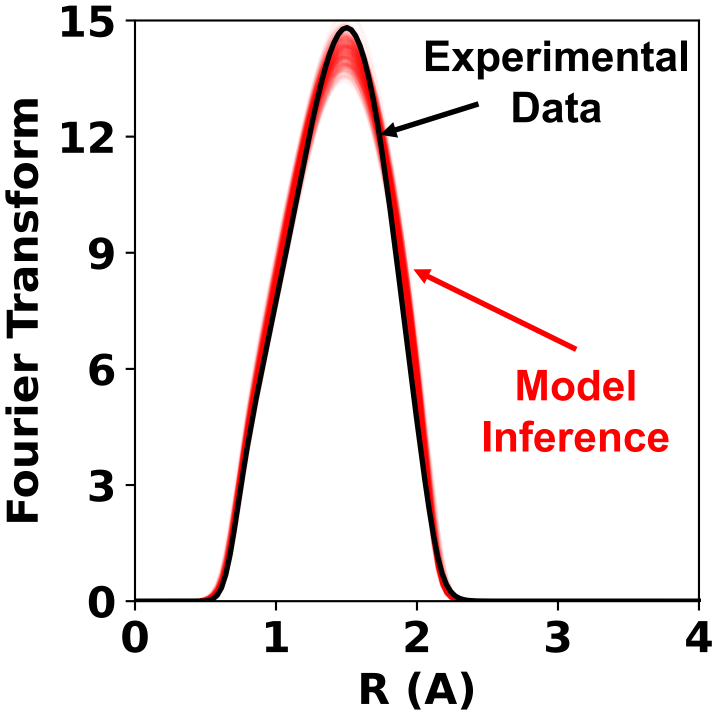
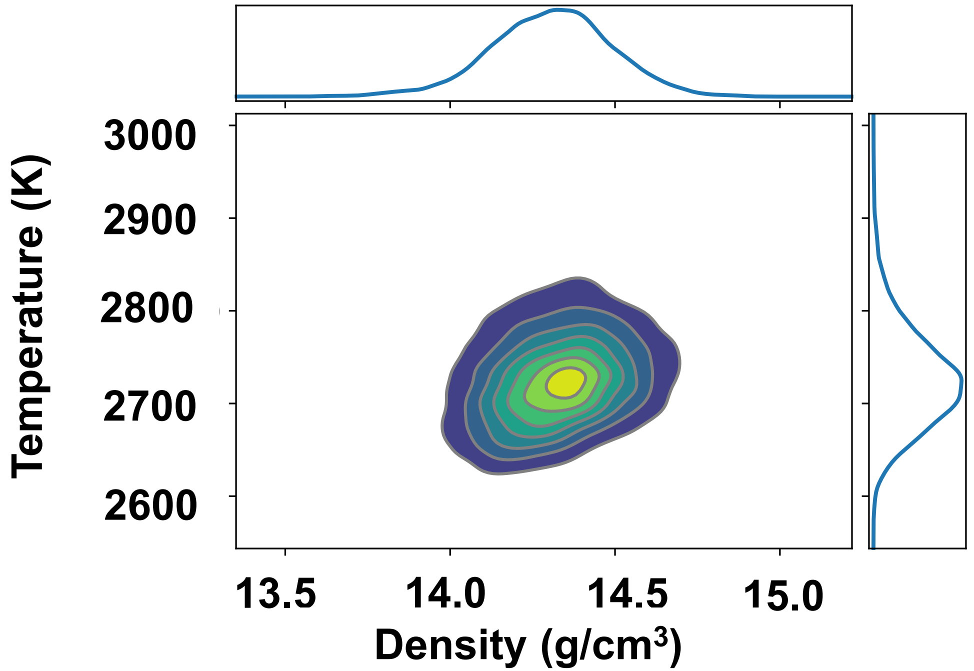
Progress towards Extended X-Ray Absorption Fine Structure (EXAFS) temperature measurements at High-Energy-Density Condition

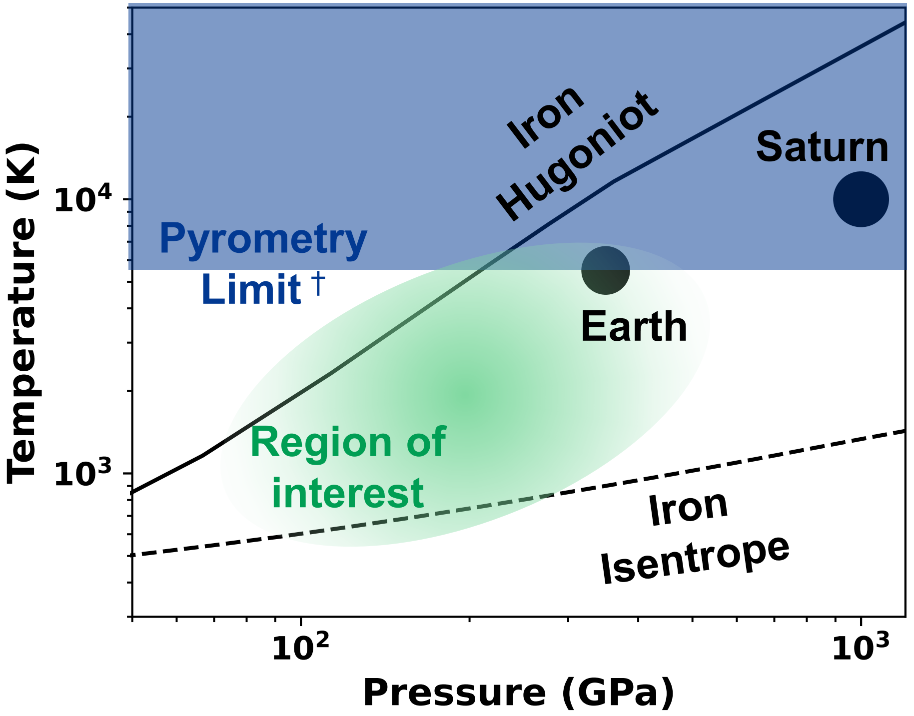




D.A. Chin
University of Rochester
Laboratory for Laser Energetics

American Physics Society Division of Plasma Physics
Spokane, Washington
October 17th 2022

Temperature is historically difficult to measure below 5000 K and above 100 GPa



EXAFS measures temperature in our region of interest

Fe Hugoniot and Isentrope from Sesame # 02140
† M.C. Gregor *et al.*, Rev. Sci. Inst. 87, 114903 (2016).

