^\pi 2\pi \sin(\theta) k^2 \mathrm{e}^{ikr\cos(\theta)} \left[S_{DH}(k) - 1\right] d\theta dk \\ &= 1 - \frac{k_D^2}{4\pi n_i r} \mathrm{e}^{-rk_D} \end{aligned}$$

A Simple Equation of State

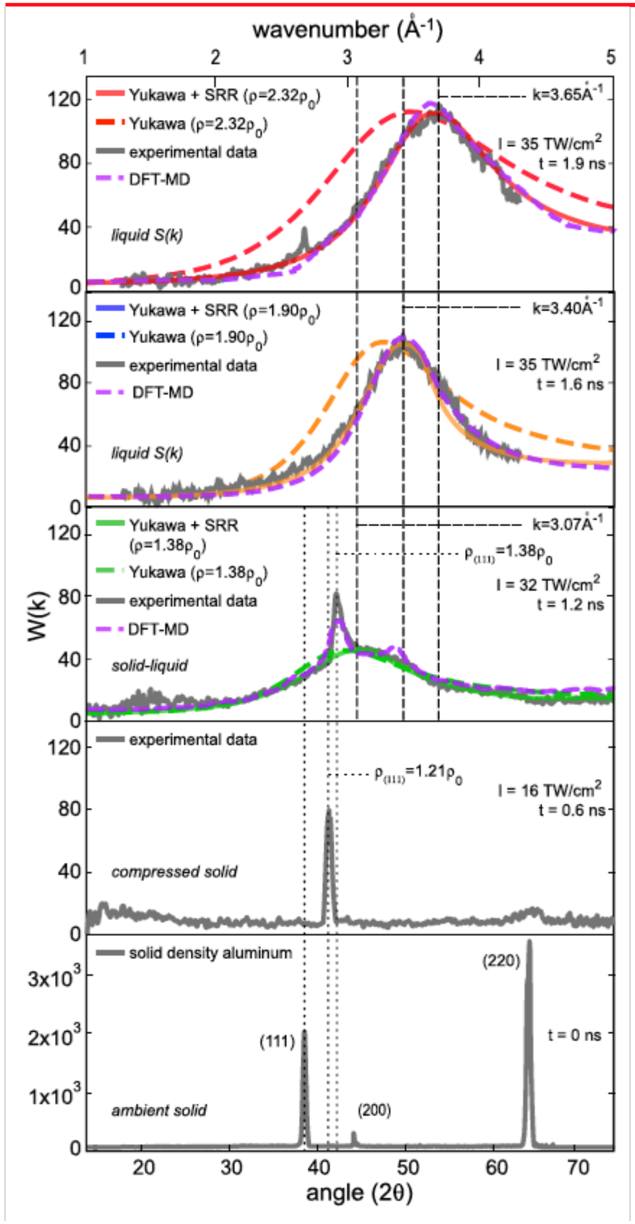
- We can use the simple Debye-Hückel model to derive a modified equation of state for a weakly coupled plasma

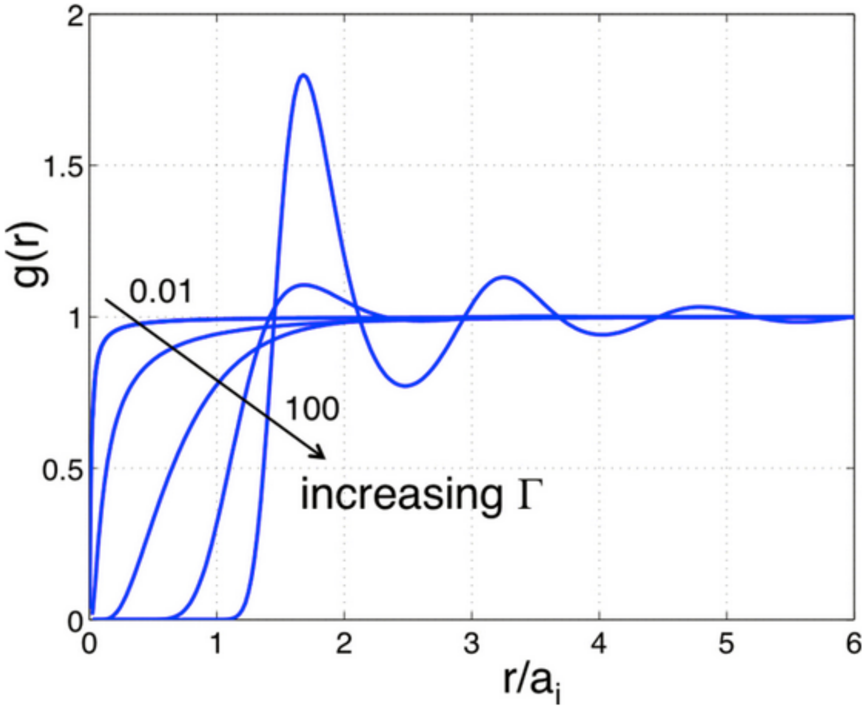
$$\frac{E_{int} - E_{int}^{ideal}}{N} = \frac{1}{4\pi^2} \int_0^\infty k^2 V_k \left[S(k) - 1 \right] dk \ = -k_B T \frac{\sqrt{3}}{2} \Gamma^{3/2}$$

Coulomb potential Debye-Hückel structure factor

- We see that the effect of weak coupling is to reduce the internal energy from the ideal value (3/2)$k_B T$
- For Γ=0.1 this accounts for about 2% reduction from the ideal value

Experiments at LCLS





See Glenzer et al, Journal of Physics B: Atomic, Molecular and Optical Physics, 49 (2016) 092001.

