Direct Temperature Measurements in Solids via Inelastic X-Ray Scattering under Shock and Ramp Compression

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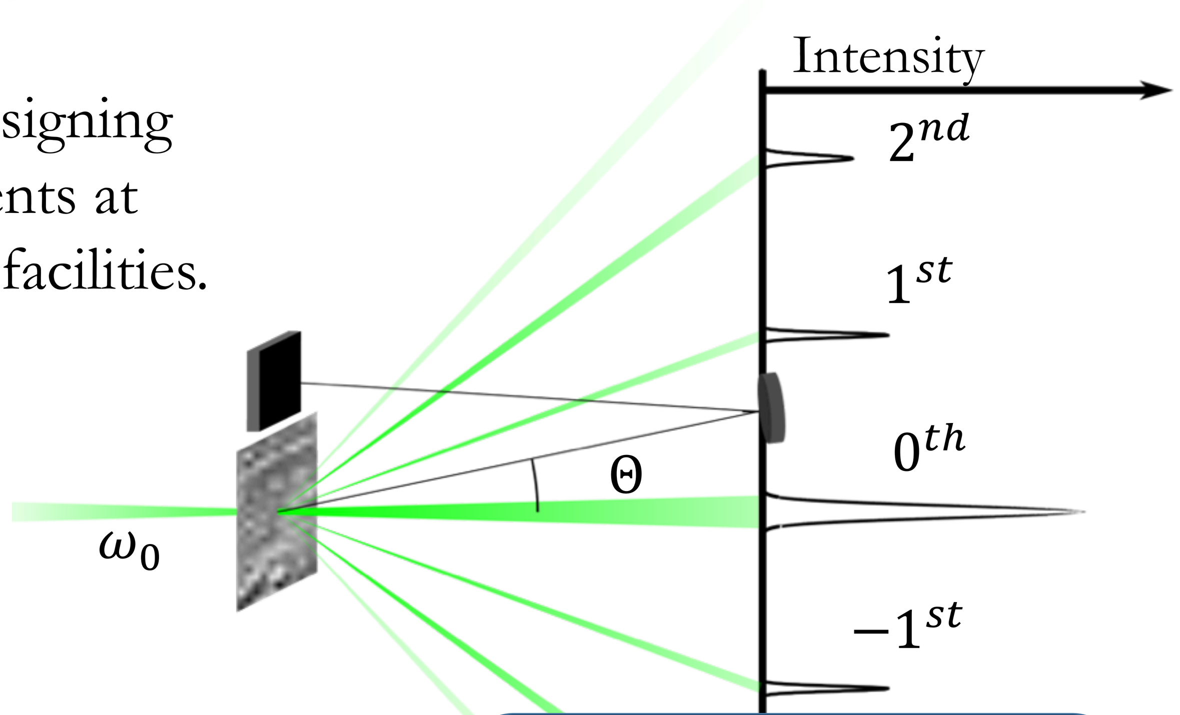
OXFORD CENTRE FOR HIGH ENERGY DENSITY SCIENCE

In the known universe, condensed matter at extreme pressures occurs in a range of unique and fascinating systems. The recent burgeoning field of exoplanet physics has created a high demand for understanding solids at terapascal pressures[1] - a regime that can be obtained through laser ablation techniques performed at high-energy laser facilities[4,5,6,12]. Whilst X-ray diffraction can obtain structural information[9,10,11,13,15] and thus density, and pressures can be inferred from Velocity Interferometry (VISAR)[2], absolute temperatures are more difficult to measure. Previous work using Extended X-ray Absorption Fine Structure (EXAFS) allows one to deduce the ratio T/T_D, where T_D is the Debye temperature, but this itself is pressure dependent, and may change drastically across a phase transition. For a deeper, fundamental understanding of these extreme phases, the absolute temperature T is of interest. We propose using inelastic X-ray scattering[3,7,8,14,16] from phonons at 4th generation sources to determine the absolute temperature[14] from the ratio of Stokes and Anti-Stokes scattering under shock and ramp compression. The feasibility is assessed through estimation of the cross sections and required resolution. Synthetic phonon spectra are calculated in various materials under shock and ramped compression using large-scale molecular dynamics simulations in LAMMPS that serve as a controlled theoretical benchmark for future experimental results.