R.F. Smith *et al.*, Nature Astronomy 2, 452 (2018).
R. Kraus *et al.*, Science 375, 202 (2022).

Techniques are under development to measure the temperature of iron compounds compressed to hundreds of GPa and thousands of Kelvin

- **Extended x-ray absorption fine structure (EXAFS) spectroscopy is a technique capable of characterizing the ion vibrations *in situ***
- **Iron compounds were ramp compressed to core Earth and super-Earth conditions (up to 400 GPa and 6000 K)**
- **Analytic radial distribution models and a Bayesian inference routine are under development to extract temperature from the EXAFS modulations**

Collaborators

P. M. Nilson, D.T. Bishel, R. Paul, E.A. Smith, M. Signor, X. Gong, M.K. Ginnane, D.N. Polsin, S.X. Hu, J. R. Rygg, and G. W. Collins
University of Rochester
Laboratory for Laser Energetics

D. Trail
University of Rochester
Department of Earth and Environmental Sciences

A. Amouretti and M. Harmand
Sorbonne Université

R. Torchio
European Synchrotron Radiation Facility

J.J. Ruby, F. Coppari, A. Coleman and Y. Ping
Lawrence Livermore National Laboratory

STEWARDSHIP SCIENCE GRADUATE FELLOWSHIP

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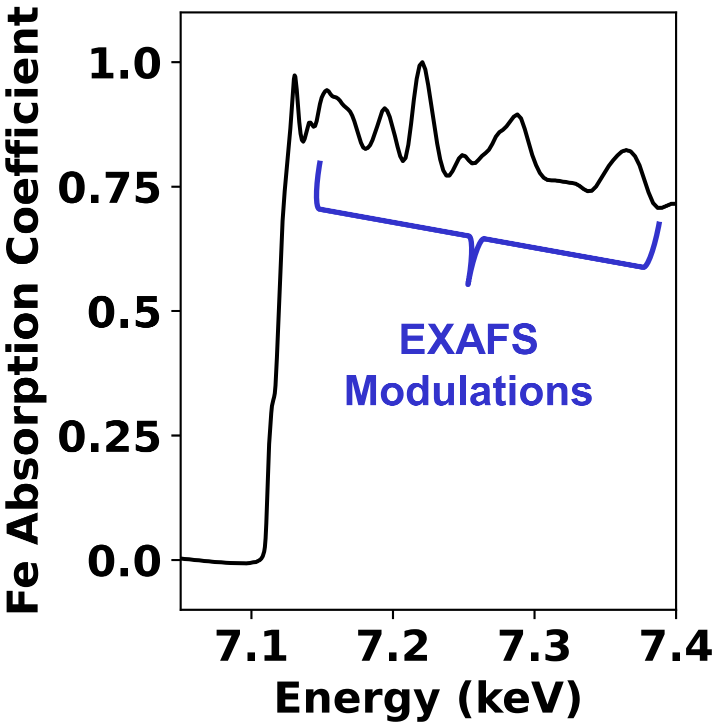
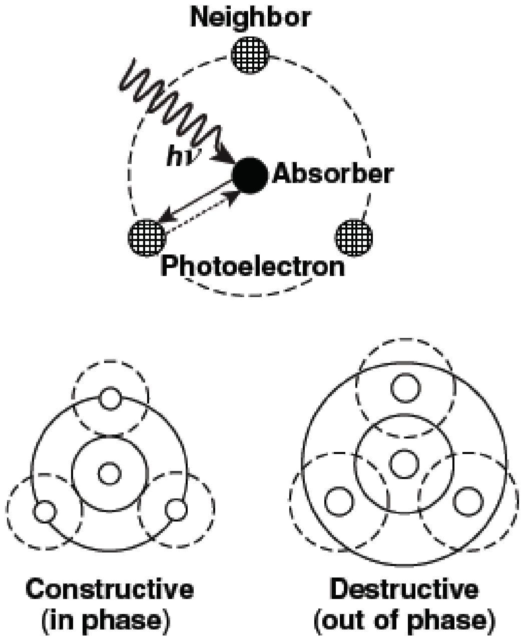
The EXAFS modulations are produced by scattering of the photoelectron off neighboring atoms



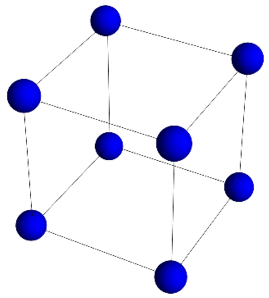


The outgoing and scattered photoelectron wavefunctions interfere, impacting the absorption coefficient

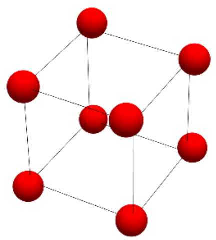
EXAFS: Extended x-ray absorption fine structure

XAFS Spectral Library, Spectrum: Fe data, 13 June 2018, http://cars.uchicago.edu/xaslib/spectrum/611.
D. C. Koningsberger *et al.*, Top. Catal. 10, 143 (2000).