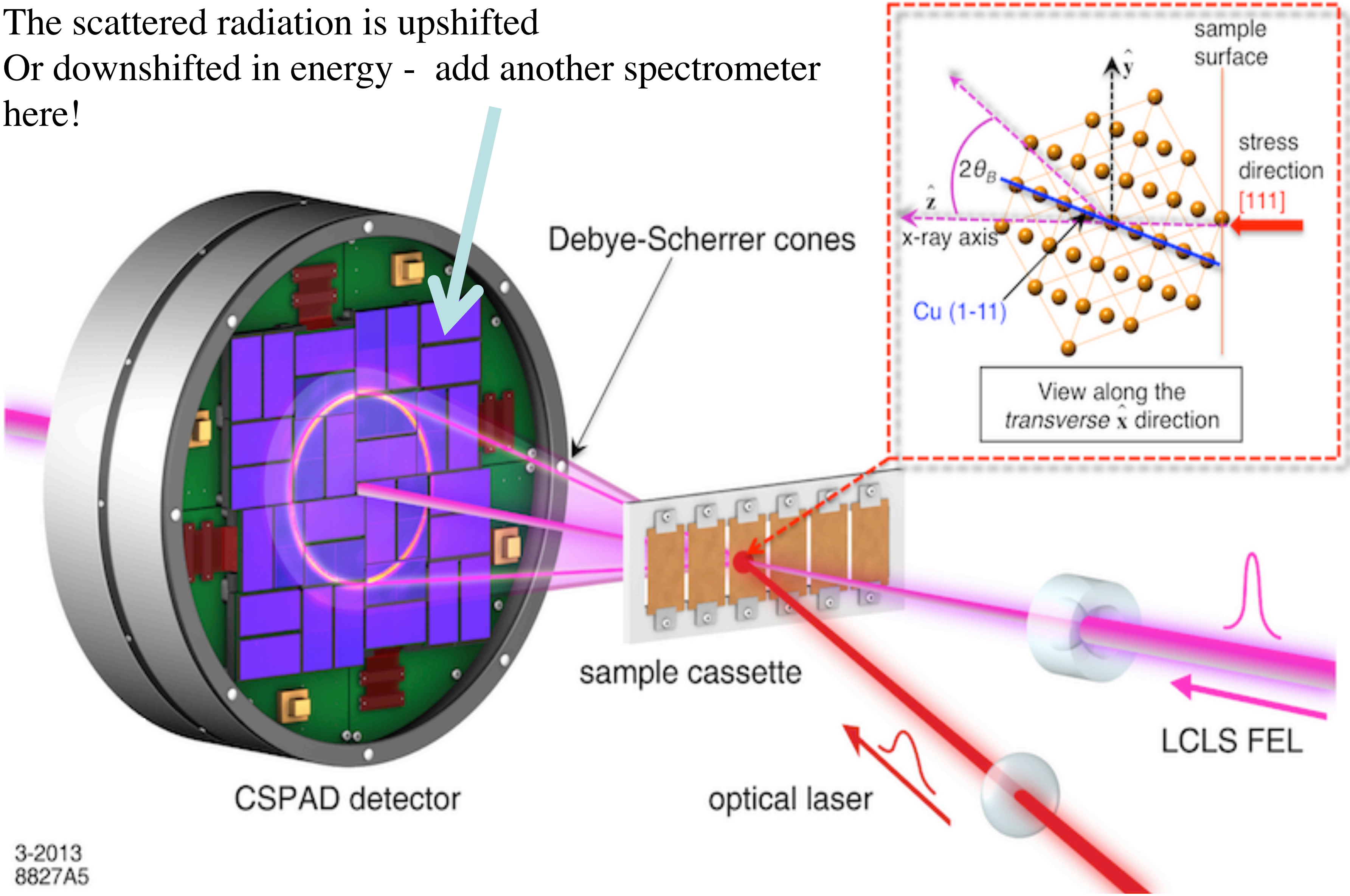
Inelastic Scattering

- The diffraction we have been discussing is the elastic scattering (from the static structure)
- As in any system, the x-rays can also gain or lose a little energy by interacting with other modes of the system - for a solid it would be phonons - for a plasma it is plasmons - which give a measure of electron density.
- In the field this is known as Thomson scattering.
- Note - in a warm 'classical' plasma the plasmon frequency depends on density and temperature…

$$\omega^2 = \omega_{\mathrm{p}}^2 + 3k^2 \frac{k_{\mathrm{B}} T}{m_e}$$

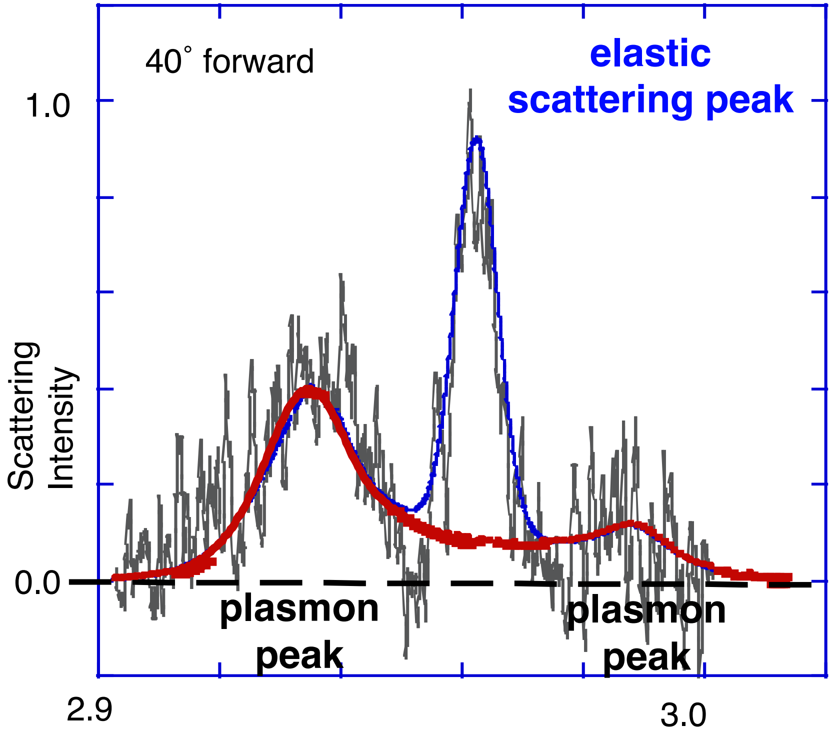
Debye-Scherrer Diffraction (inelastic scattering)

The scattered radiation is upshifted
Or downshifted in energy - add another spectrometer here!

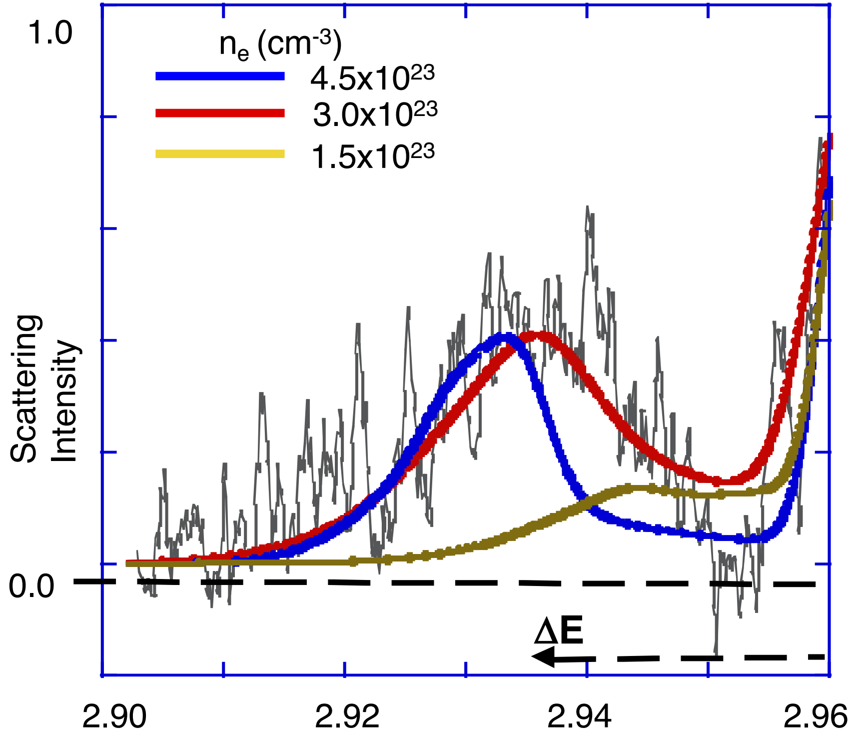


Thomson forward scattering – laser plasma source plasmons provide additional data

