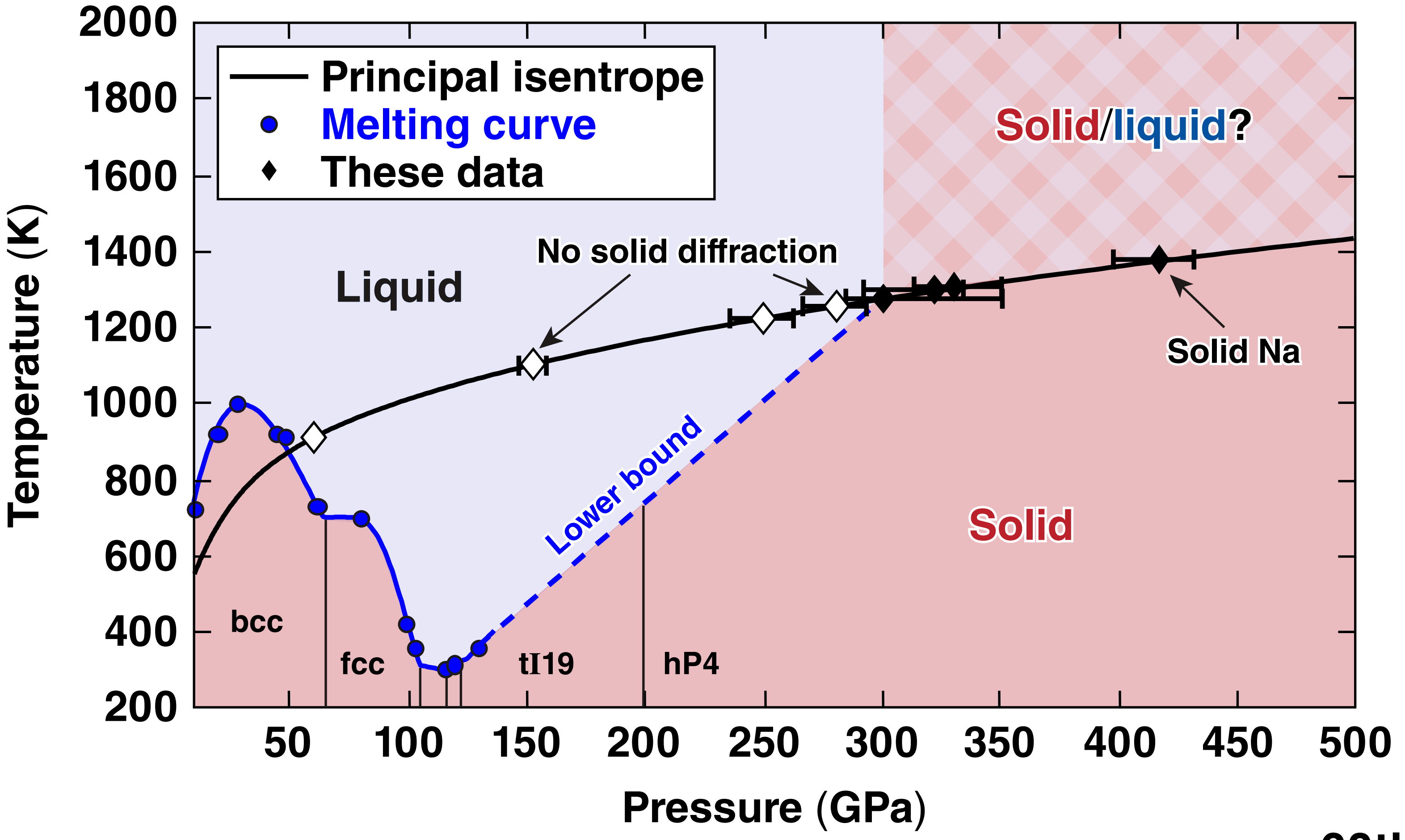
X-Ray Diffraction of Ramp-Compressed Sodium



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Summary

Melting and resolidification of Na to a solid phase is observed for pressures greater than 300 GPa

- Alkali metals, including Na, serve as a prototypical system to guide theory for ultradense matter beyond the Thomas–Fermi model
- High-power lasers ramp compressed Na and nanosecond *in-situ* x-ray diffraction (XRD) was used to measure the crystal structure at pressures up to 418 GPa
- Solid Na was observed at 301 to 418 GPa; therefore, the melting temperature increases dramatically above 140 GPa
- Simultaneous XRD and optical reflectivity measurements reveal that Na remains reflective to at least 290 GPa, in striking contrast to room-temperature static-compression measurements

Collaborators

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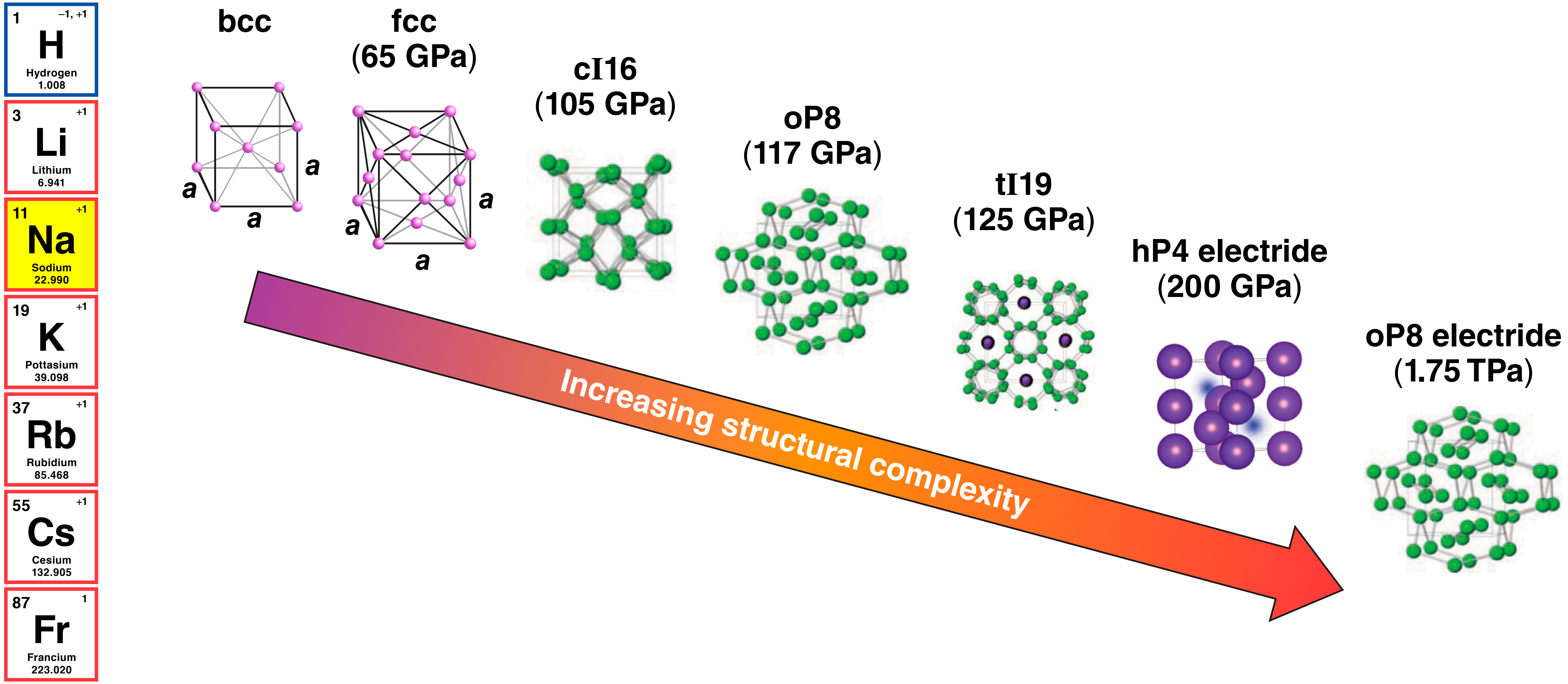
Motivation

Alkali metals, including sodium, are an ideal test bed for studying high-pressure structural phenomenon



1	−1, +1	H	Hydrogen	1.008
3	+1	Li	Lithium	6.941
11	+1	Na	Sodium	22.990
19	+1	K	Pottasium	39.098
37	+1	Rb	Rubidium	85.468
55	+1	Cs	Cesium	132.905
87	1	Fr	Francium	223.020

bcc

fcc (65 GPa)

cI16 (105 GPa)

oP8 (117 GPa)

tI19 (125 GPa)

hP4 electride (200 GPa)

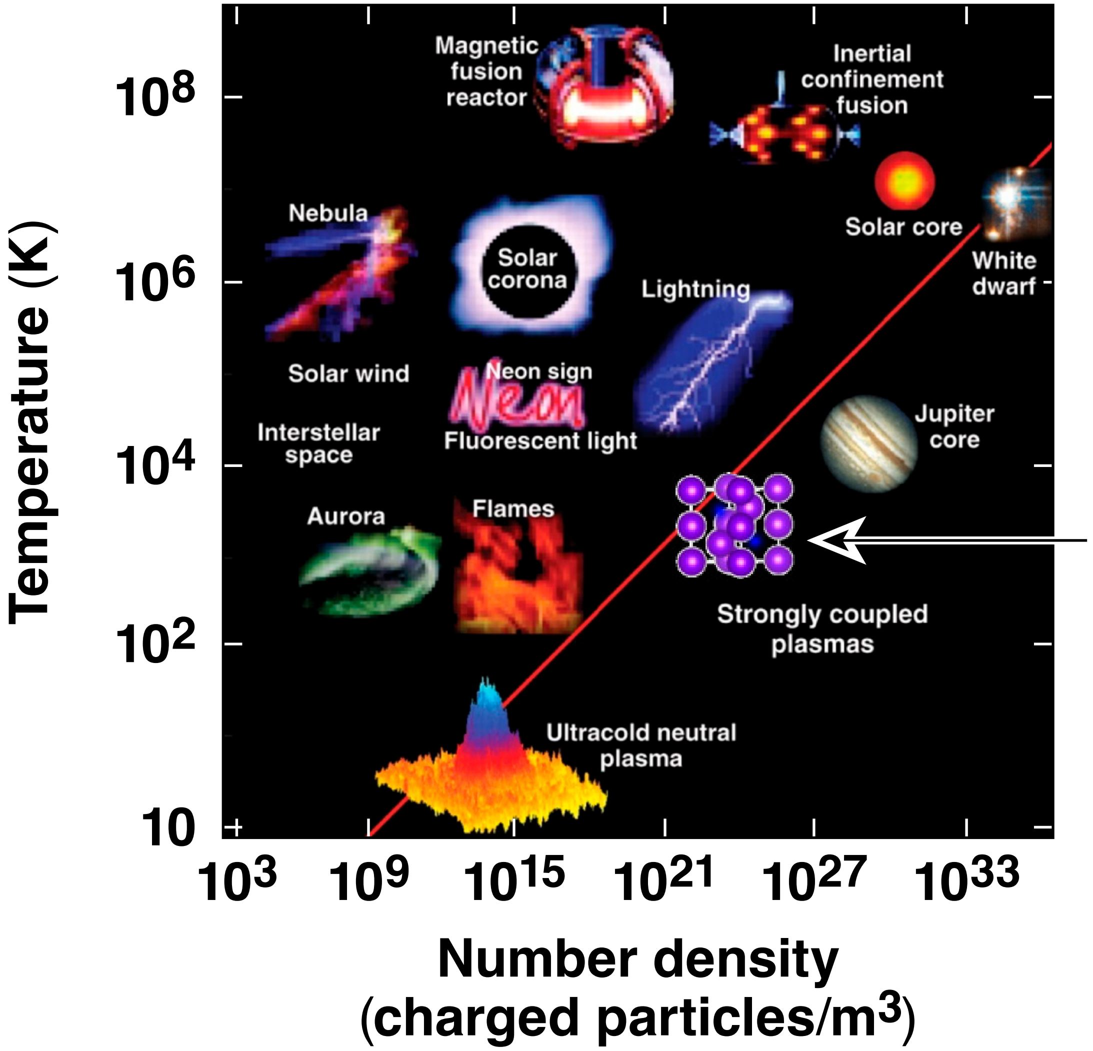
oP8 electride (1.75 TPa)