Increasing density increases the period of the modulations while increasing the temperature damps out the modulations



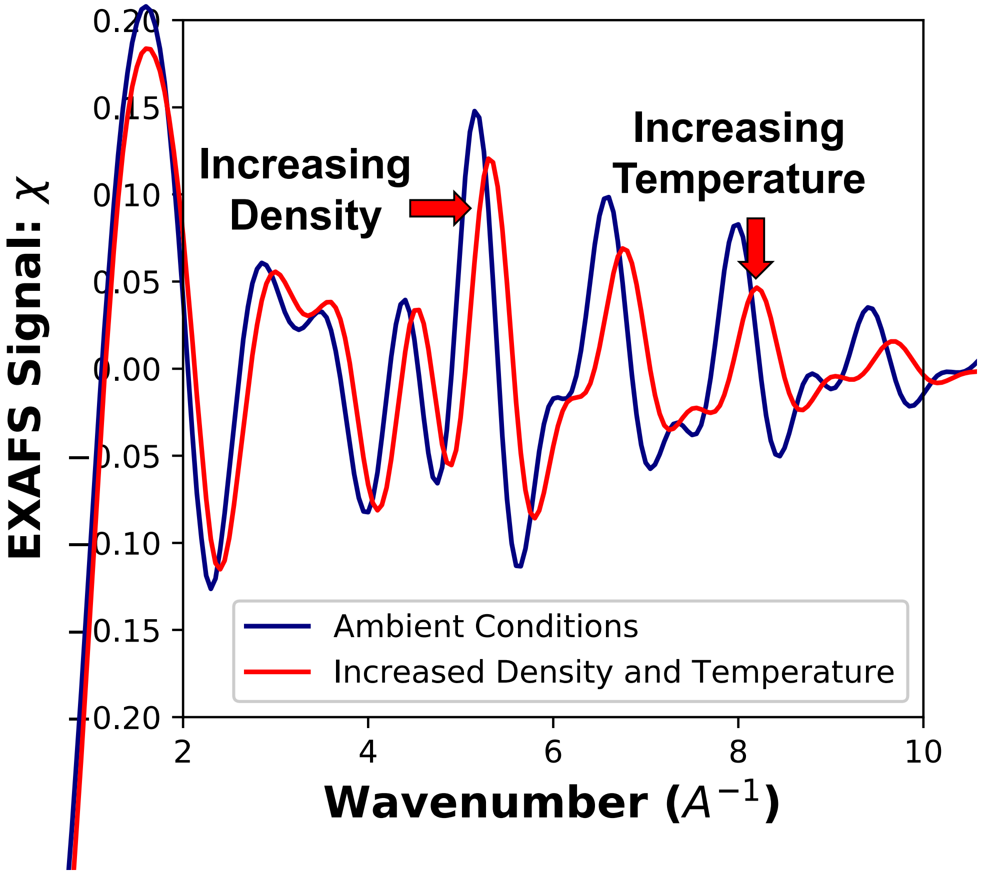
Ambient Conditions



Increased Density and Temperature

The EXAFS modulations are isolated to obtain χ

$$\chi(k) = \frac{\mu(E) - \mu_0(E)}{\mu_0(E)}$$



E: Energy of photon
k: Electron wavenumber
$\mu(E)$: absorption coefficient
$\mu_0(E)$: absorption coefficient without neighboring atoms

J.J. Rehr *et al.*, Phys. Chem. Chem. Phys., 12 5503-5513 (2010).

The EXAFS modulations depend upon the radial distribution of atoms surrounding the absorbing atom

The EXAFS equation for each scattering path is an integral over the real distribution ($p(r)$):

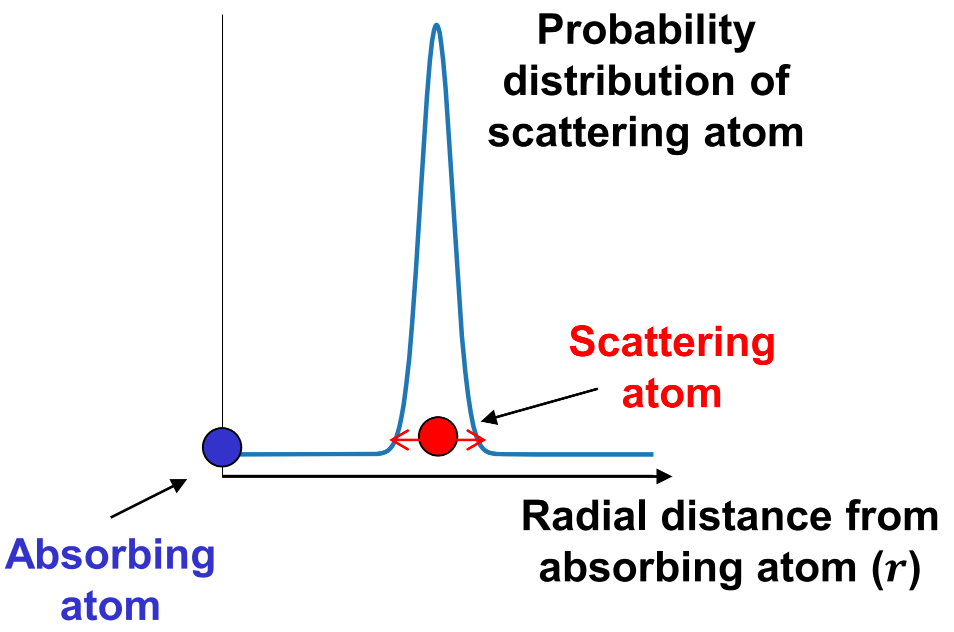
$$\chi(k) = \int A(k,r)\,p(r)\,\sin\big(2kr + \delta(k)\big)\,dr$$