Best fit found at 12 eV from scattering from Be

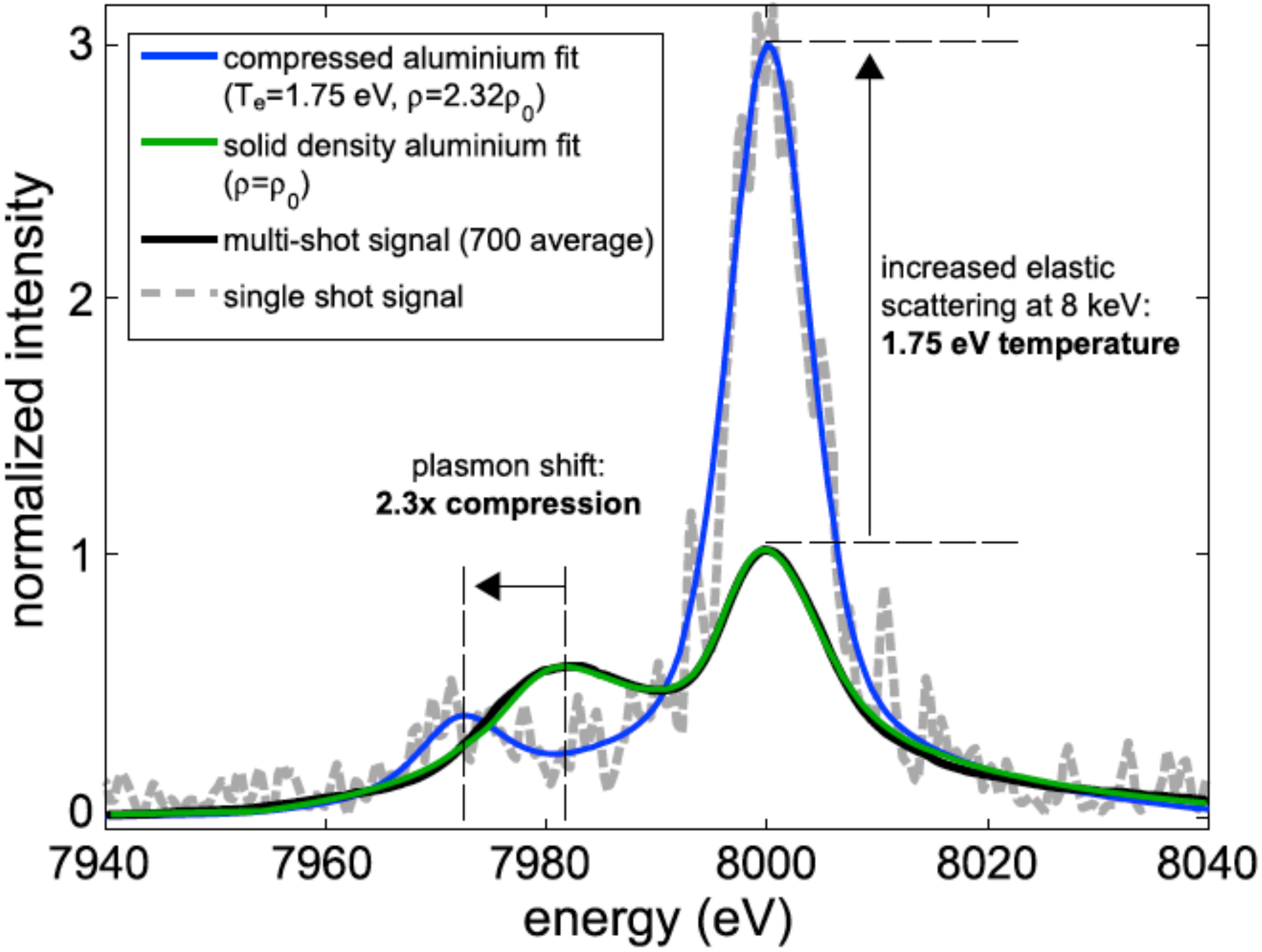


Best fit found at $3x10^{23}$ cm^{-3} from plasmon spectrum



- Plasmon peak intensity related by detailed balance.
- Experiments with independent T_e measurement are needed to determine correct approximation for collisions

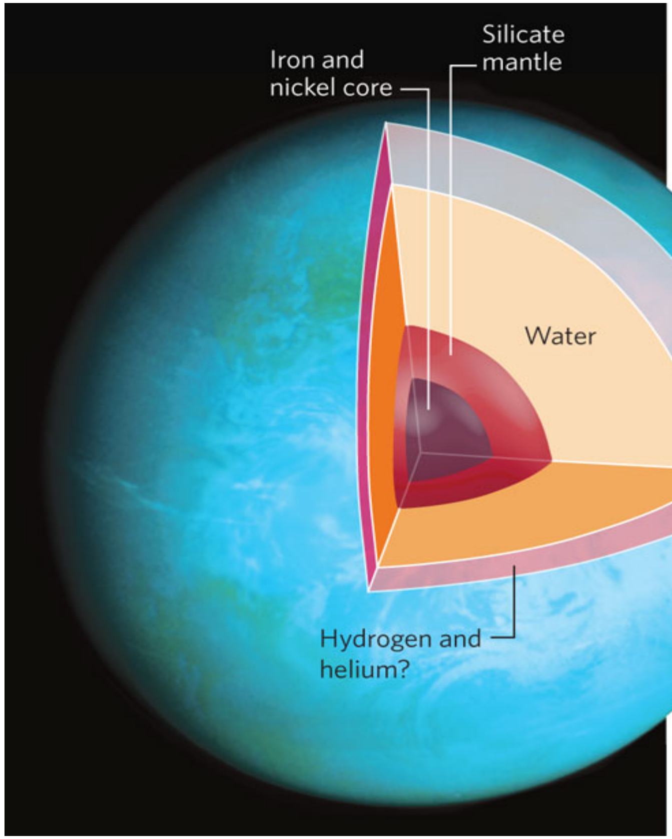
Thomson scattering – recent LCLS data



Quasi-Isentropic Compression

- Thus far we have been considering creating transient high pressure solid-state matter via shock-compression.
- Shocks generate copious entropy, and the material is heated. Typical metals shock-melt (the Hugoniot crosses the melt line) between 0.5-3.0 Mbar.
- Exploring solid-state matter at higher pressures requires ramp ‘quasi-isentropic’ compression, by ‘slowly’ applying pressure to the material (how slow is slow?)
- Motivation – example - exoplanets.