Increasing structural complexity

bcc: body centered cubic
fcc: face centered cubic

Y. Ma *et al.*, Nature 458, 182 (2009);
E. Gregoryanz *et al.*, Science 320, 1054 (2008).

High-energy-densities are achieved in these experiments through ramp-compression

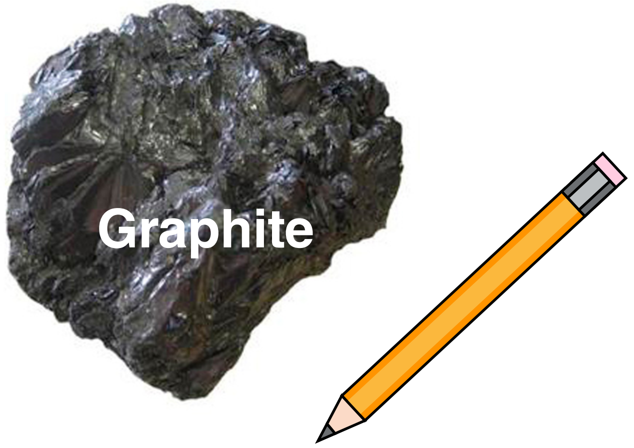


Solid density and ~a few 1000 K

T. Killian, Science 316, 5825 (2007).

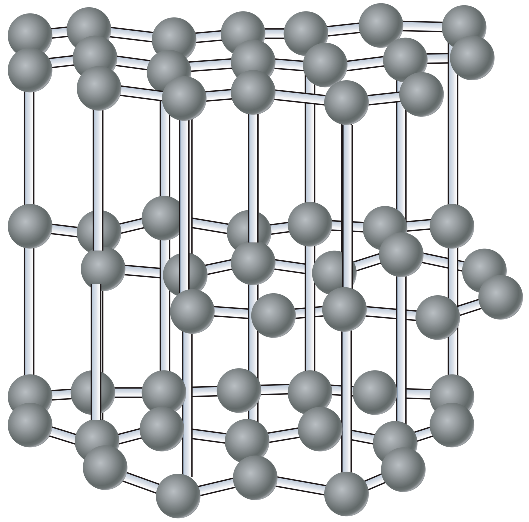
Atomic structure affects many properties of materials

(Carbon) crystals | **Physical properties** | **Atomic structure**



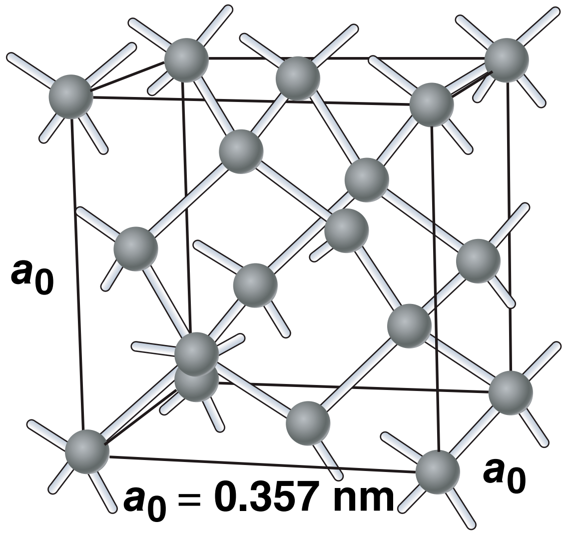
Density
2.2 g/cm^3

Yield strength
<0.2 GPa



Density
3.51 g/cm^3

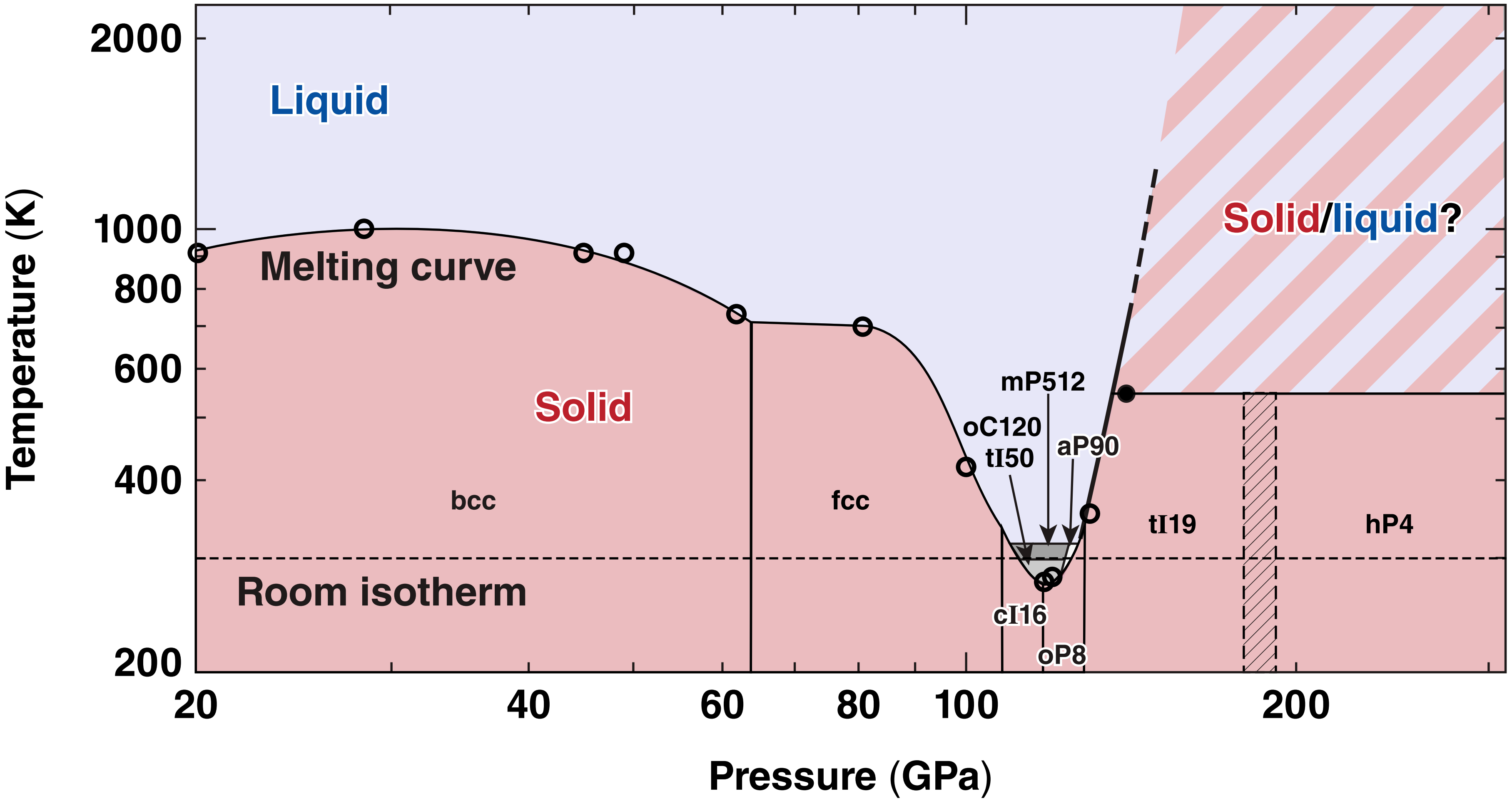
Yield strength
>110 GPa



There is a 1.6× difference in density, 10× difference in compressibility, and 500× difference in strength.

At high pressures, Na has a unique melting curve that exhibits a minimum at 120 GPa, then rises steeply

Sodium**



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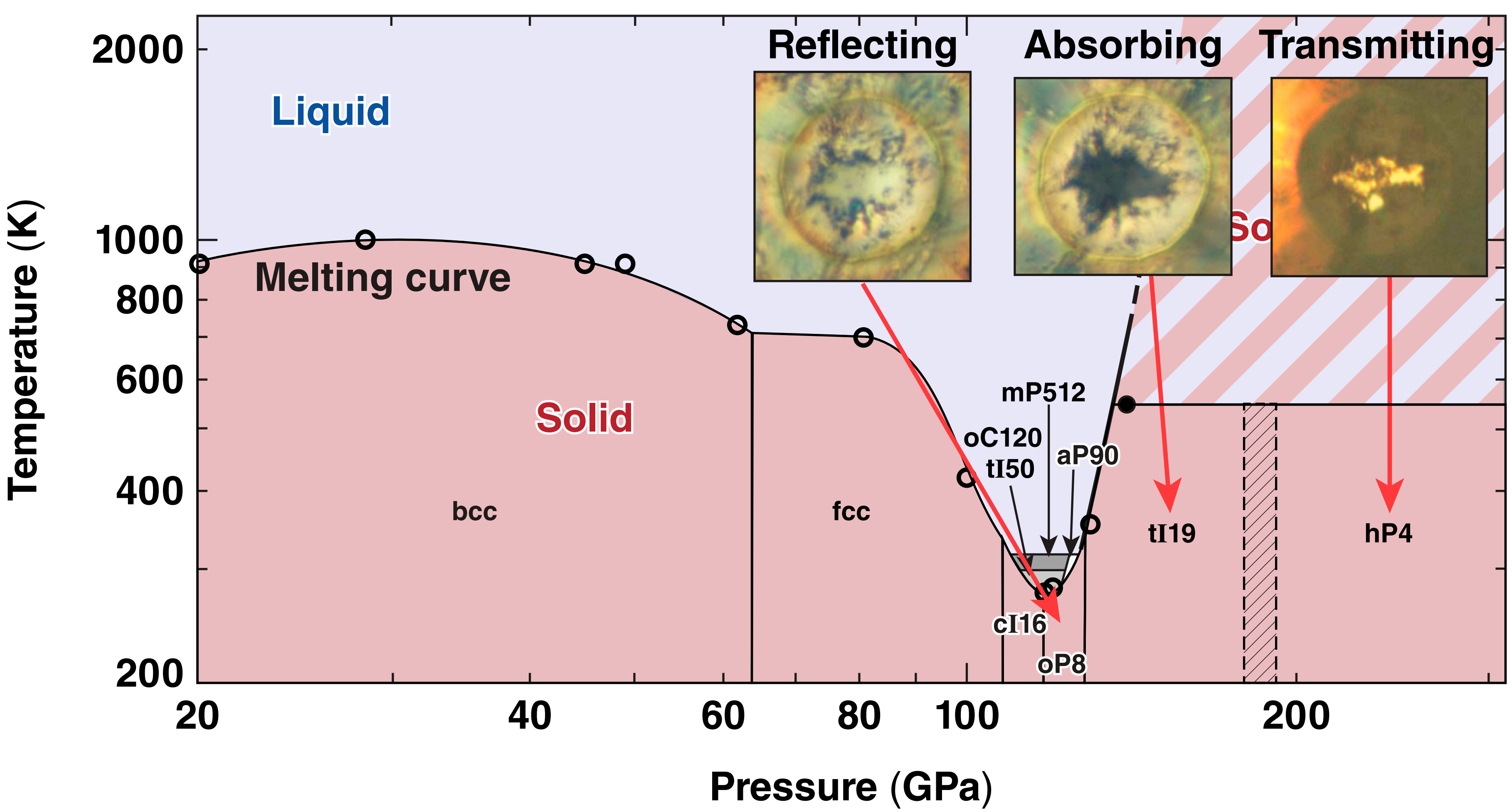
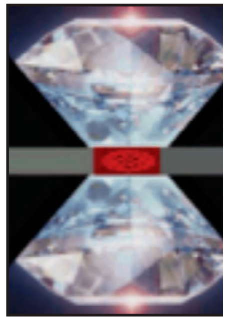
*E. Gregoryanz *et al.*, Science 320, 1054 (2008).
**M. Marqués *et al.*, Phys. Rev. B 83, 184106 (2011).