Amplitude → $A(k,r)$

Real Distribution → $p(r)$

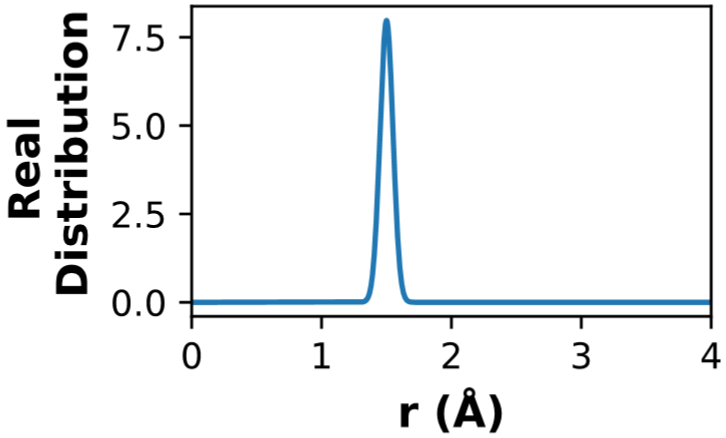
Phase shift → $\delta(k)$

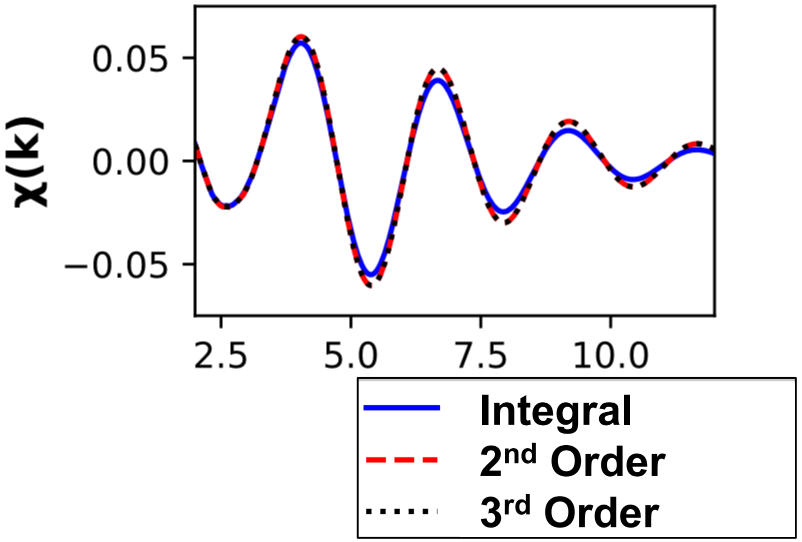
Real Distribution: $p(r)$



G. Bunker *Introduction to XAFS* University Press, Cambridge, (2010).

The EXAFS equation is often modeled with a moment expansion





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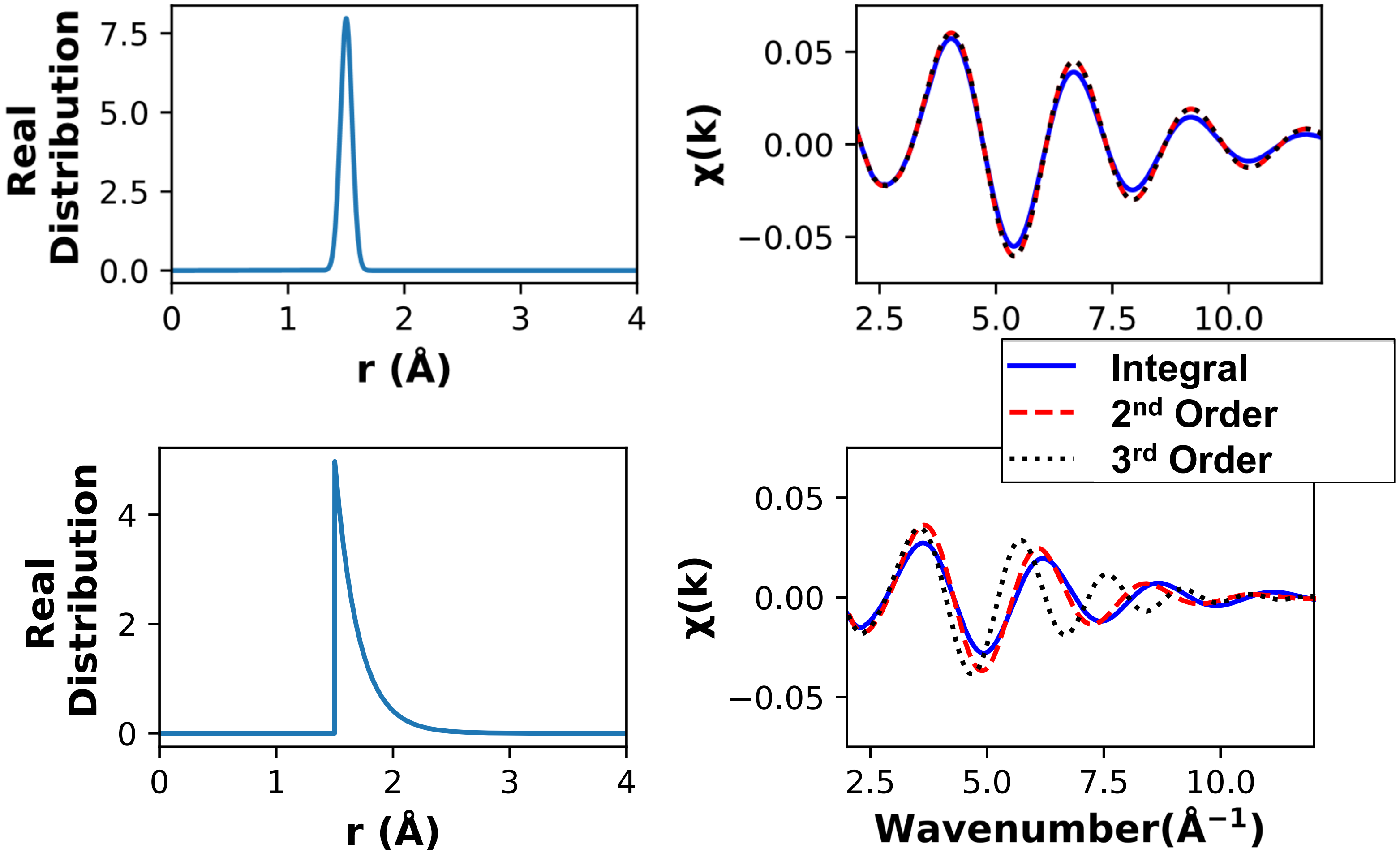
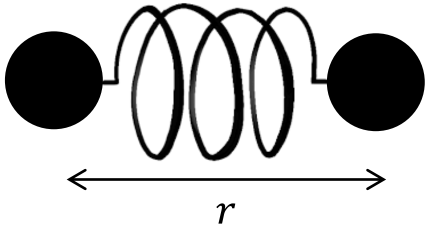