Masses and radii of planets



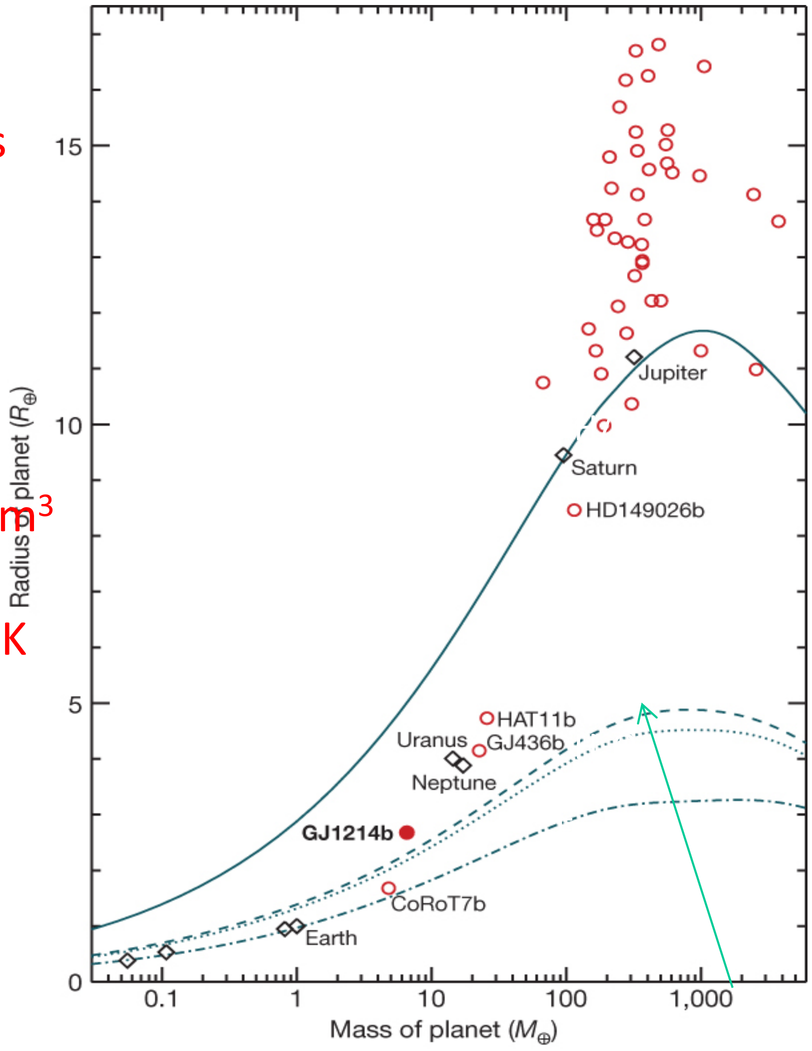
Orbital Period: 1.58 days

Mass: $6.55 \pm 0.98\ M_E$

Radius: $2.68 \pm 0.13 R_E$

Density: $1.87 \pm 0.40\ kg/m^3$

Equilibrium Temp: ~500 K

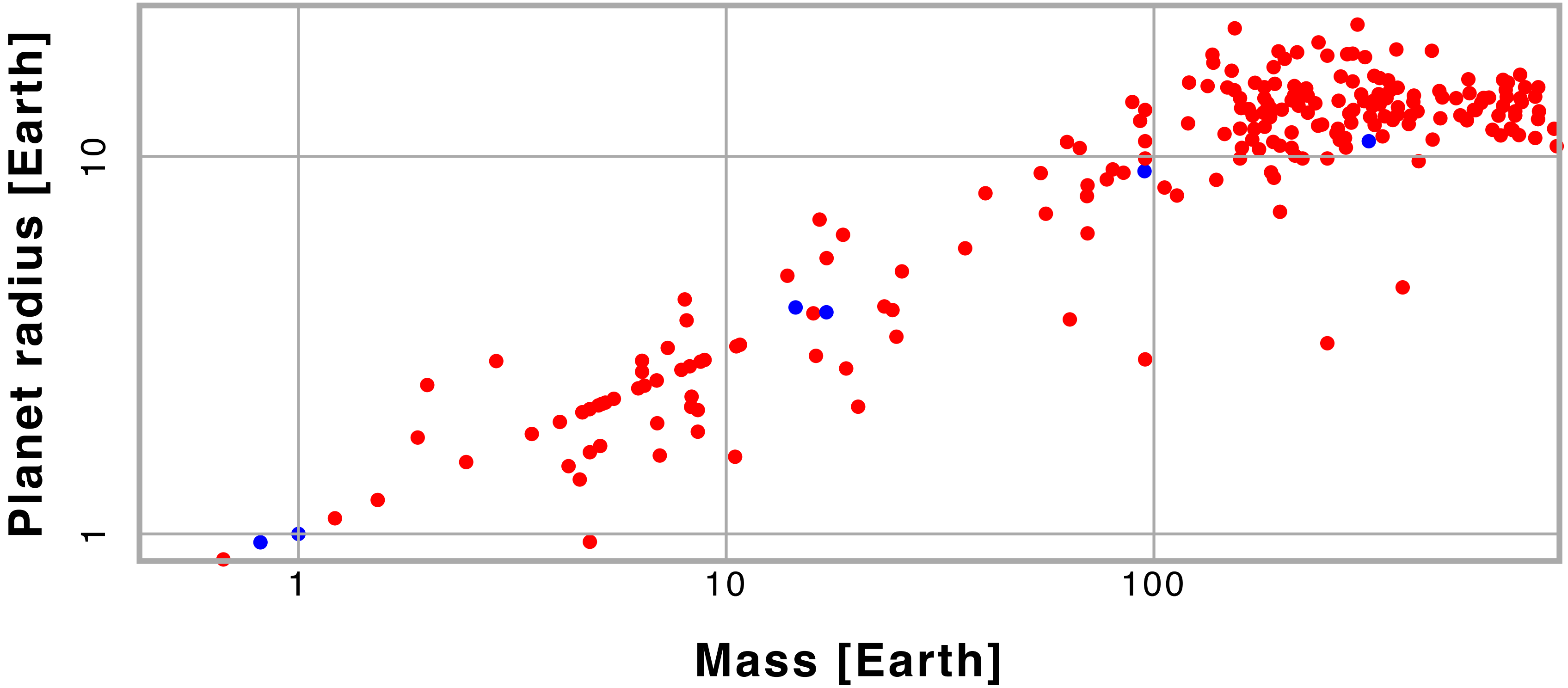


75%H_2O, 22% sil., 3% Fe

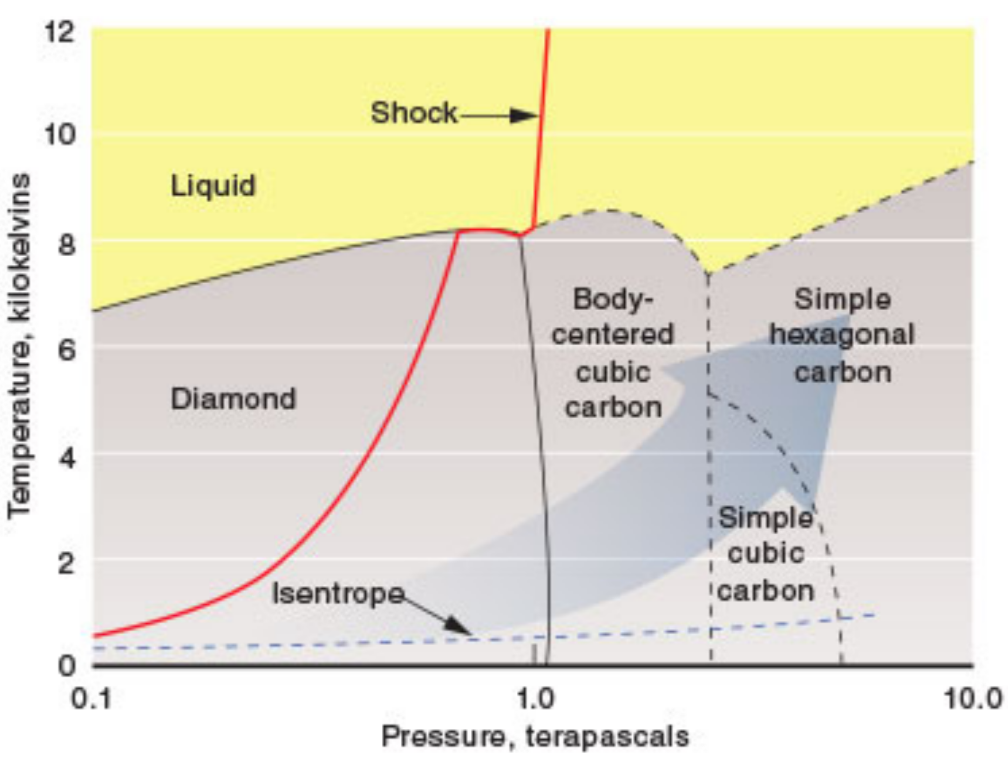
GJ1214b – A Water Planet?

Currently we rely on DFT Calculations for compressiblity. Can we measure this directly experimentally?

Mass and Radius Constrain Composition



Shocks and Isentropes



PHYSICAL REVIEW B **85**, 024112 (2012)

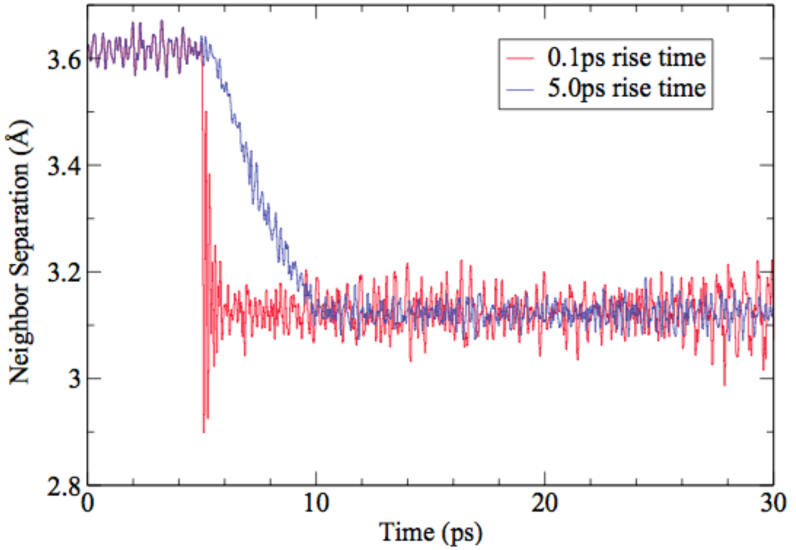


FIG. 1. (Color online) The interatomic spacing as a function of time for two particular atoms in a quasi-1D chain that has been subjected to ramp compressions with rise times of 0.1 ps and 5 ps. Note that the thermal motion is identical in the two cases prior to compression.