High Energy Density Physics

- High energy physics at terapascal scale for fusion and planets
- Currently no method to find temperature in dynamic experiments
  - The Debye-Waller factor W is function of $\frac{T}{T_D}$.
  - Then again, Debye temperature T_D function of T.
- To properly interpret experiments and obtain a complete equation of state p, V, and T, one needs to find either T or T_D separately.
- At this, we are designing ablation experiments at high-energy laser facilities.

$$Q = 2k \sin\frac{\Theta}{2}$$



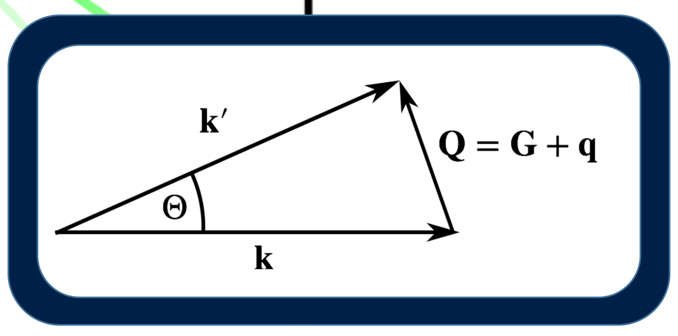


Figure 1: Typical experimental geometry in a transmissive scatter experiment. Laue diffraction via Thomson scattering off a thin sample foil with elastic peak projection in green.

Inelastic X-Ray Scattering

- We are investigating inelastic X-ray scattering (IXS) in solids
- From differential cross section:

$$\frac{I_1(\omega_0-\omega_q)}{I_2(\omega_0+\omega_q)} = e^{\frac{\hbar\omega_q}{k_B T}}.$$

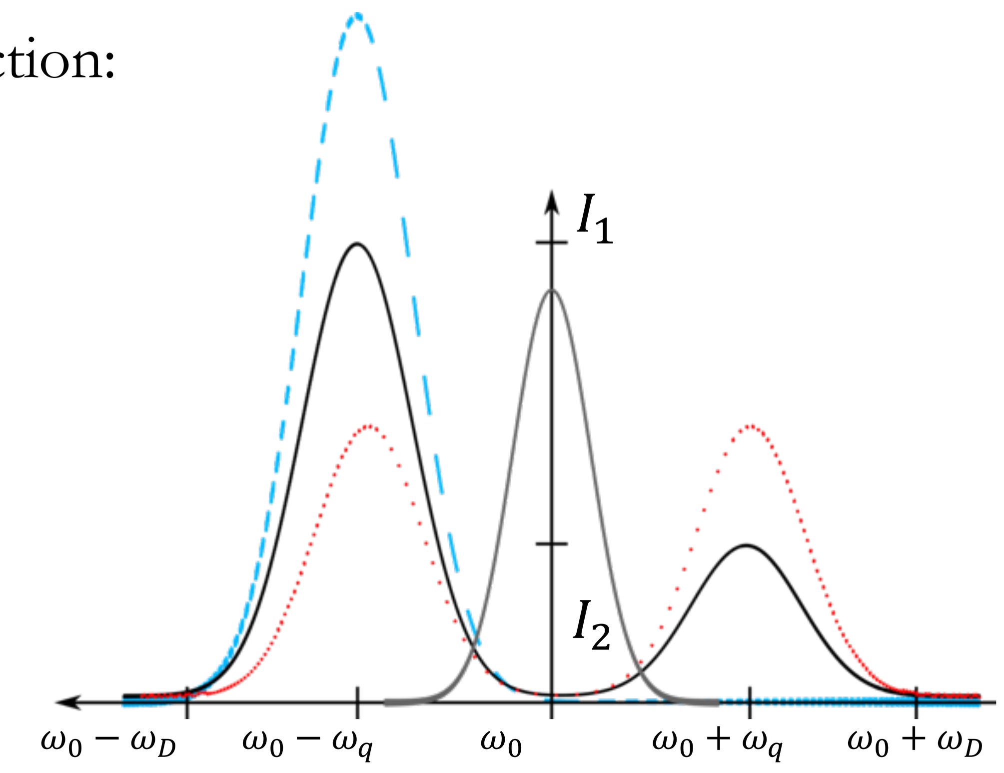


Figure 2: Stokes and Anti-Stokes peaks. Their typical spacing is ±40 meV with an width of roughly 50 meV. Blue line for a cold sample, red hot and black close to the Debye temperature.