This moment expansion does not always converge

As an alternative to the moment expansion, the real distribution can be modeled directly

- **Reduced system of 2 ions**



- **The Hamiltonian of the system:**

$$H = \frac{1}{2}\mu\,(\dot{r}^2 + \omega_E^2 r^2)$$

- **The energy in the system is given by kT**

μ: Reduced mass
ω_E: Einstein frequency

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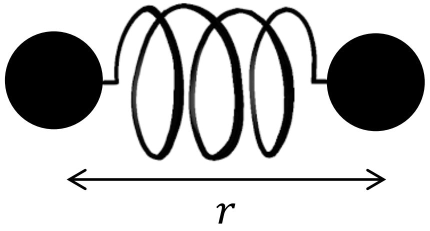

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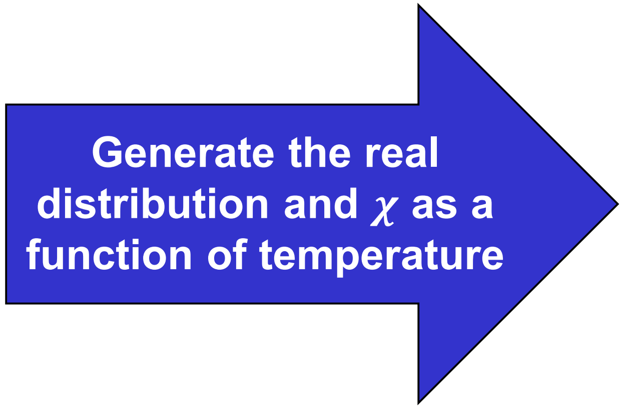

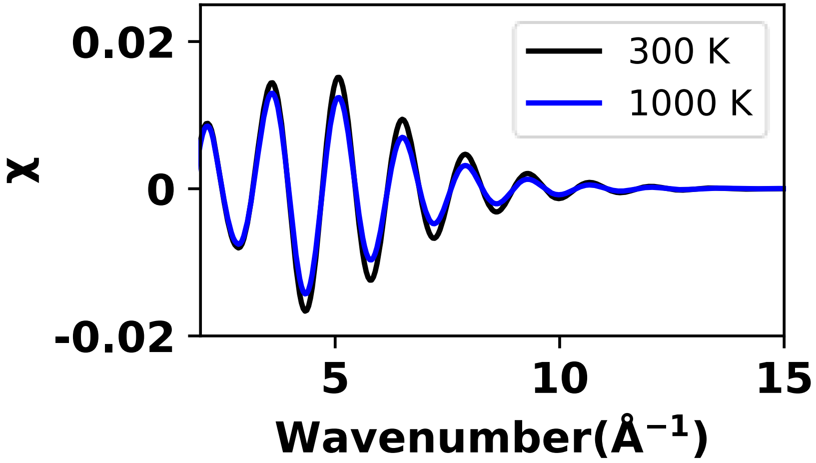

μ: Reduced mass
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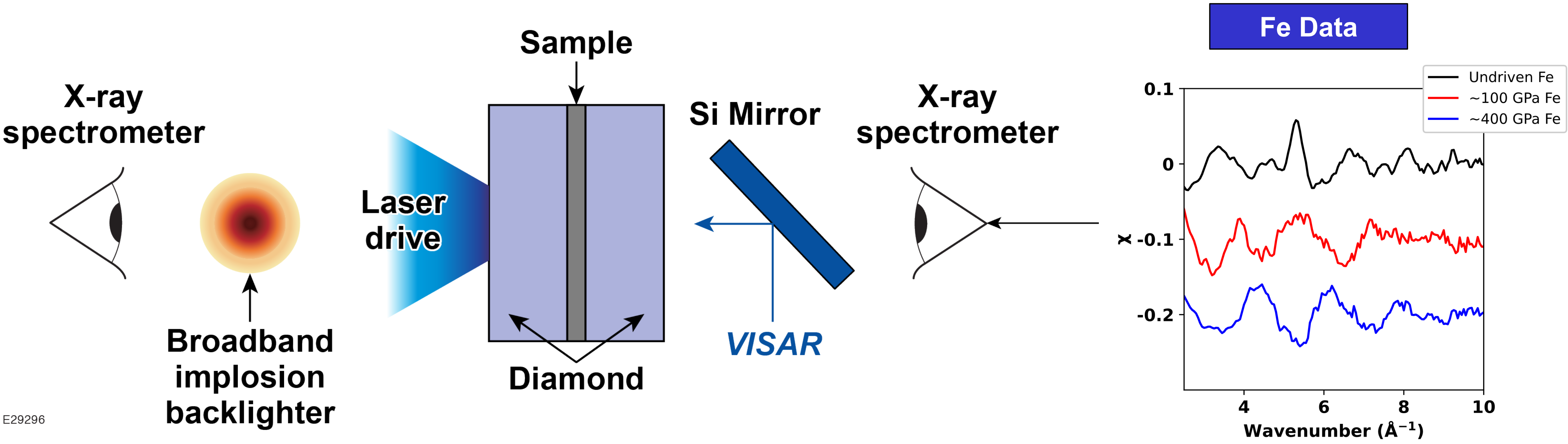
J.J. Rehr *et al.*, Phys. Chem. Chem. Phys., 12 5503-5513 (2010).