Isentropic compression (usually) is below the melt line

Gruneisen parameter for lattice spacing a, sound speed c:

$$\Gamma = \left(\frac{\partial \ln T}{\partial \ln \rho}\right)_S = \left(\frac{\partial \ln \theta_D}{\partial \ln \rho}\right)_S$$

$$\theta_D \approx \frac{\hbar c}{a k_B} \propto c\rho^{1/3}$$

$$M < x^2 > \omega_D^2 \approx k_B T$$

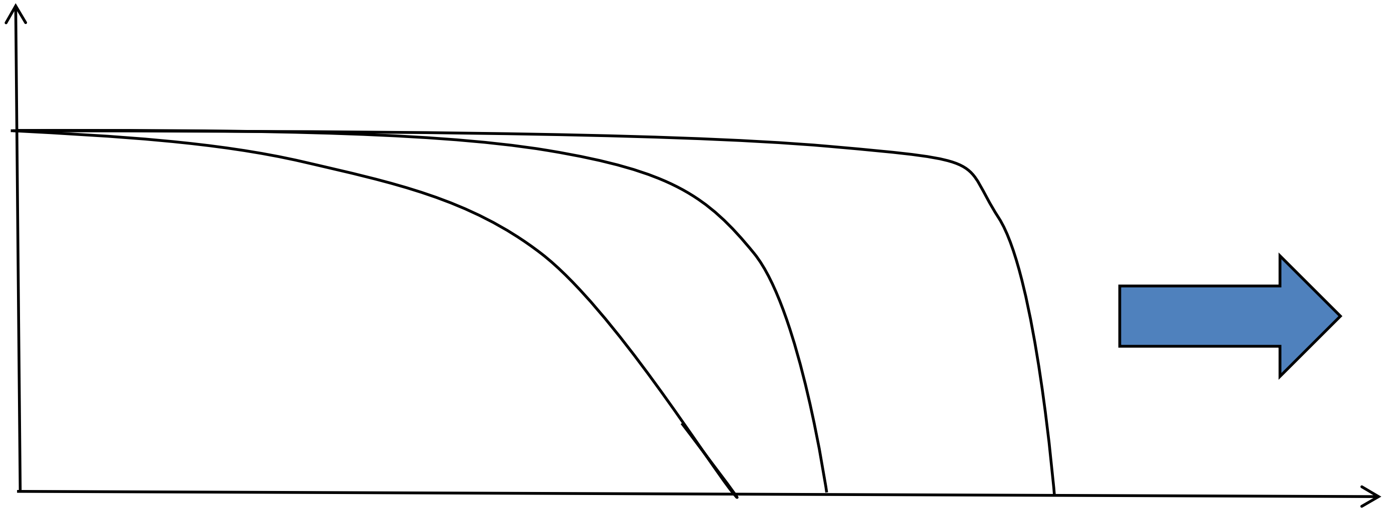
Lindemann melting criterion is x ~ 0.1 a

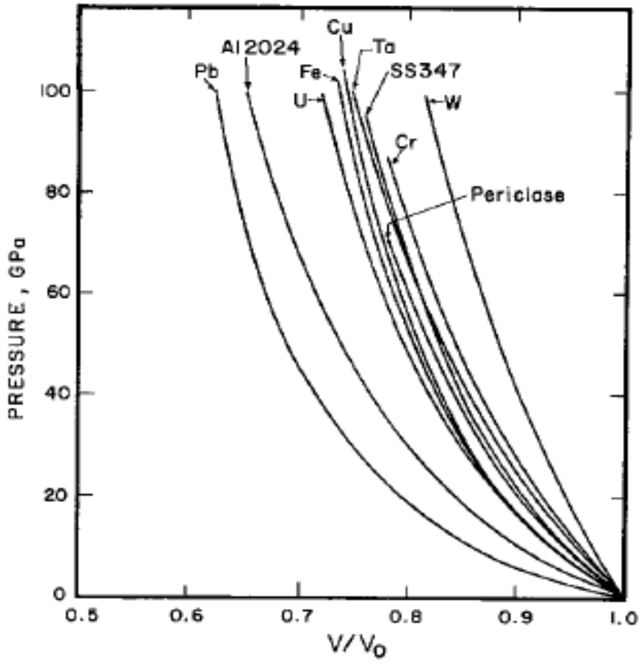
$$T_{\rm melt} = \theta_D^2 \rho^{-2/3}$$

$$\frac{d \ln T_{\rm melt}}{d \ln \rho} = 2(\Gamma - 1/3)$$

So in this simple model the isentrope lies below the melt line for Γ > 2/3

Shock/Ramp Compression





As the speed of sound generally increases with compression, a strong compression wave will steepen into a shock, where there is a discontinuity in density, temperature, and energy across the shock front.

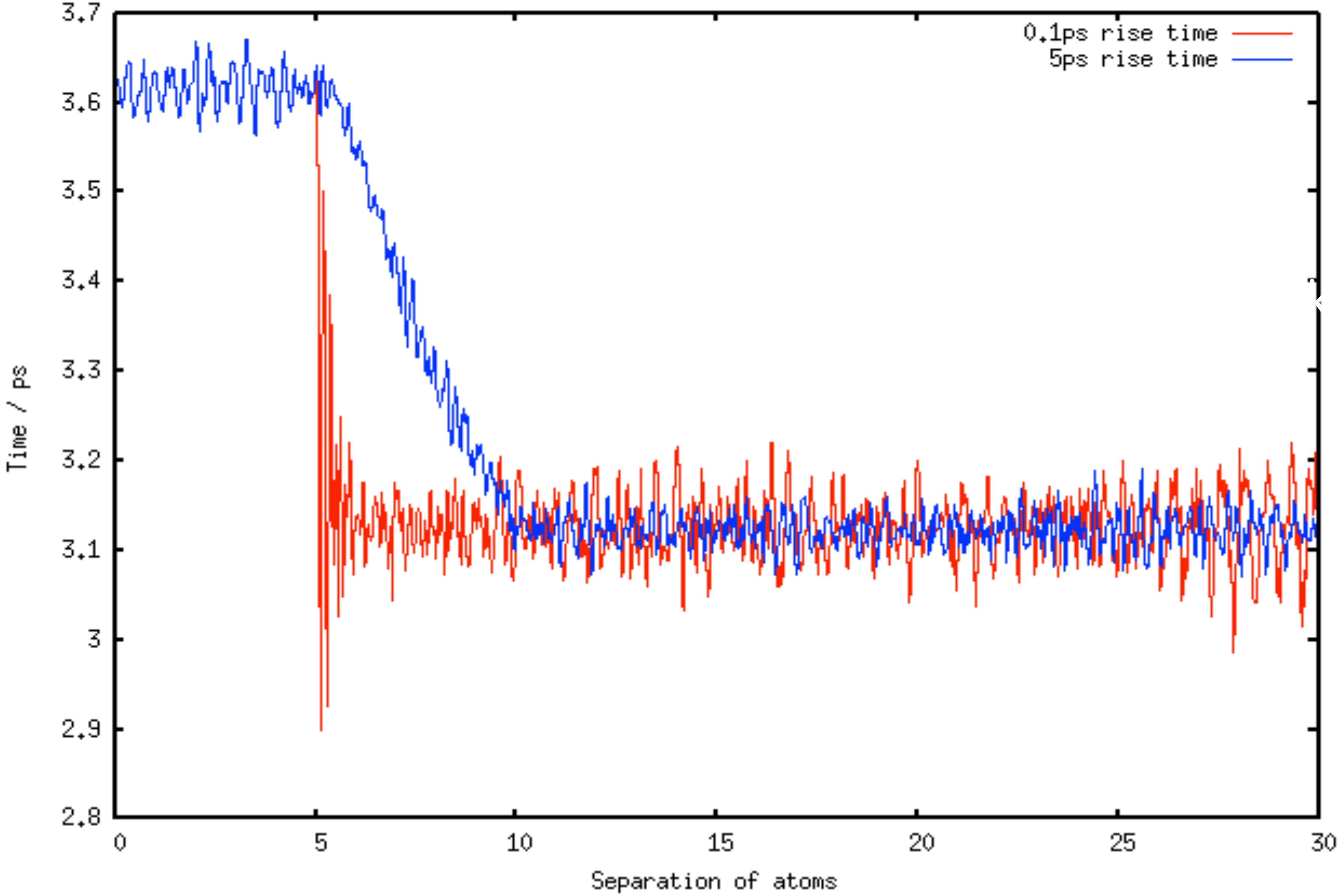
We need to ‘ramp’ compress our sample, but probe before it steepens to a shock.