Diamond-anvil cell (DAC) experiments* show that Na transforms into an optically transparent phase at 200 GPa





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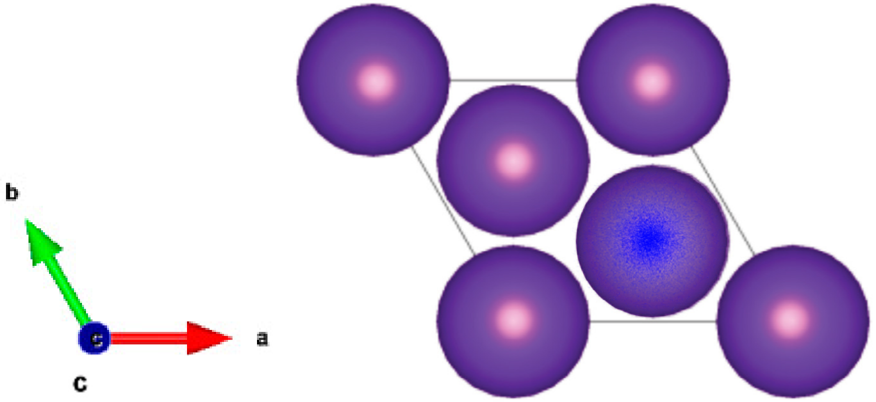
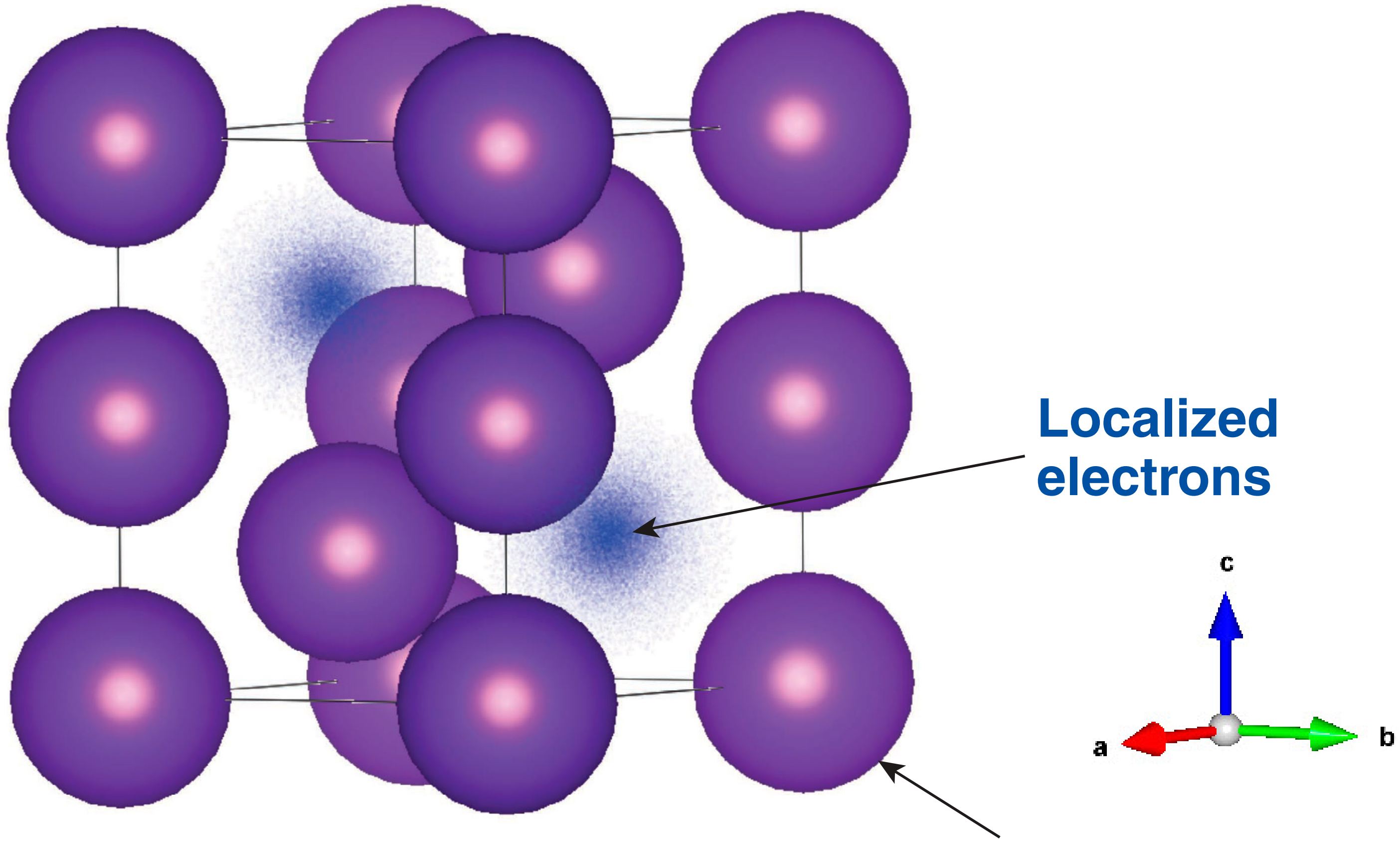
*Y. Ma *et al.*, Nature 458, 182 (2009).

The transparent phase is predicted* to be an electride hP4 structure, where conduction electrons are "trapped" in interstitial wells, producing an insulating behavior

hP4: double hexagonal close-packed (dhcp) structure, squeezed more than twice along the *c* axis





Localized electrons

Na^+ ions

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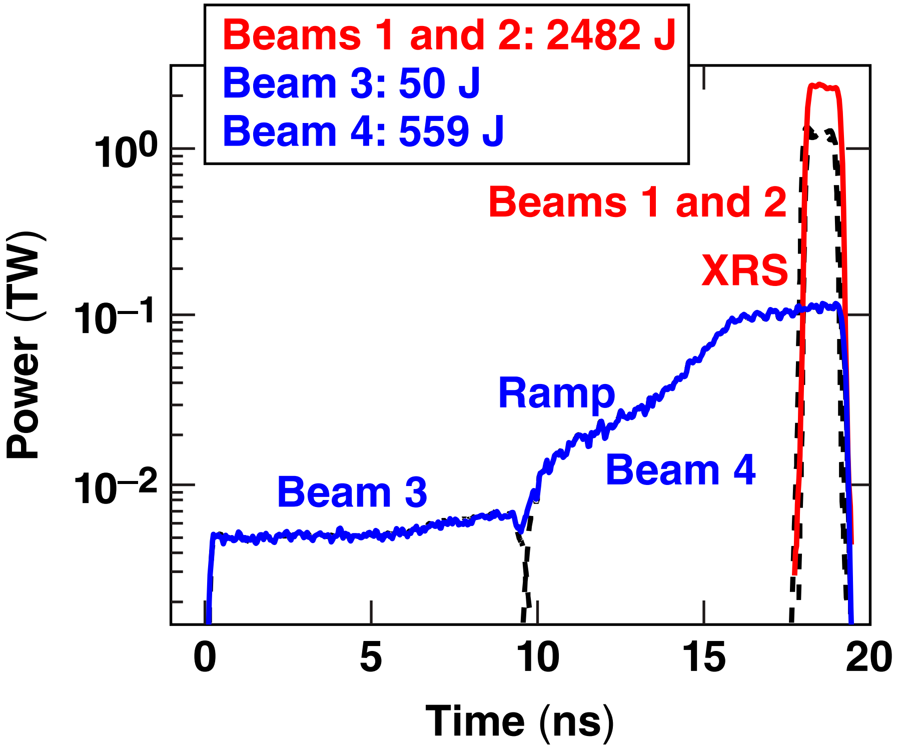
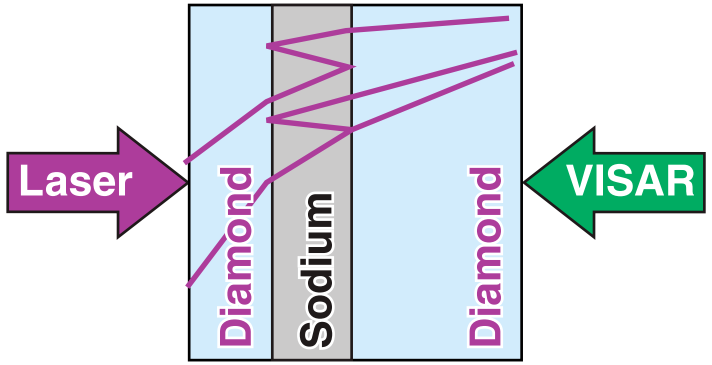
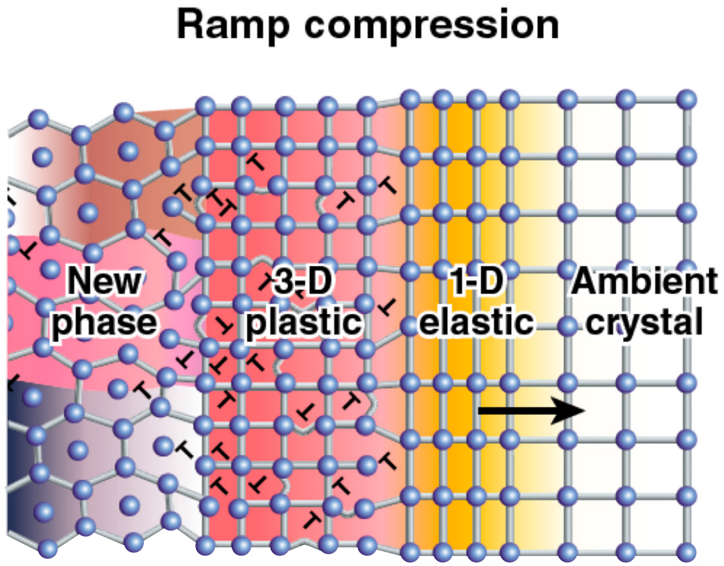
*Y. Ma *et al.*, Nature 458, 182 (2009).