Samples were compressed over a few nanoseconds and probed with x-rays over a few hundred picoseconds

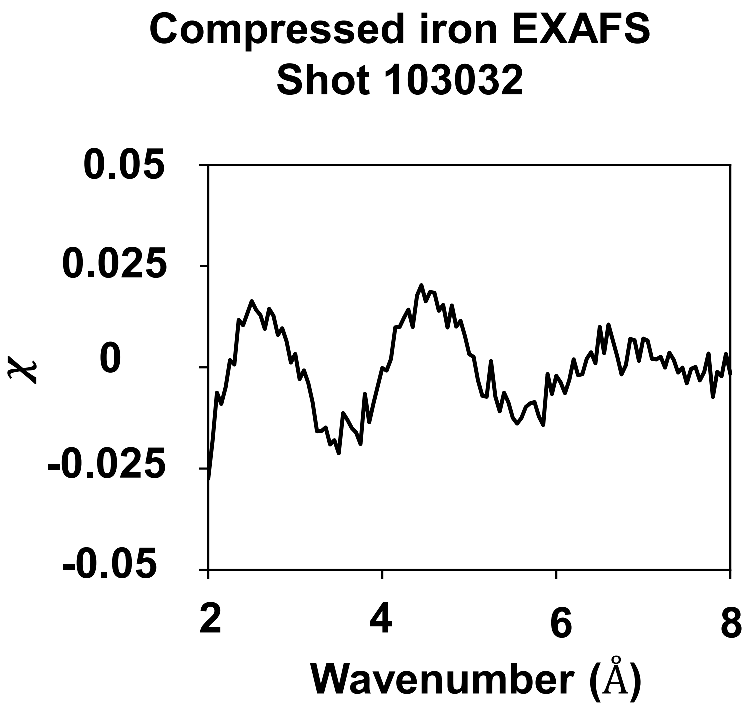


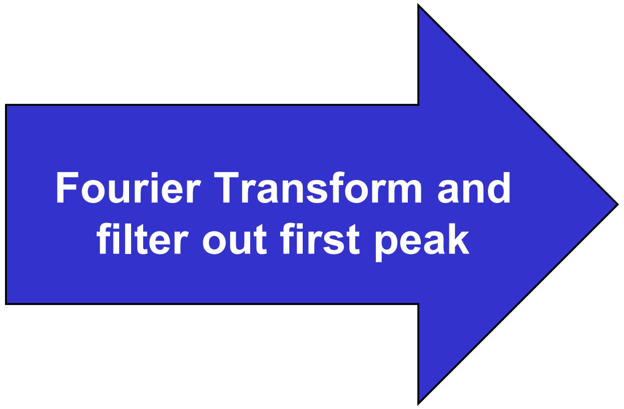
E29296

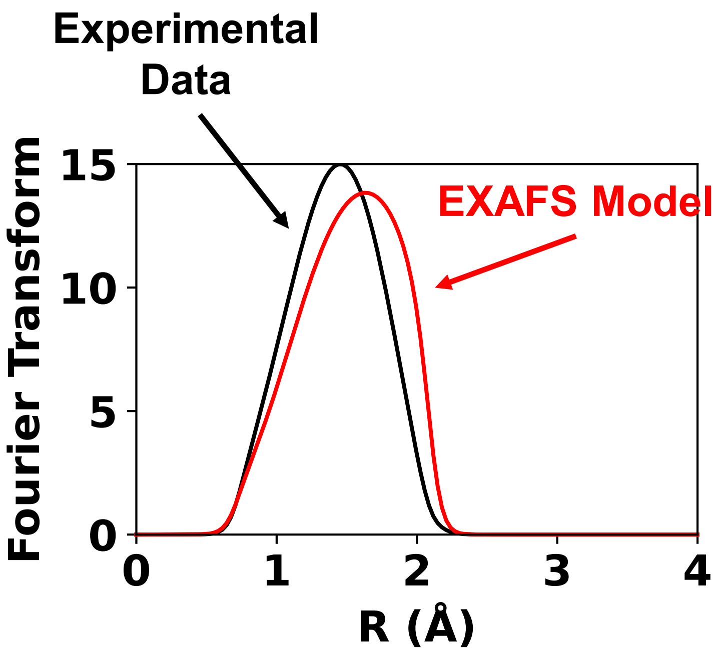
VISAR – Velocity interferometry system for any reflector

Y. Ping *et al.*, Phys. Rev. Lett. 111, 065501 (2013).
D.A. Chin *et al.*, Phys. Plasmas 29, 052702 (2022).

The measured data was transformed into Fourier Space and compared with different EXAFS models