When is a shock a shock?

How fast is the compression in a shock wave?

Alternatively, how slowly do we need to compress solids to keep them on (or at least close to) isentropes?

1-D Elastic Shock/Ramp



Swegle-Grady 4th Power Law

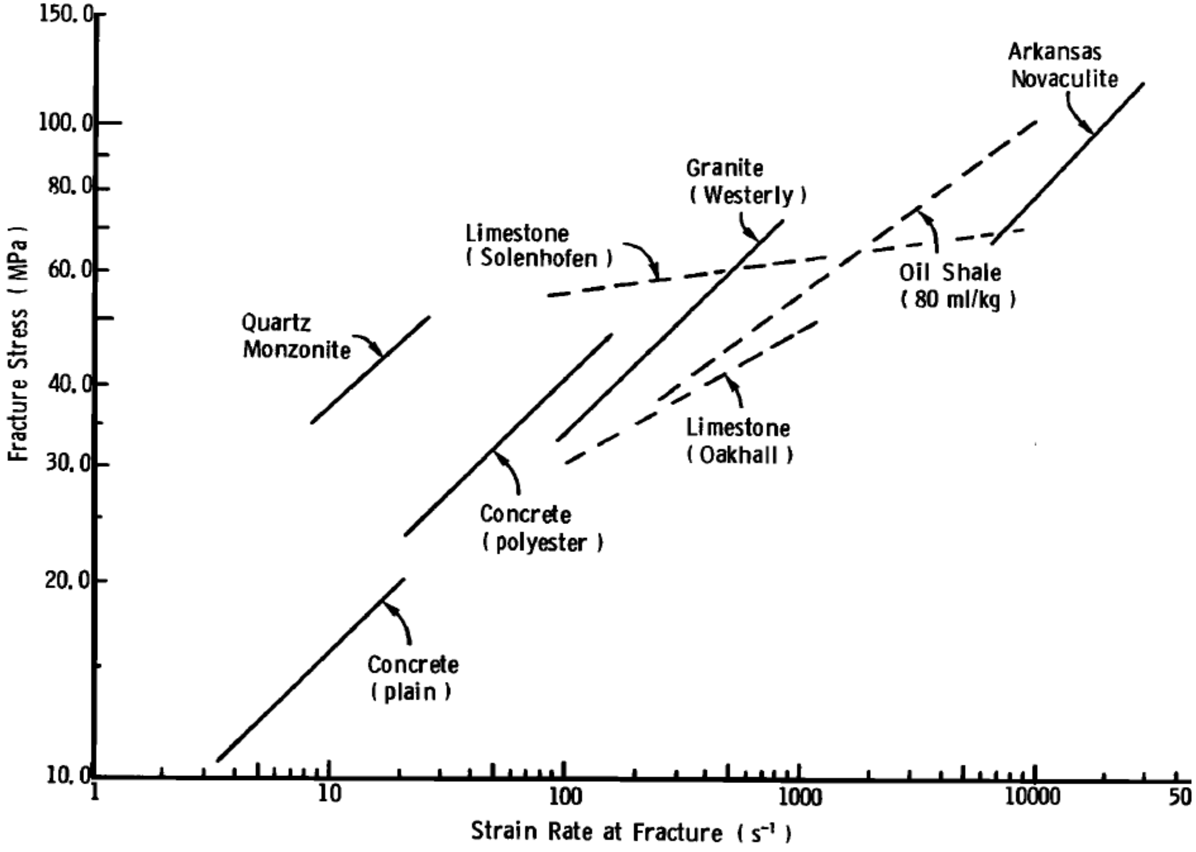
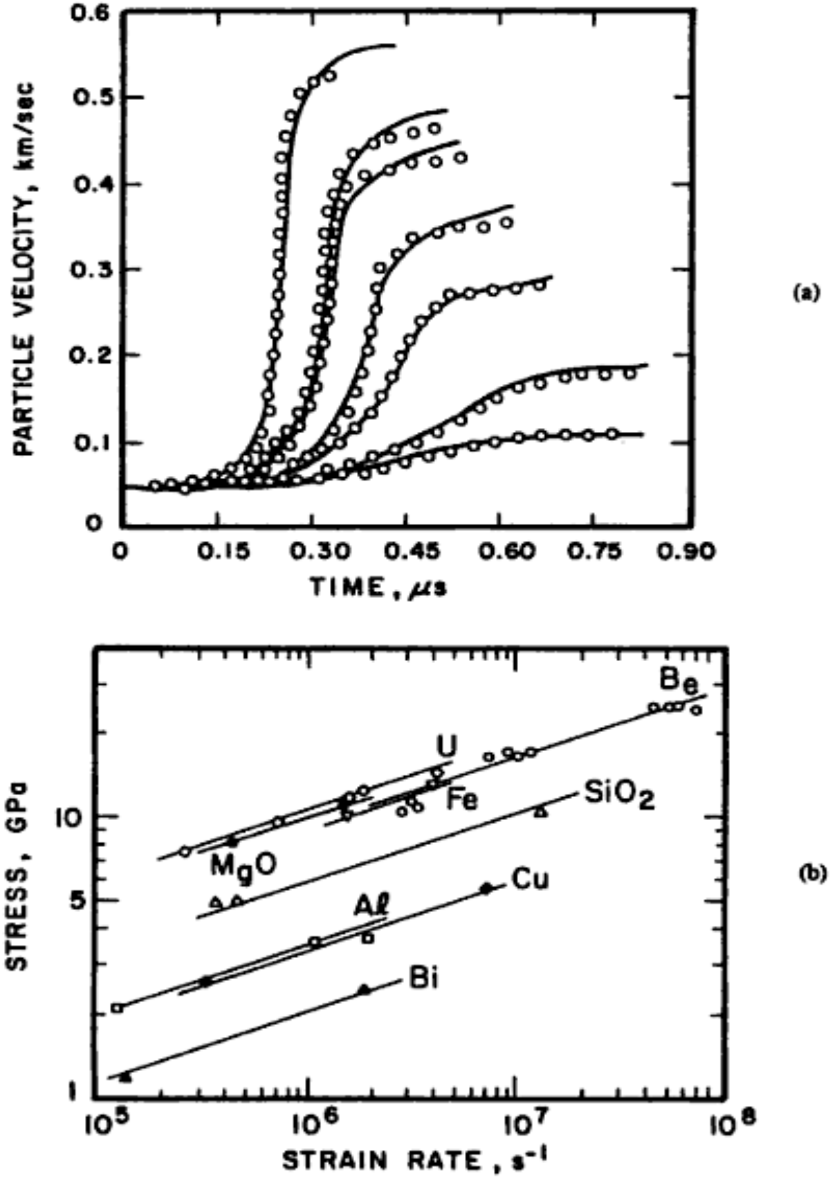


Fig. 1. Strain rate dependence of fracture stress.