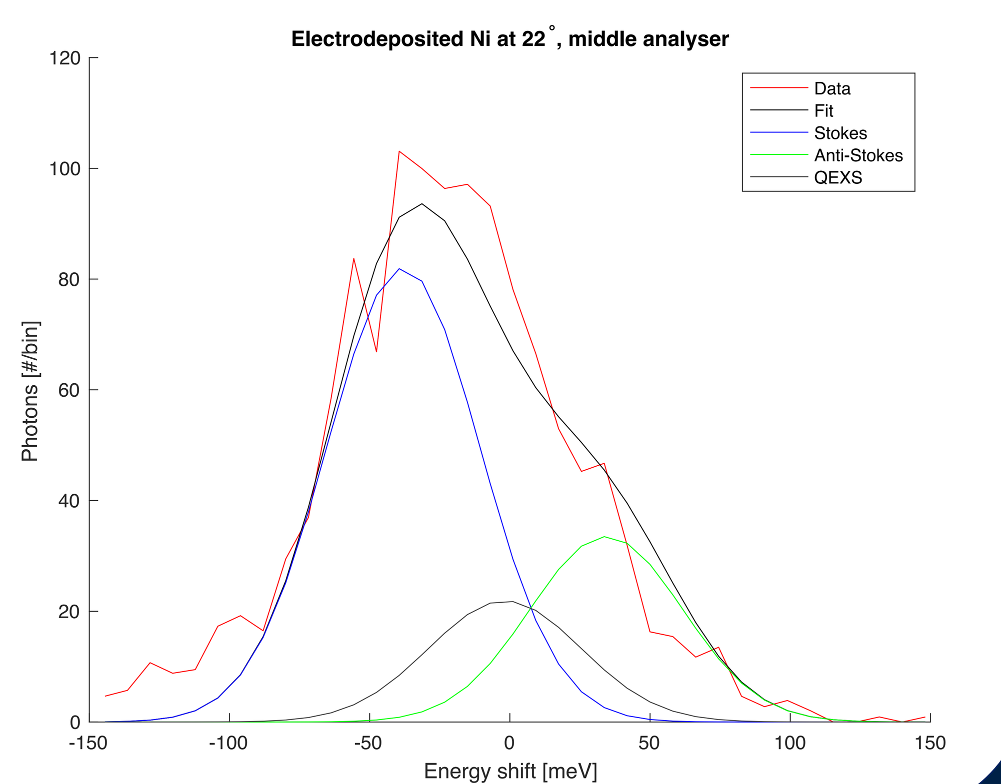
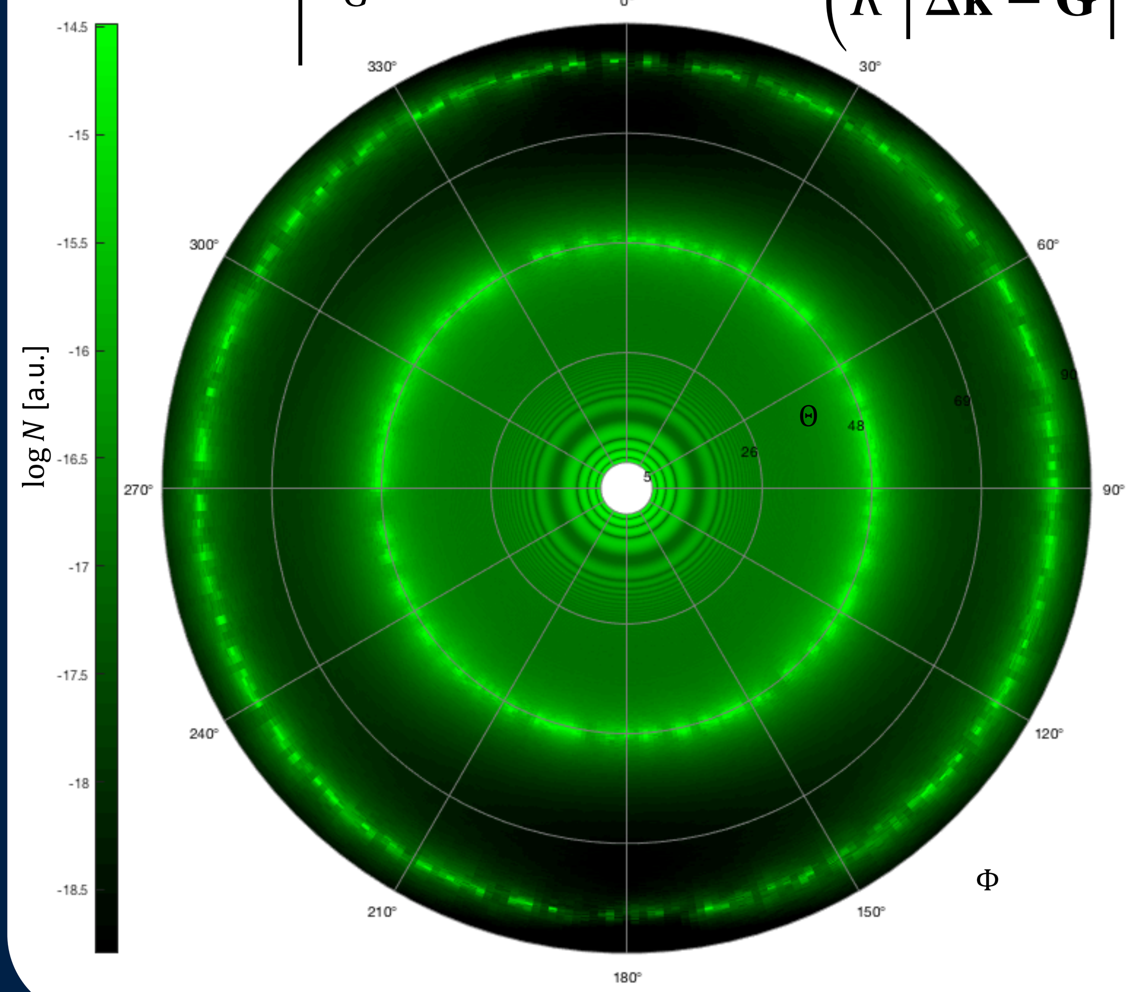