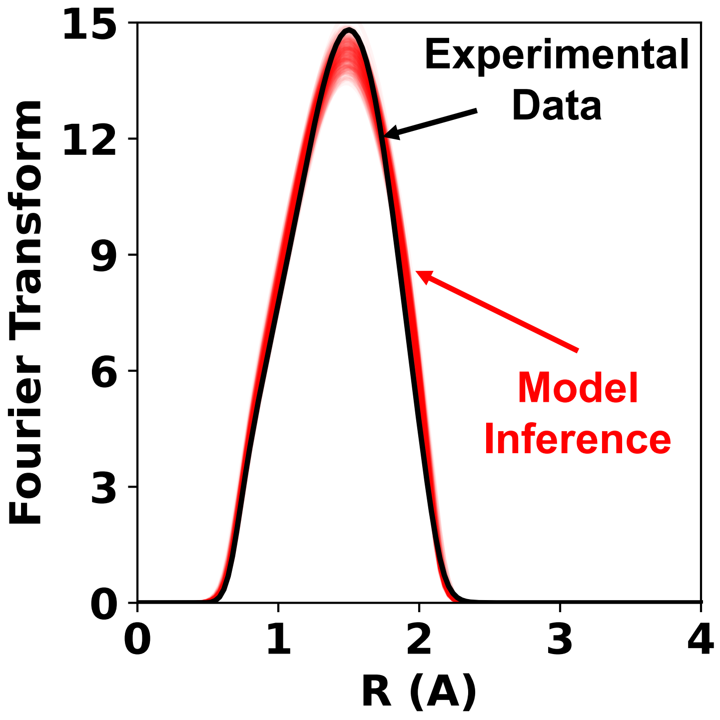


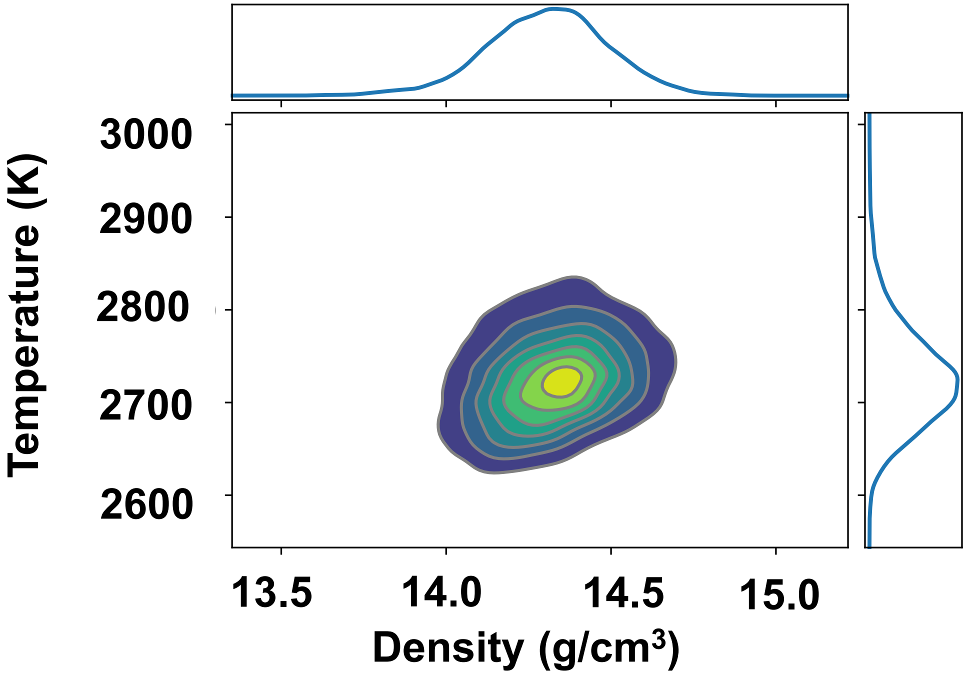




A Bayesian inference routine was used to extract density and temperature for each model

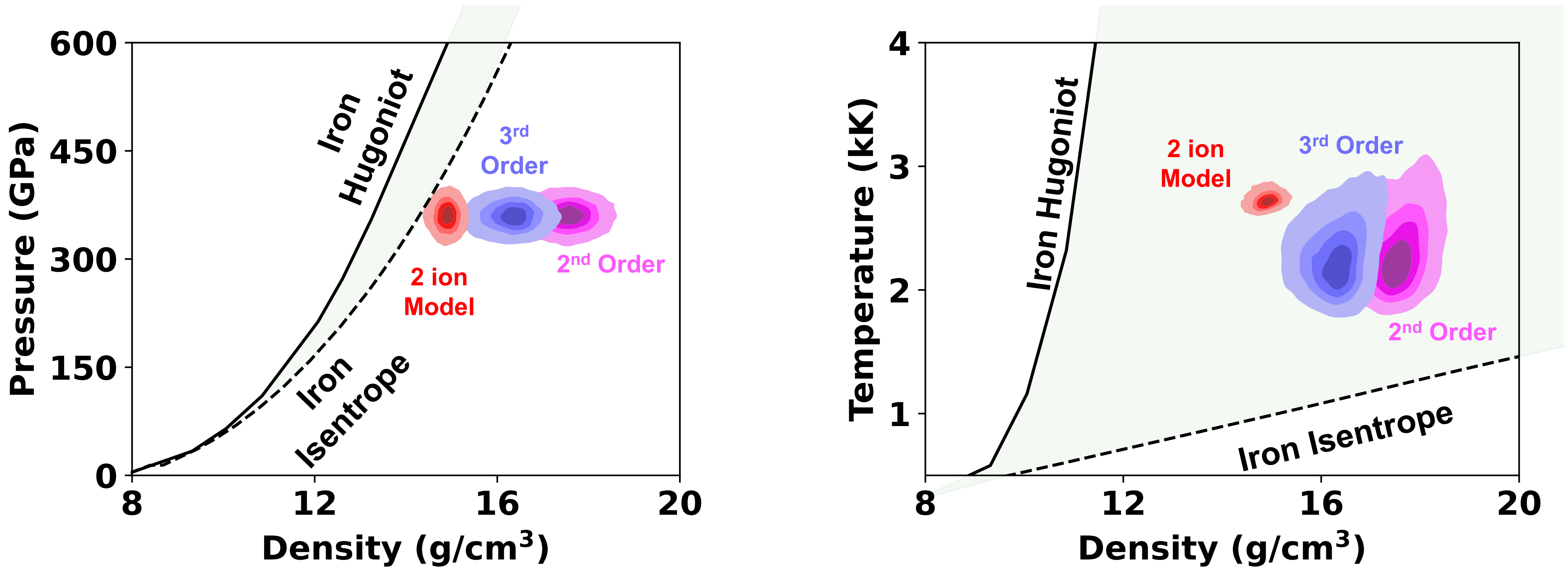
Analysis of compressed iron EXAFS data with the 2 ion model





D. Phan *et al.*, Statistics Archive https://arxiv.org/abs/1912.11554 (2019).
E. Bingham *et al.*, J. Mach. Learn. Res. 20, 973 (2019).