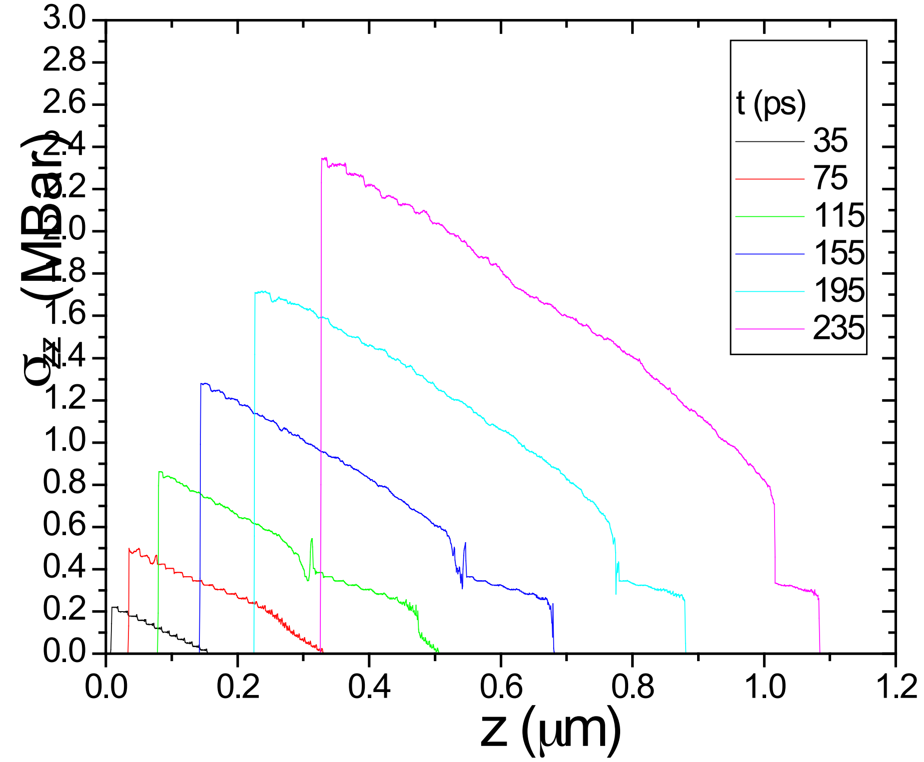


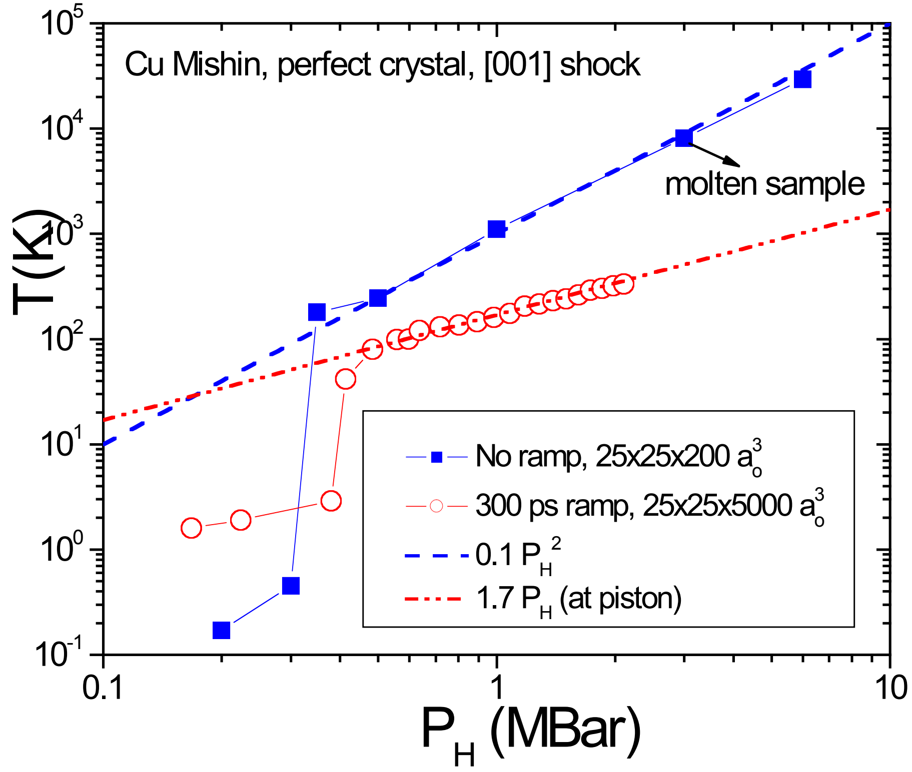
Grady & Lipkin, Geophys. Res. Lett., 7, 25

“Dynamic Behavior of Materials” Marc Meyers

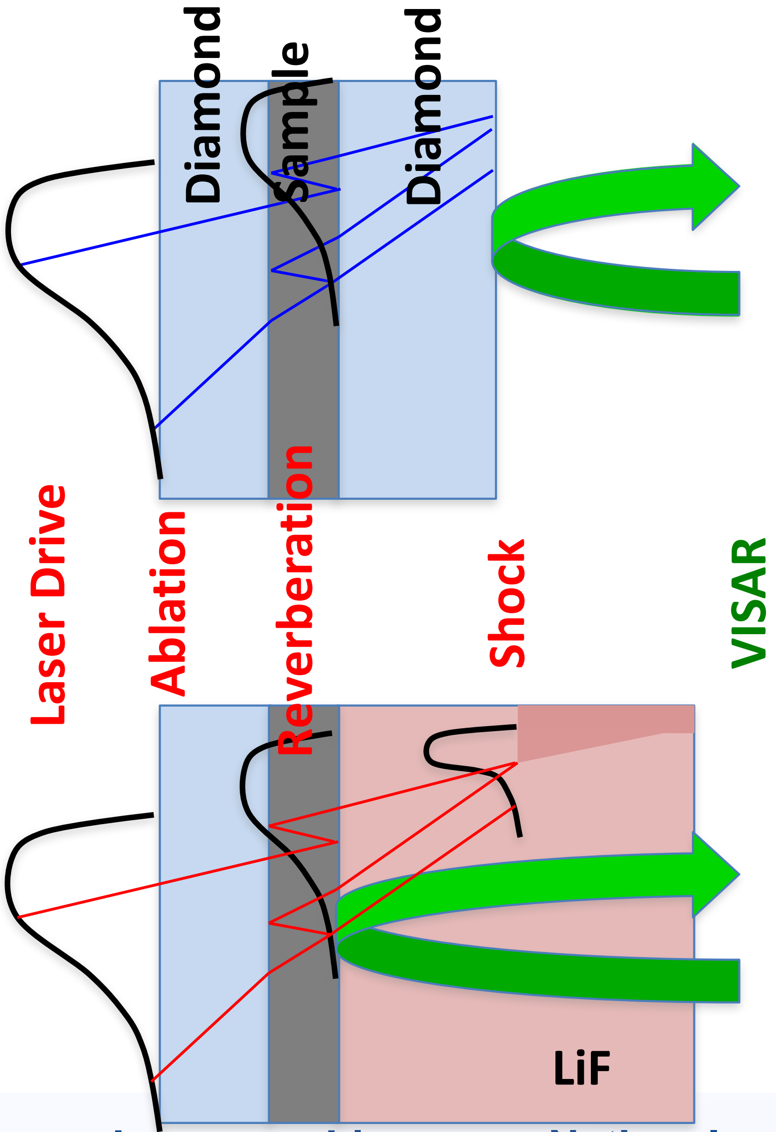
Molecular Dynamics Simulations of ramp compression

Cu Mishin [001], 25x25x5000 fcc cells (1.8 μm), 300 ps ramp, $U_{p\text{-}max}$=3.5 km/s ➔P~2.5 Mbar