Ramp compression is used to follow a nearly isentropic compression path to investigate the solid phases of Na

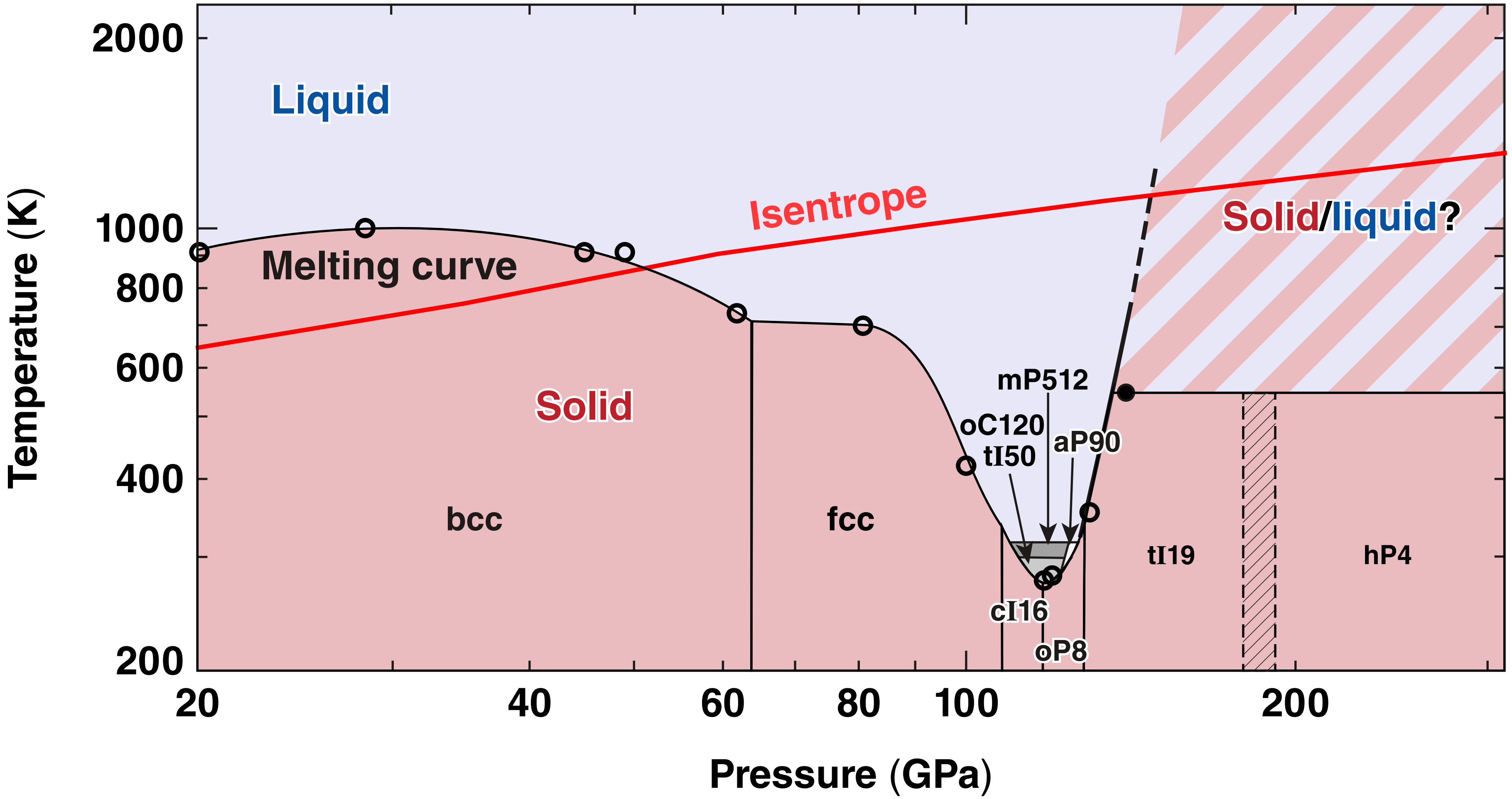






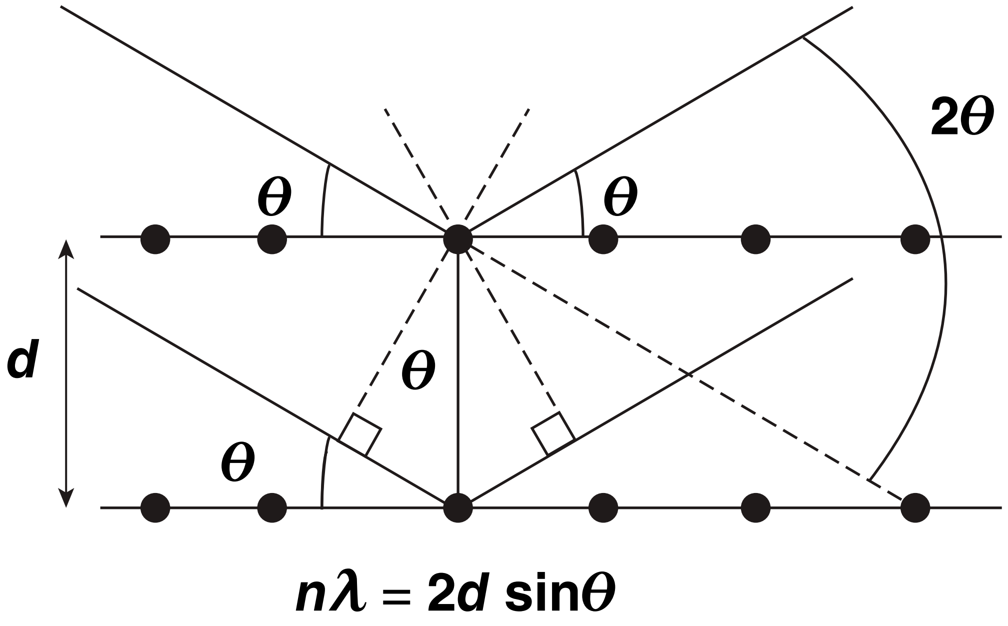
VISAR: velocity interferometer system for any reflector
XRS: x-ray source

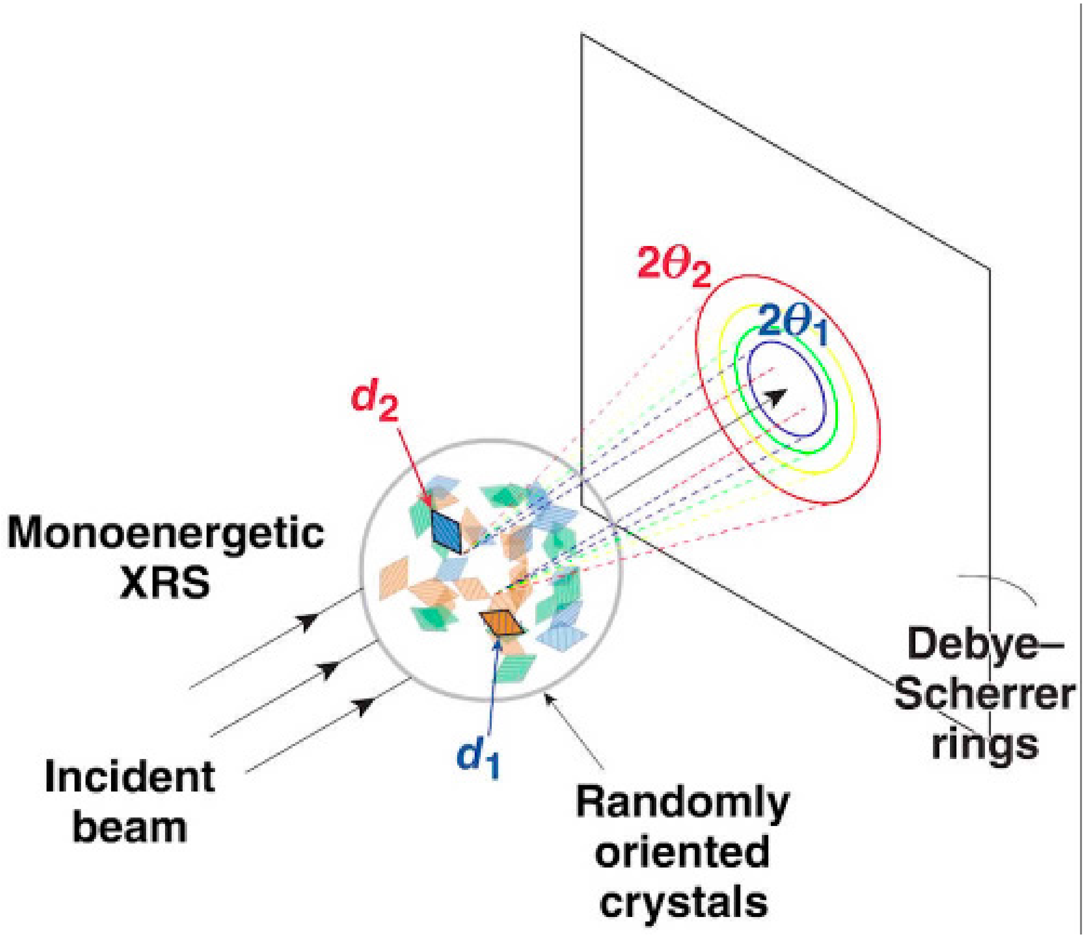
Assuming isentropic compression, Na will melt in the bcc phase and a solid phase will be observed if the melting curve continues to sharply increase



M. Marqués *et al.*, Phys. Rev. B 83, 184106 (2011).

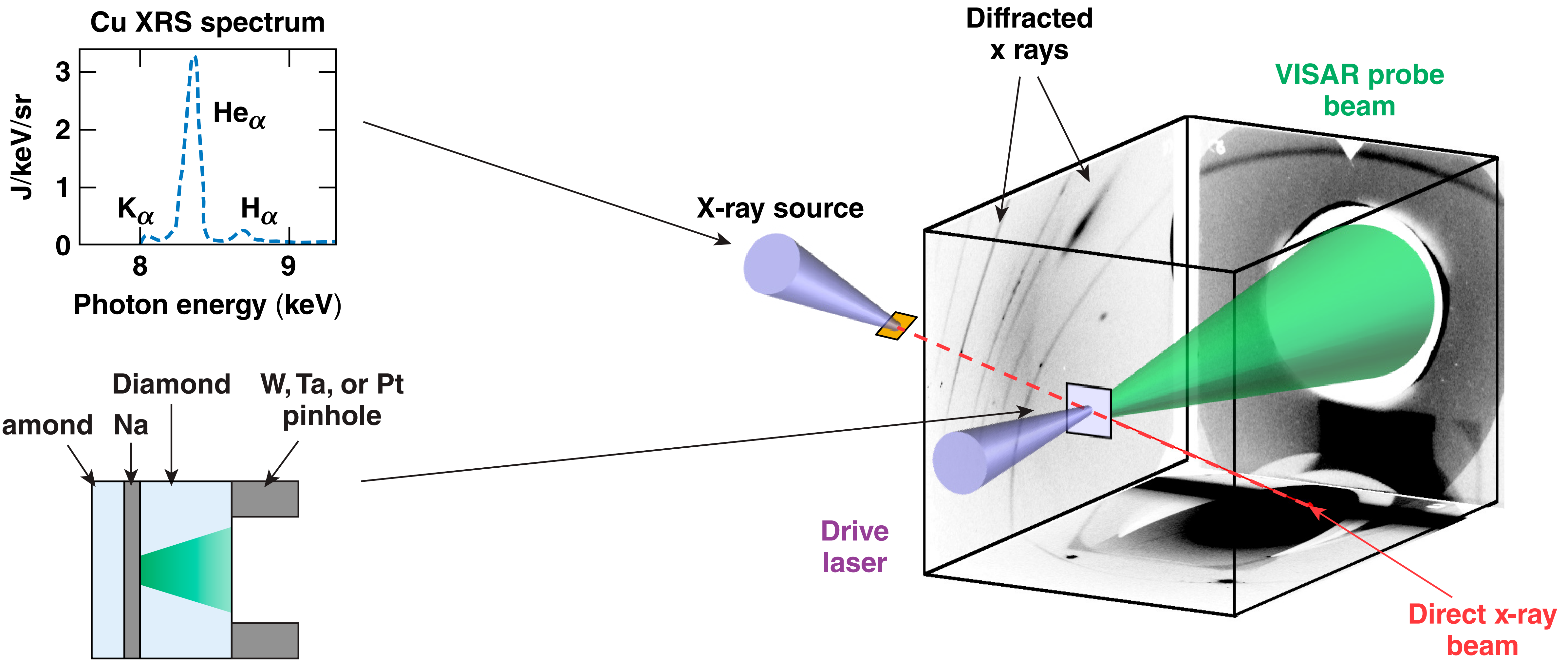
Monoenergetic x rays incident on an ideal powder sample will diffract in rings of uniform intensity at angles 2θ with respect to the x-ray beam





Compression of interatomic spacing (*d* spacing) is determined by measuring diffraction angles (2θ) and x-ray wavelength (λ).