Three different EXAFS models were used to extract temperature and density from the same shot



Preliminary results indicate the density from the 2 ion model is more consistent with the iron equation of state

Fe Hugoniot and Isentrope from Sesame # 02140

Future work is planned to further improve the inference

1. Additional free parameters are required in the models ($\omega_E \ldots$)*

2. A more sophisticated likelihood is needed to better account for data uncertainty**

3. Uncertainty in the spectrometer energy dispersion relation must be accounted for during the inference†

These EXAFS models will be benchmarked against other published nickel EXAFS datasets at lower temperatures and densities‡

*L.S. Dubrovinsky *et al.,* Am. Min. 85, 386 (2000).
** P. Marjoram OA Genetics 01, 1(1):3 (2013).
†D.A. Chin *et al.,* EXAFS flat crystal (EFX) high-resolution x-ray spectrometer, Rev. Sci. Instrum. *In review*
‡S. Boccato *et al.,* J. Geo. Res. Solid Earth 122 9921 (1017).

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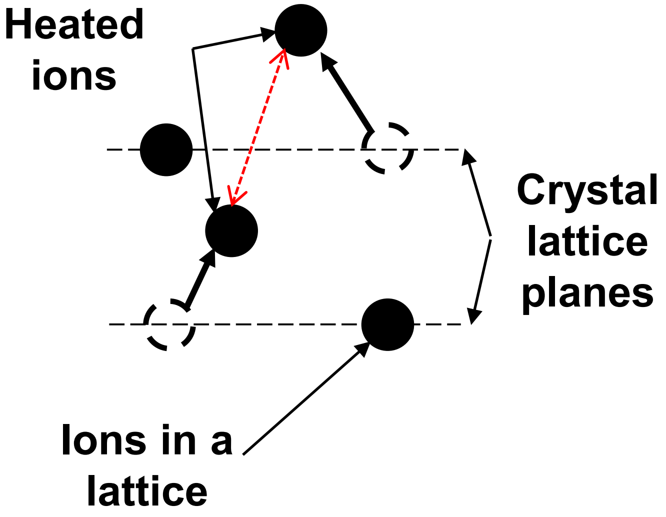
Backup

There are a variety of techniques under development to measure temperature in this low temperature HED regime

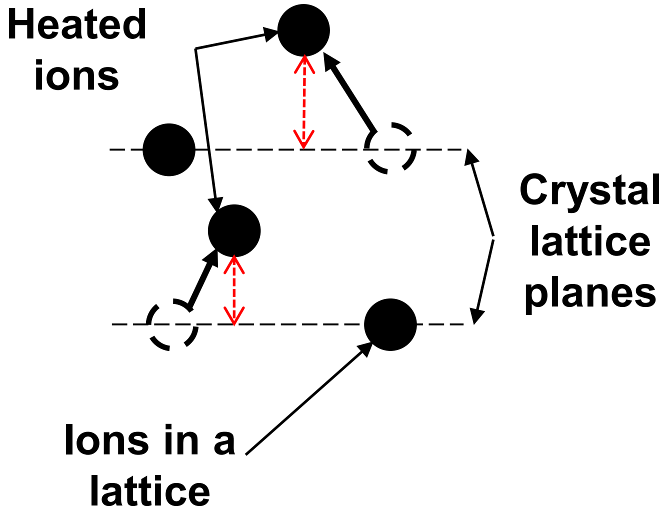
Extended X-ray Absorption Fine Structure (EXAFS)[1,2]

Measures the variation in the distance between ions

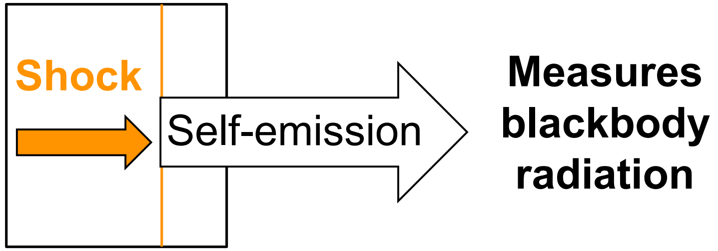


X-Ray Diffraction (XRD)[3]

Measures the variation in the distance between the ion and the lattice plane



Streaked Optical Pyrometry (SOP)[4]



X-Ray Absorption Near Edge Spectroscopy (XANES)[5]

Measures electron density of states

- **Transition rate $R \sim [1 - f(E)]$**
- **f is the Fermi-Dirac distribution**

$$f(E) = \left[1 + \exp\left(\frac{E - \mu(T_e)}{k_B T_e}\right)\right]^{-1}$$

UNIVERSITY of ROCHESTER

[1]E. Sevillano *et al.*, Phys. Rev. B 20, 4908 (1979).
[2]Y. Ping et al., Phys. Rev. Lett. 111, 065501 (2013).
[3]B.E. Warren “X-Ray Diffraction” (Dover Publications, 1990)
[4]M.C. Gregor *et al.*, Rev. Sci. Instrum. 87, 114903 (2016)
[5]F. Dorchies *et al.*, Phys. Rep. 657, 1-26(2016)