Sandwich Ramp-Compression



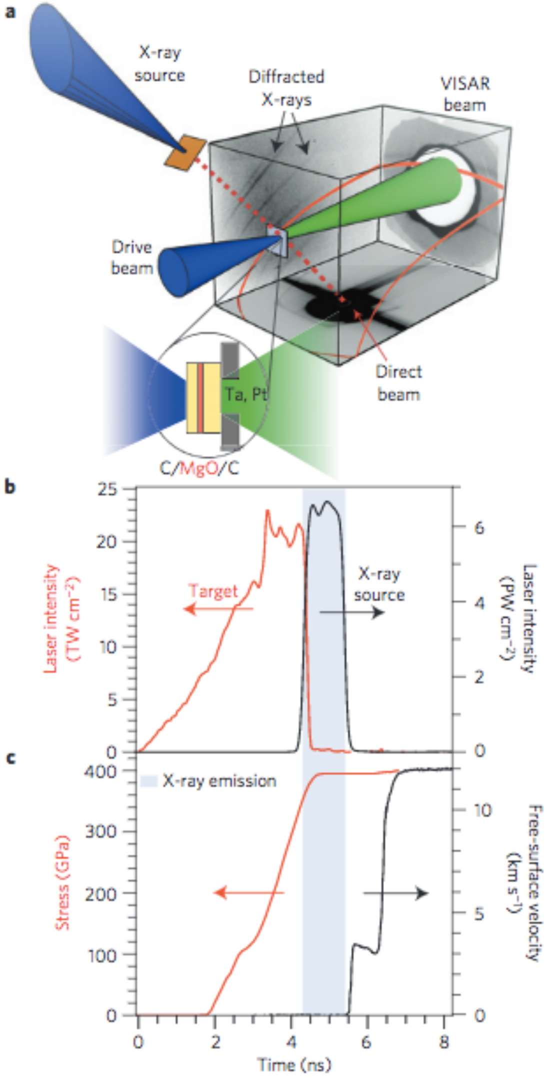
The LiF or Diamond interfacial pressure is the same as in sample

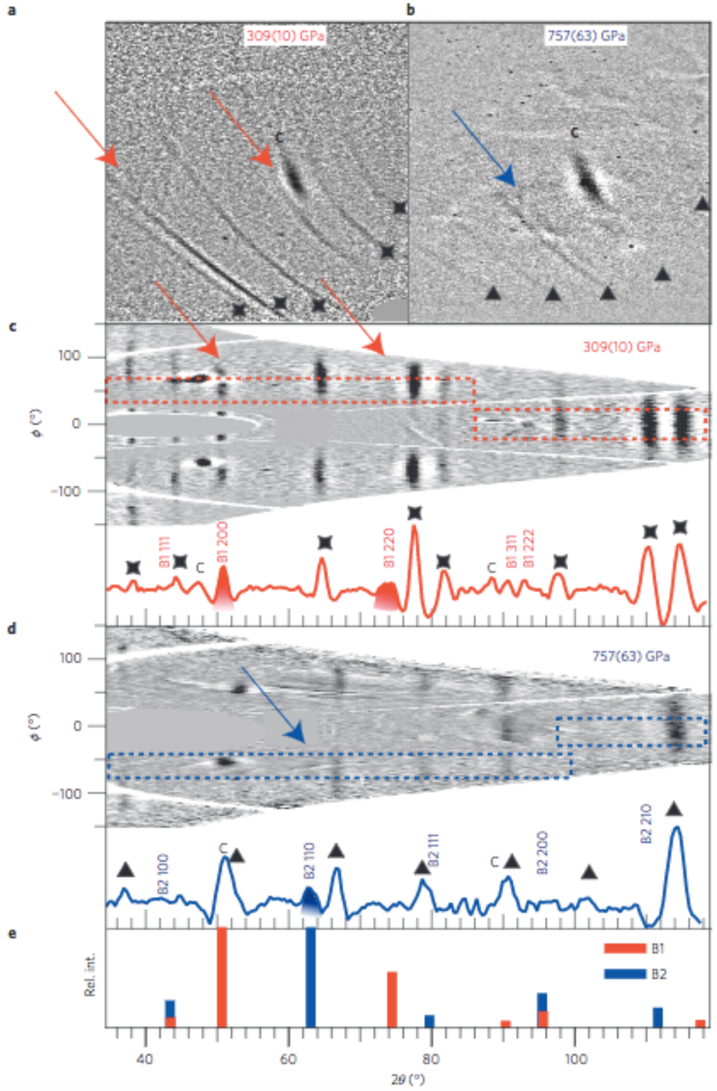
If we know the EOS of LiF or Diamond we can find the Pressure in the sample using the VISAR diagnostic

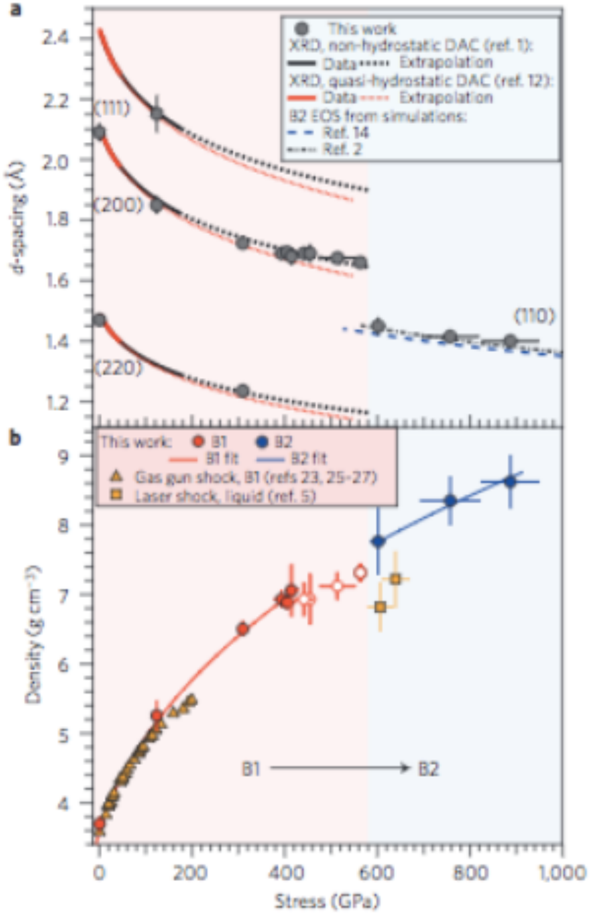
Using this target design, we believe we can ramp compress samples to ~30 Mbar, Hold the state for several ns, Determine the pressure, and Make a measurement.

XRD, XAFS, XANES, Reflectivity, ... *Temperature remains the most important parameter that we do not know how to measure.*

Diffraction at TPa Pressures







F. Coppari *et al.*, Nature Geoscience, 6, 926 (2013).

Future prospects

- Solids under TPa pressures are only just starting to be explored.
- We are commissioning a 100 J optical laser on XFEL that will operate at 10 Hz and allow data collection on this timescale.
- Future challenges include making temperature measurements (as well as pressure and density), to get fuller information on the phase diagram.

The dipole laser at HiLASE - a copy will be installed at XFEL

Summary

- TPa -10^{12} Nm^{-2} (10 Mbar) environments occur in hot dense plasmas, as well as very cold dense solids:
  - Hot – halfway to the centre of the sun.
  - Cold – the centre of the giant planets.
- New opportunities to make hot (several hundred eV - 10^6 K, plasmas) at exactly solid density, exist with femtosecond X-Rays.
- Standard Plasma theory breaks down for dense plasmas, where there are few (<1) particles in a Debye Sphere. We need to measure EOS, transport, ionisation etc etc etc.
- X-Ray FELs are allowing us to study how solids deform rapidly, and to see rapidly formed new phases. Ablation pressures are much higher than pressures that can be achieved statically.
- Quasi-isentropic compression of solids via the application of ramped laser-plasma pressures has allowed the creation of solids at densities never before created on earth, and at pressures of relevance to the science of the giant planets and exoplanets.