The powder x-ray diffraction image-plate (PXRDIP*) platform is used to record diffraction patterns on OMEGA EP



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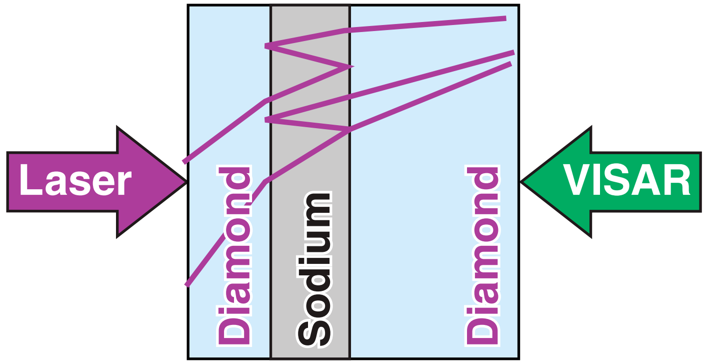
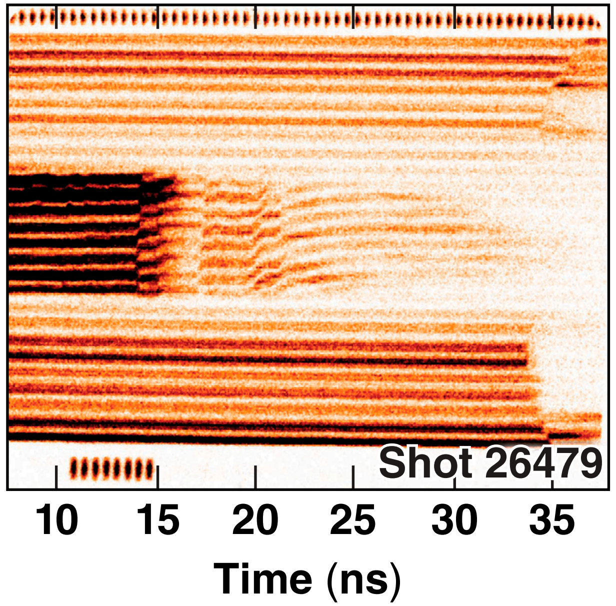
*J. R. Rygg *et al.*, Rev. Sci. Instrum. 83, 113904 (2012).

The velocity of the diamond free surface is back-propagated to determine the pressure in the Na sample





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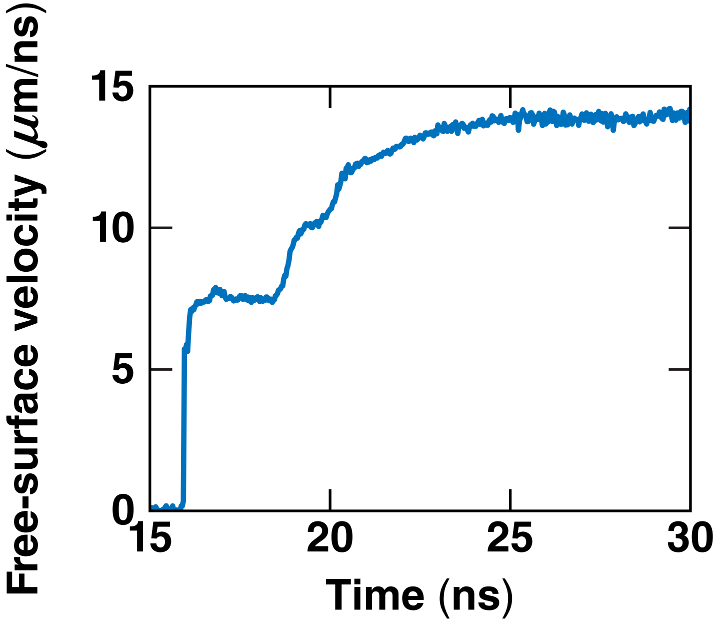

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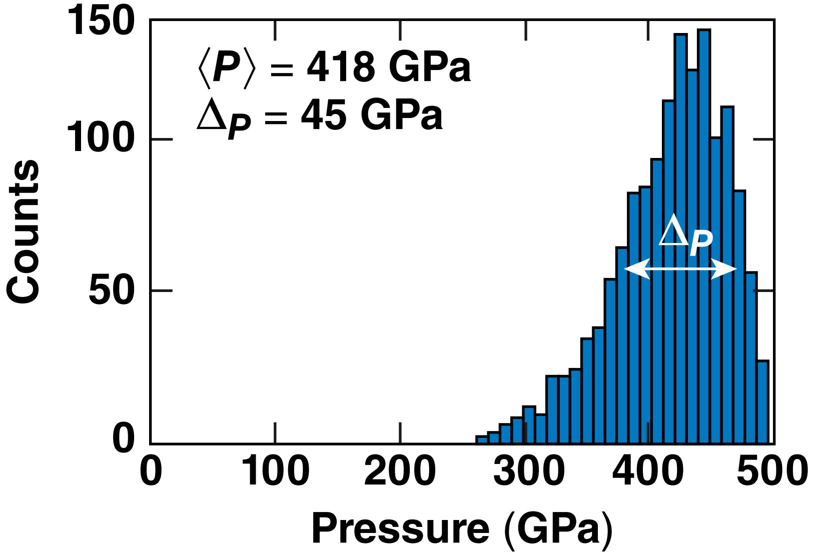
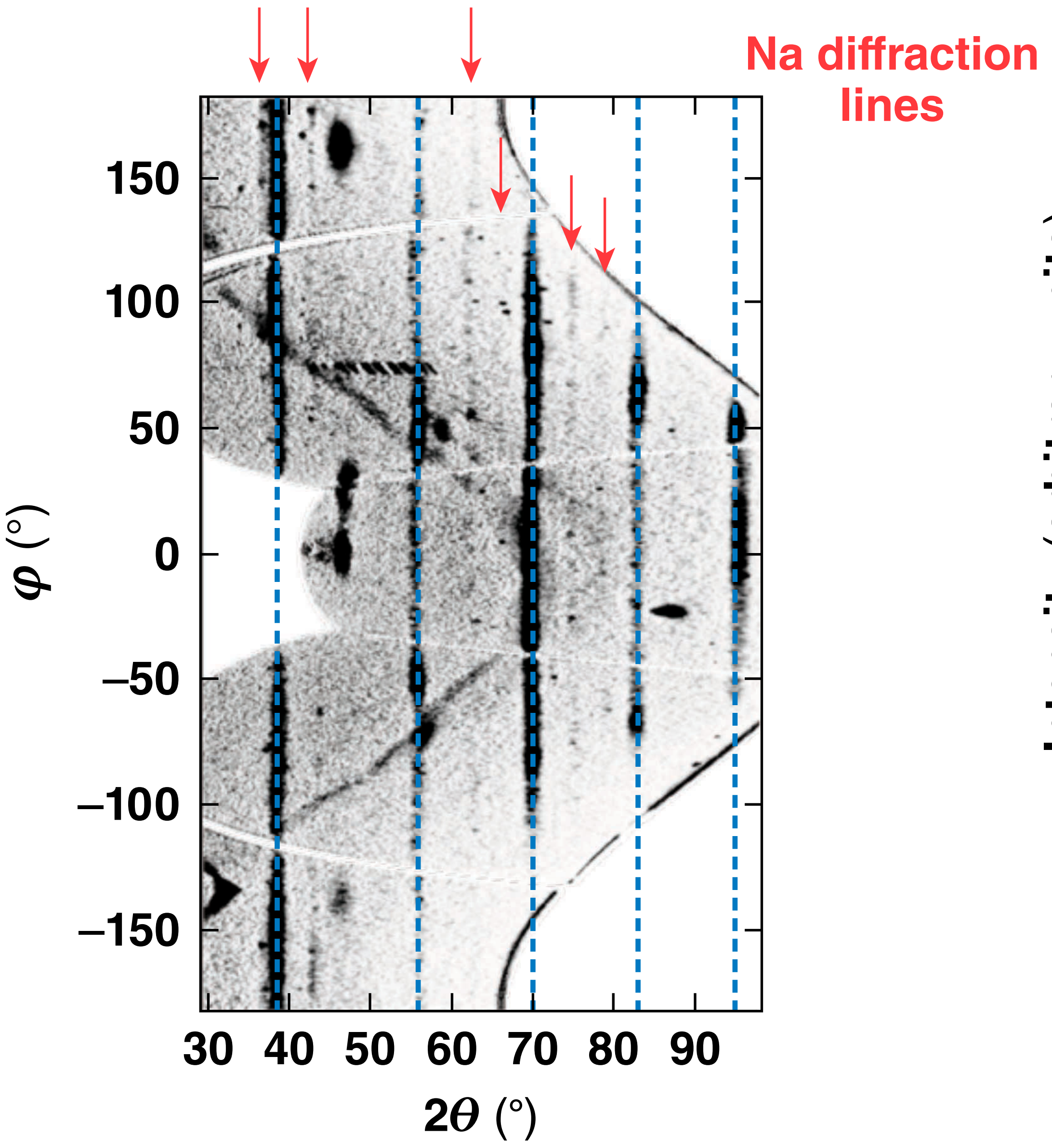
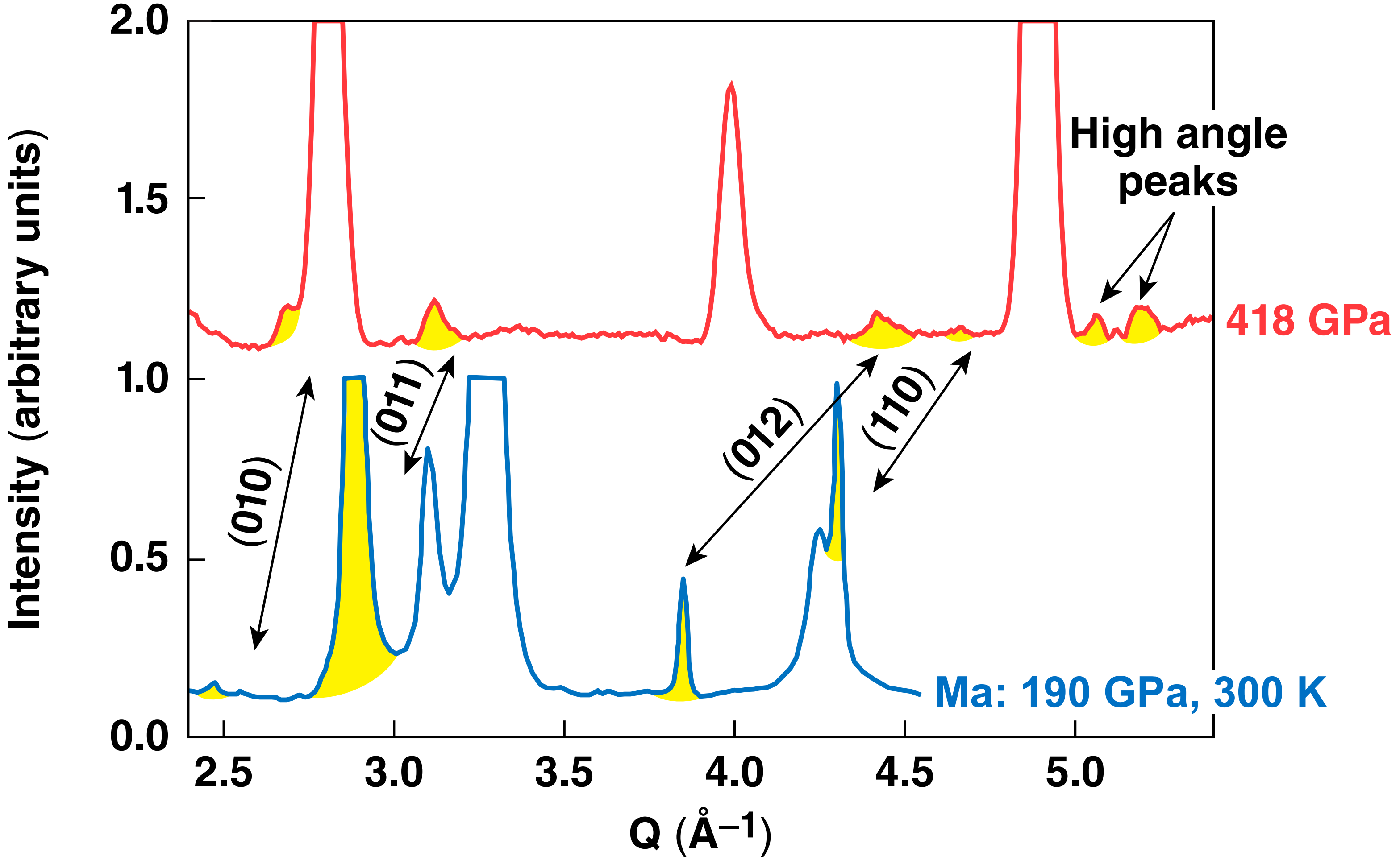
Backward characteristics algorithm