Figure 3: Exemplary result of EuXFEL p2191 experiment in Hamburg, Germany with Quasi-Elastic X-Ray Scattering (QEXS) in grey.

Deviation from Perfect Crystal

- In polycrystalline targets, IXS is competing with noise and quasi-elastic scattering (QEXS) arising from deviations from a perfect crystal, for example the shape function of each crystallite.

$$N \propto \left| \sum_G \frac{\sin\left(R\left|\Delta\mathbf{k}-\mathbf{G}\right|\right) - R\left|\Delta\mathbf{k}-\mathbf{G}\right| \cos\left(R\left|\Delta\mathbf{k}-\mathbf{G}\right|\right)}{\left(R\left|\Delta\mathbf{k}-\mathbf{G}\right|\right)^3} \right|^2$$



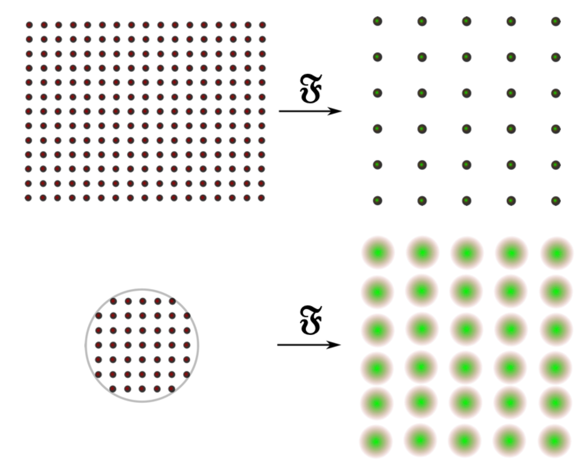


Figure 4: Quasi-elastic scattering from 1000 randomly orientated spherical diamond crystallites with R=500 nm and λ=1.66 Å.