Evidence of solid Na was observed at 418 GPa with up to six diffraction peaks from compressed Na





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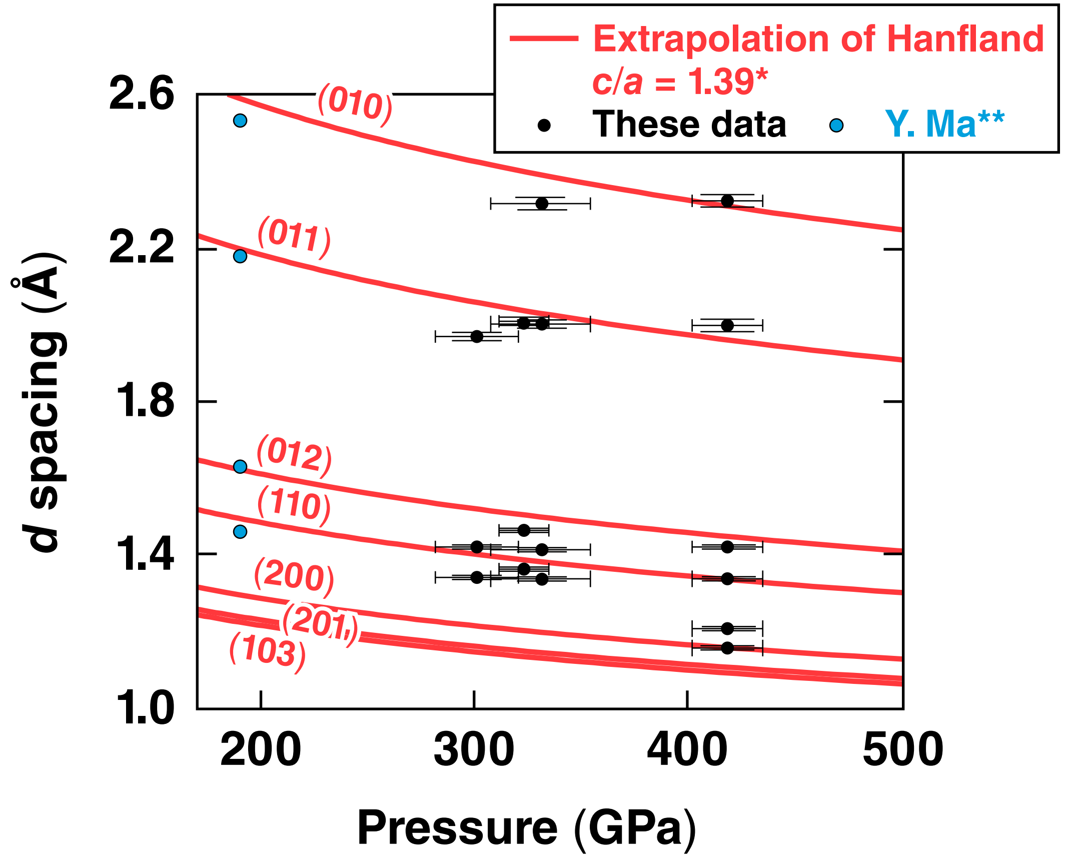
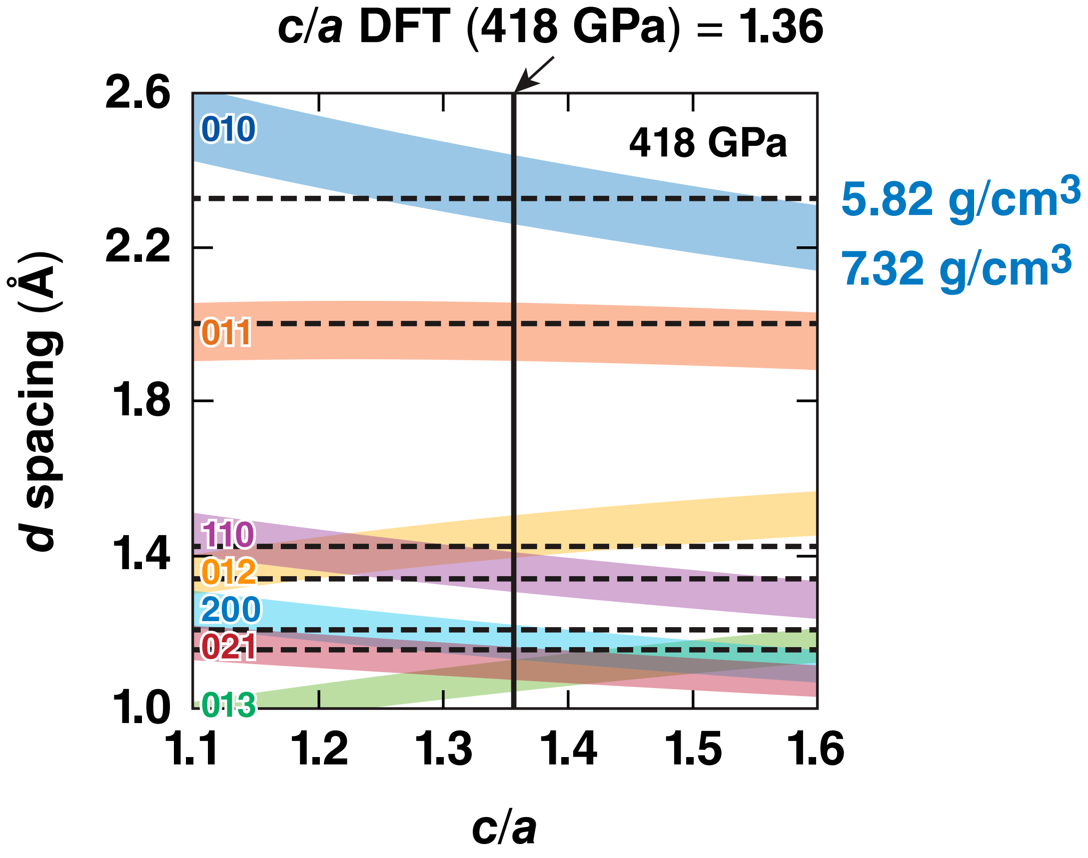
Y. Ma *et al.*, Nature 458, 182 (2009).

A single, reproducible high-pressure solid phase is observed in Na ramp compressed to 300 to 418 GPa



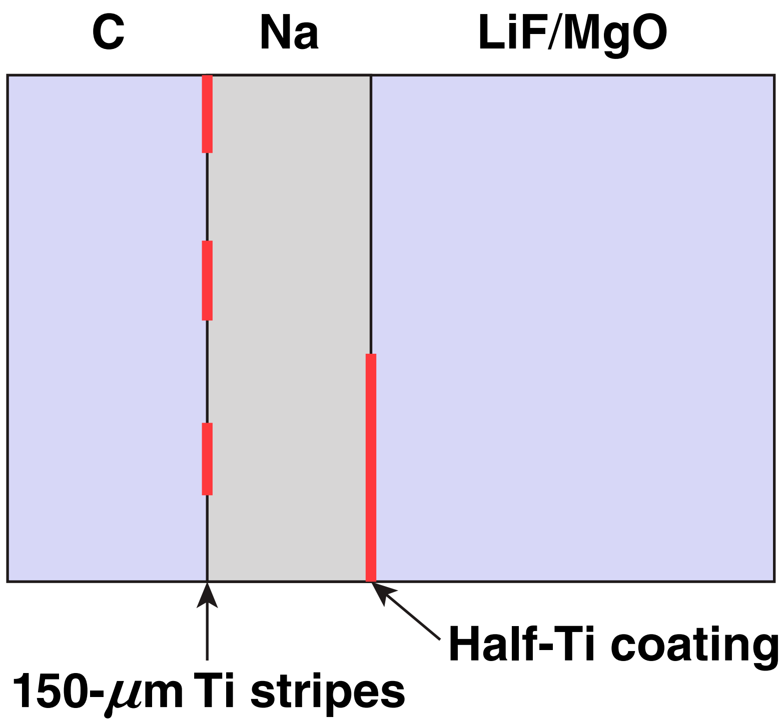


This phase exhibits sixfold compression and is not fully consistent with the hP4 phase.

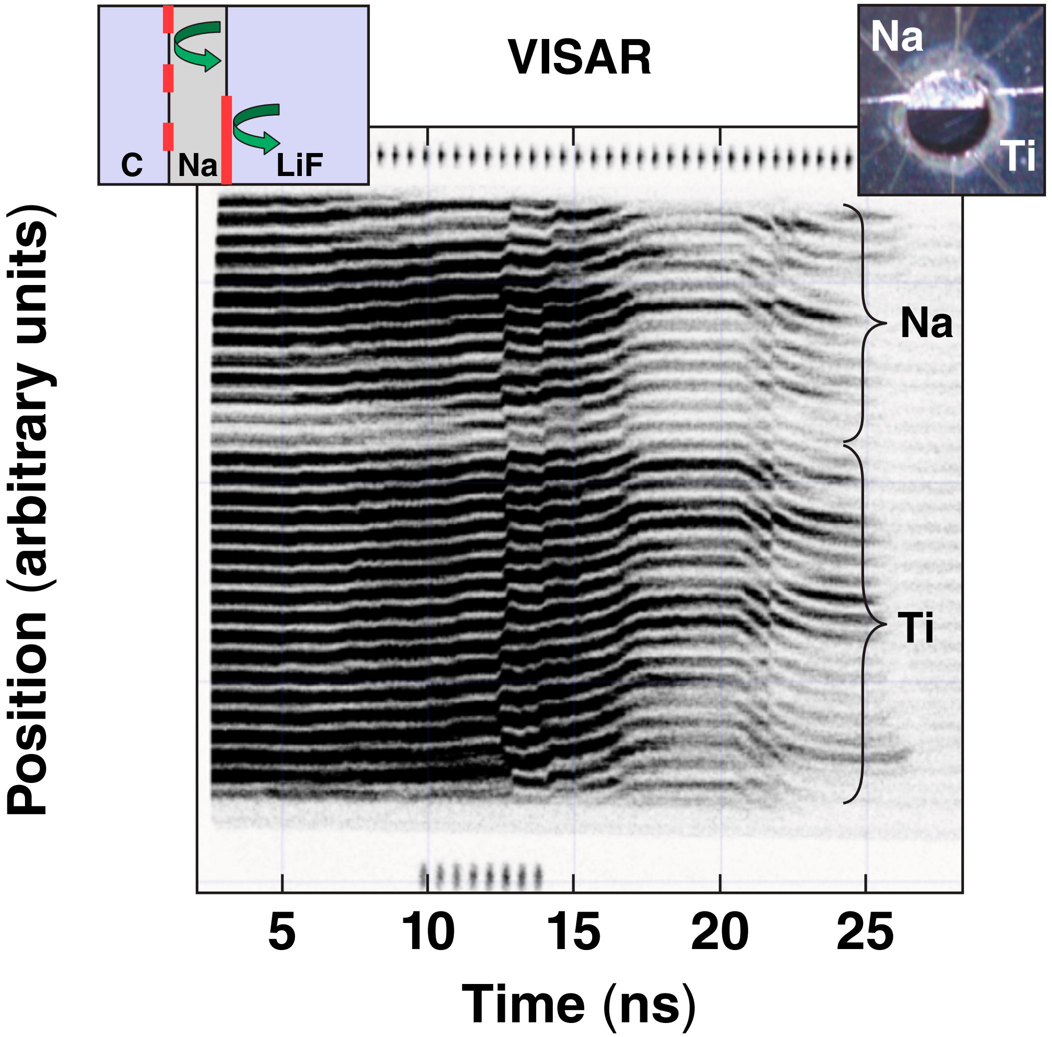
DFT: density functional theory

[*] M. Hanfland *et al.*, Phys. Rev. B. 65, 184109 (2002).
[**] Y. Ma *et al.*, Nature 458, 182 (2009)

A transparent window with a reflectance pattern of Ti stripes is used to detect Na transparency at 532 nm; Na remains reflective to at least 290 GPa



Elevated temperatures may cause delocalization of the electrons and thermal population of the conduction band in Na.



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A simple semiconducting Drude picture is used to constrain the bandgap and temperature of dense Na

Temperature-activated carrier concentration

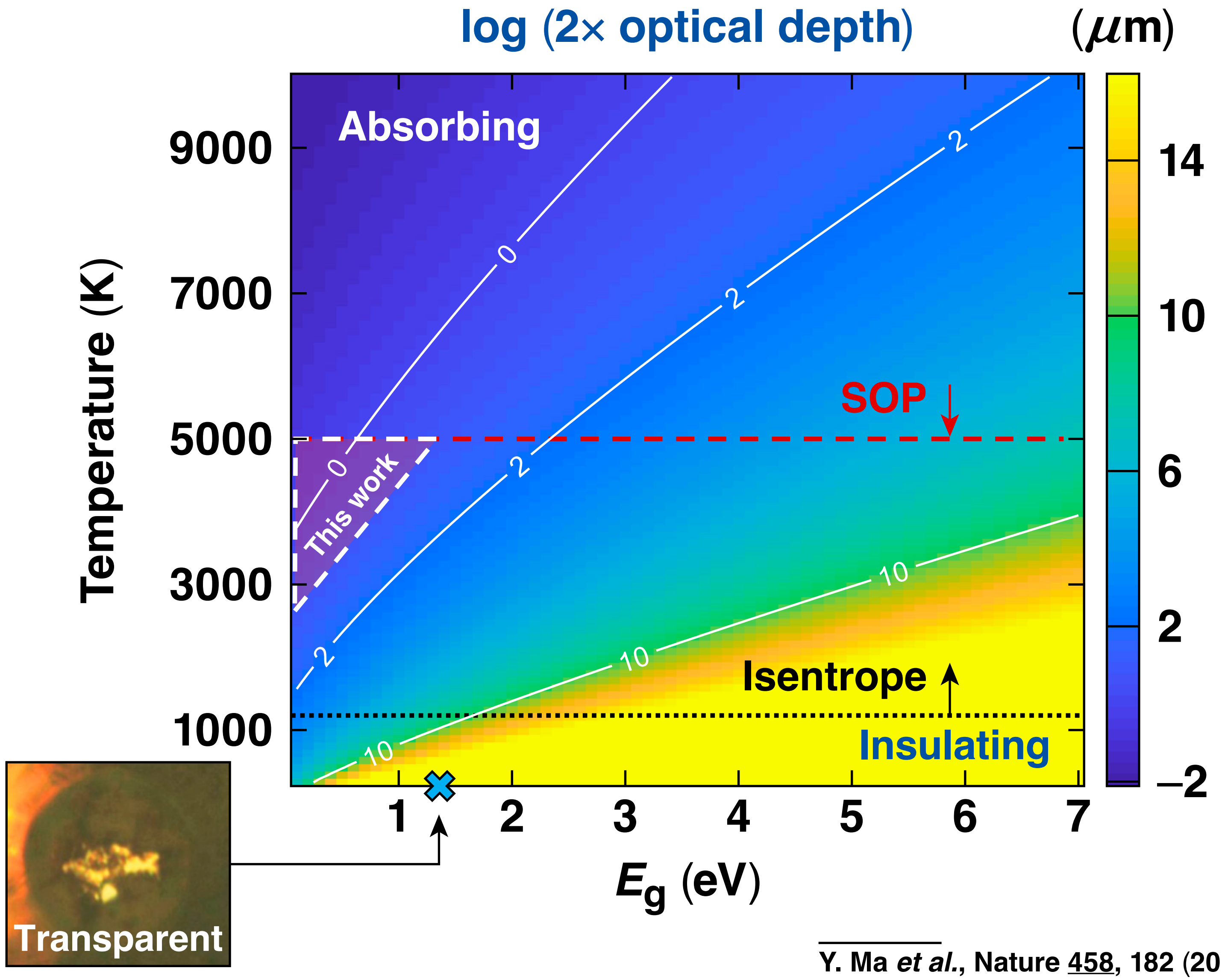
$$N_{\mathrm{e}} = 2\left(\frac{m_{\mathrm{eff}} k_{\mathrm{B}} T}{2\pi\hbar^{2}}\right)^{\frac{3}{2}} F\left(-E_{\mathrm{g}}/2k_{\mathrm{B}}T\right)$$

Dielectric function

$$\epsilon = \epsilon_{\mathrm{b}} + \frac{i}{\omega\epsilon_{0}}\sigma$$

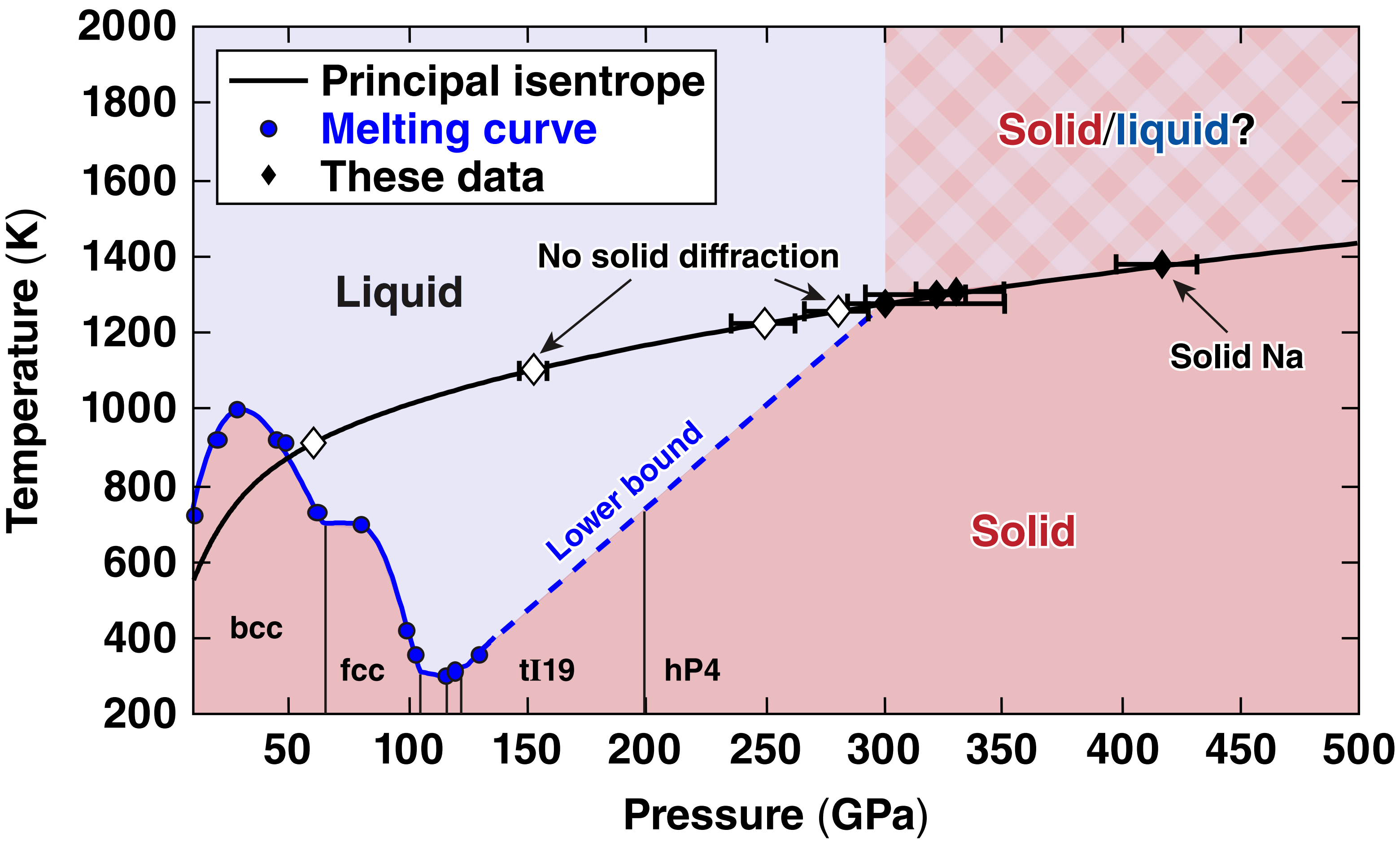
Bound electrons

Free carriers (Drude conductivity)



Y. Ma *et al.*, Nature 458, 182 (2009).
SOP: streaked optical pyrometer

We are mapping out the Na phase diagram to 500 GPa



A dramatic increase in the melting temperature occurs above 140 GPa.