Molecular Dynamics with LAMMPS

- In shocked samples, stacking faults can affect signal

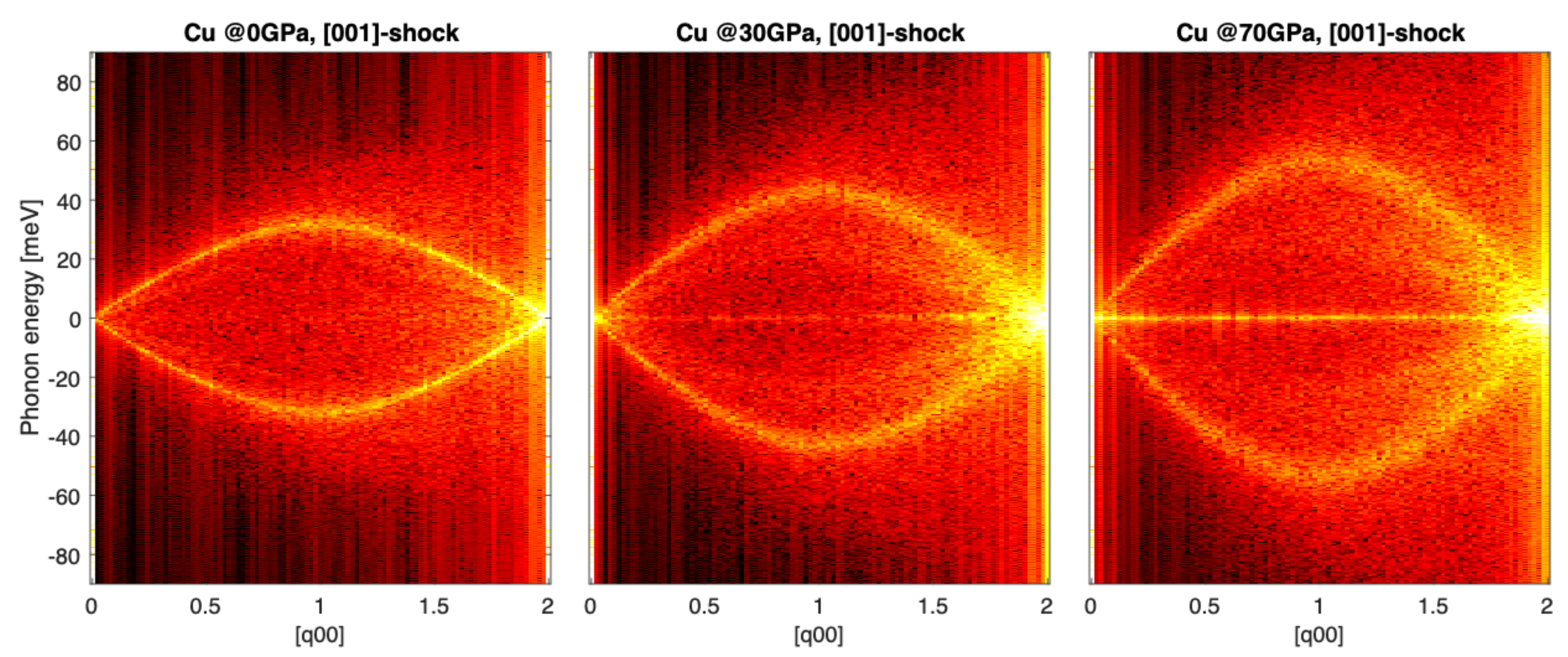


Figure 5: Stokes and Anti-Stokes modes in Cu obtained from LAMMPS. Shocked to 0, 30 and 70 GPa, the onset of QEXS at 0 meV is linked to stacking faults.

Figure 6: Box of corresponding shocked Cu atoms. Stacking faults highlighted in red by the centrosymmetry parameter.

- Design dynamic temperature measurement experiments with MD
- Data analysis of EuXFEL p2191 in progress
- XFELs at full capacity now powerful enough to obtain T
- Combine with drive lasers

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