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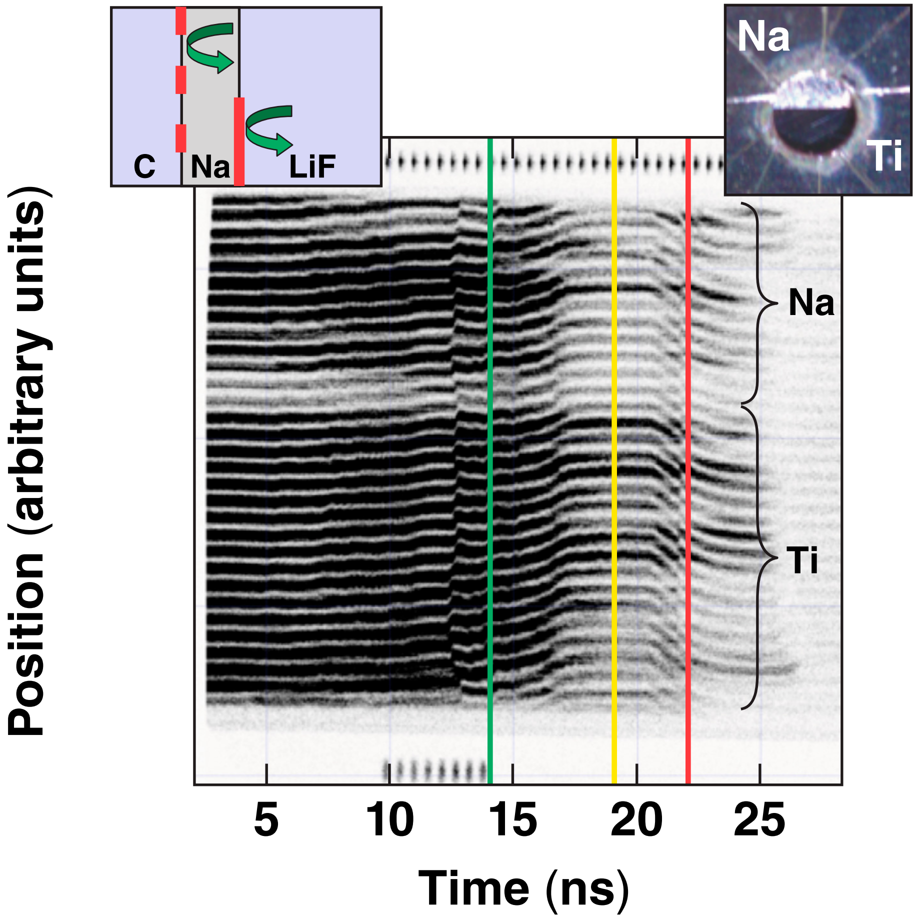
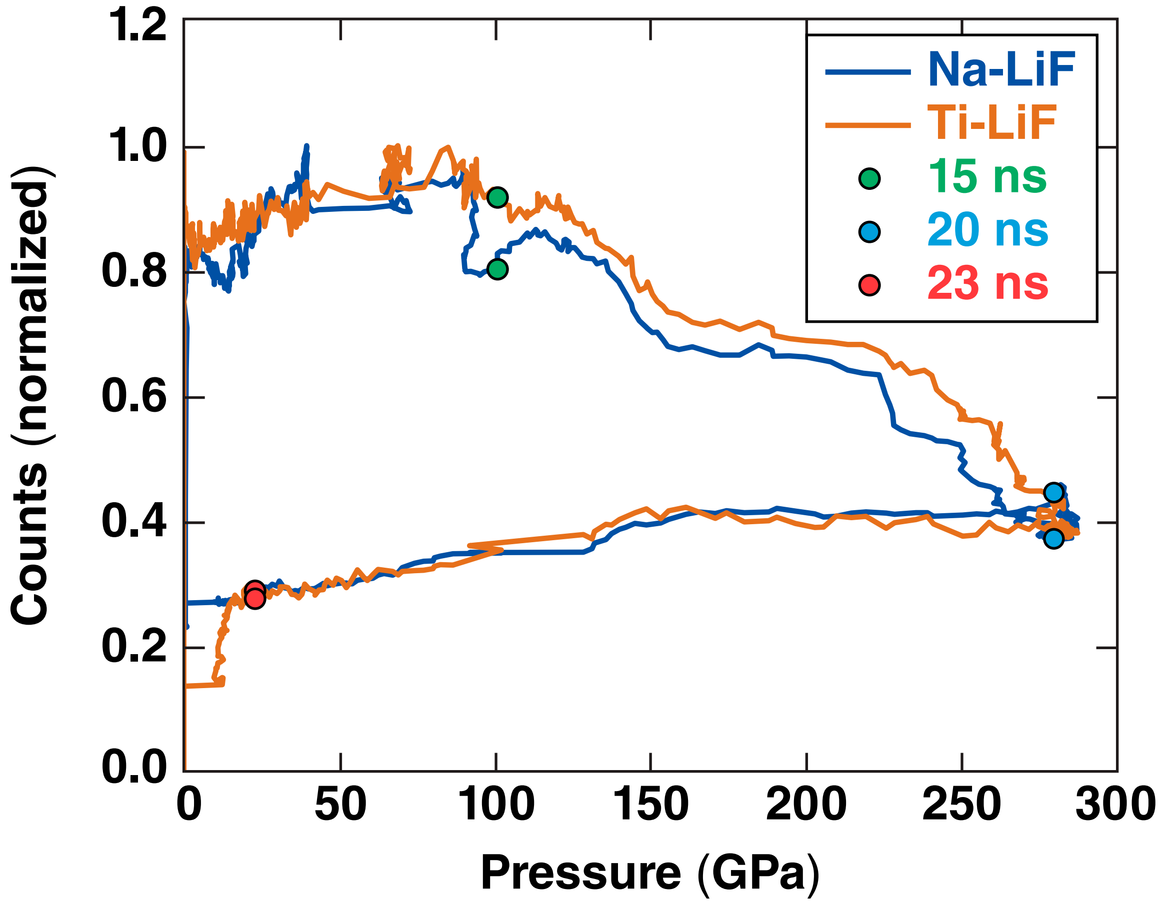
Summary/Conclusions

Melting and resolidification of Na to a solid phase is observed for pressures greater than 300 GPa

- Alkali metals, including Na, serve as a prototypical system to guide theory for ultradense matter beyond the Thomas–Fermi model
- High-power lasers ramp compressed Na and nanosecond *in-situ* x-ray diffraction (XRD) was used to measure the crystal structure at pressures up to 418 GPa
- Solid Na was observed at 301 to 418 GPa; therefore, the melting temperature increases dramatically above 140 GPa
- Simultaneous XRD and optical reflectivity measurements reveal that Na remains reflective to at least 290 GPa, in striking contrast to room-temperature static-compression measurements

Na remains reflective to at least 290 GPa, and the variation in the reflectivity is caused by changes in the window

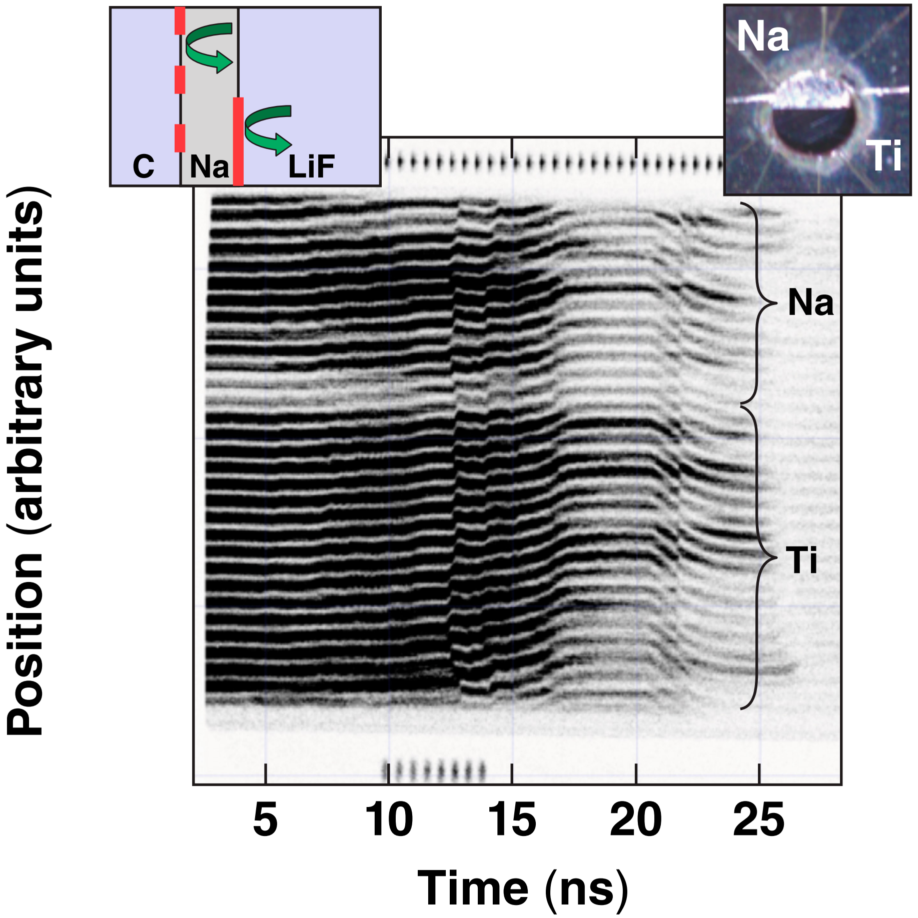


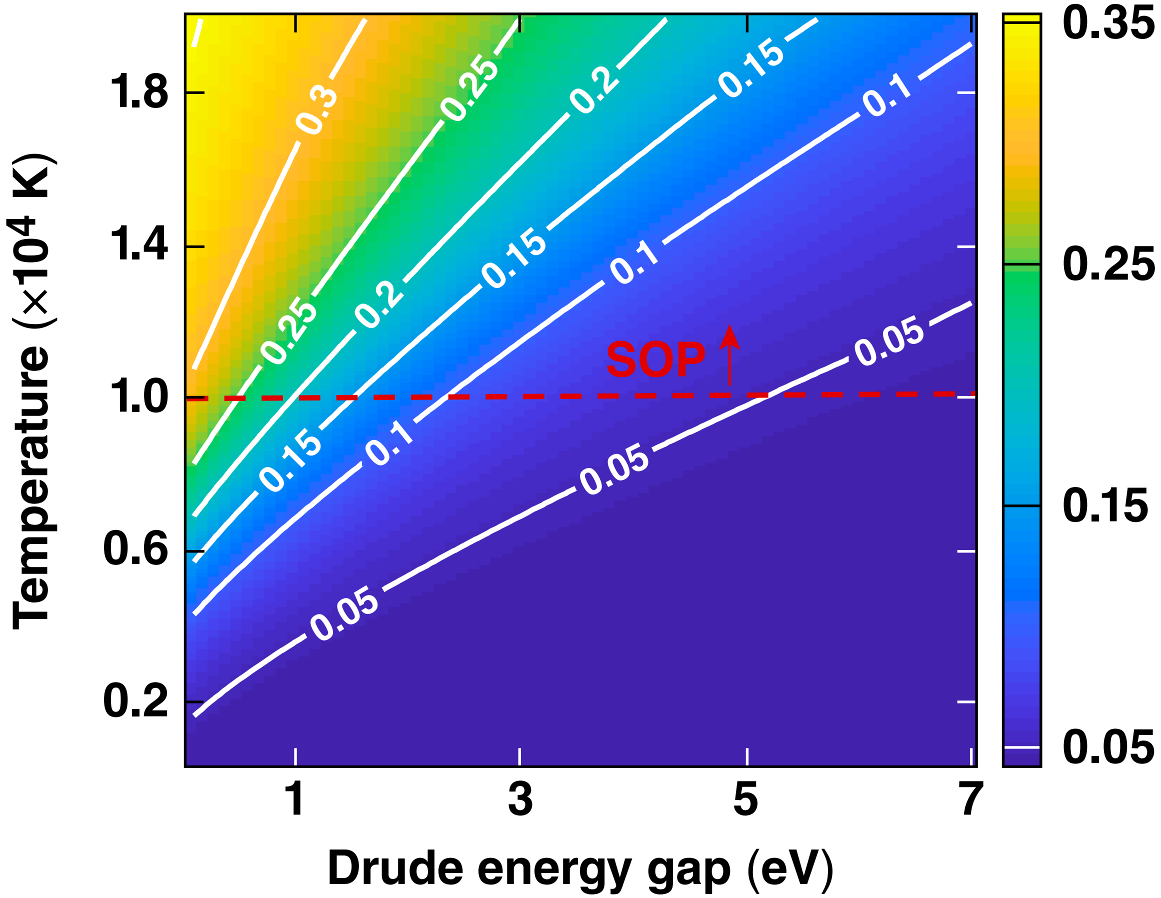


Elevated temperatures may cause delocalization of the electrons and thermal population of the conduction band in Na.

E27659b

Reflectivity measurements of Na suggest a gap has opened to 2 eV at 290 GPa





Elevated temperatures may cause delocalization of the electrons and thermal population of the conduction band in Na.

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