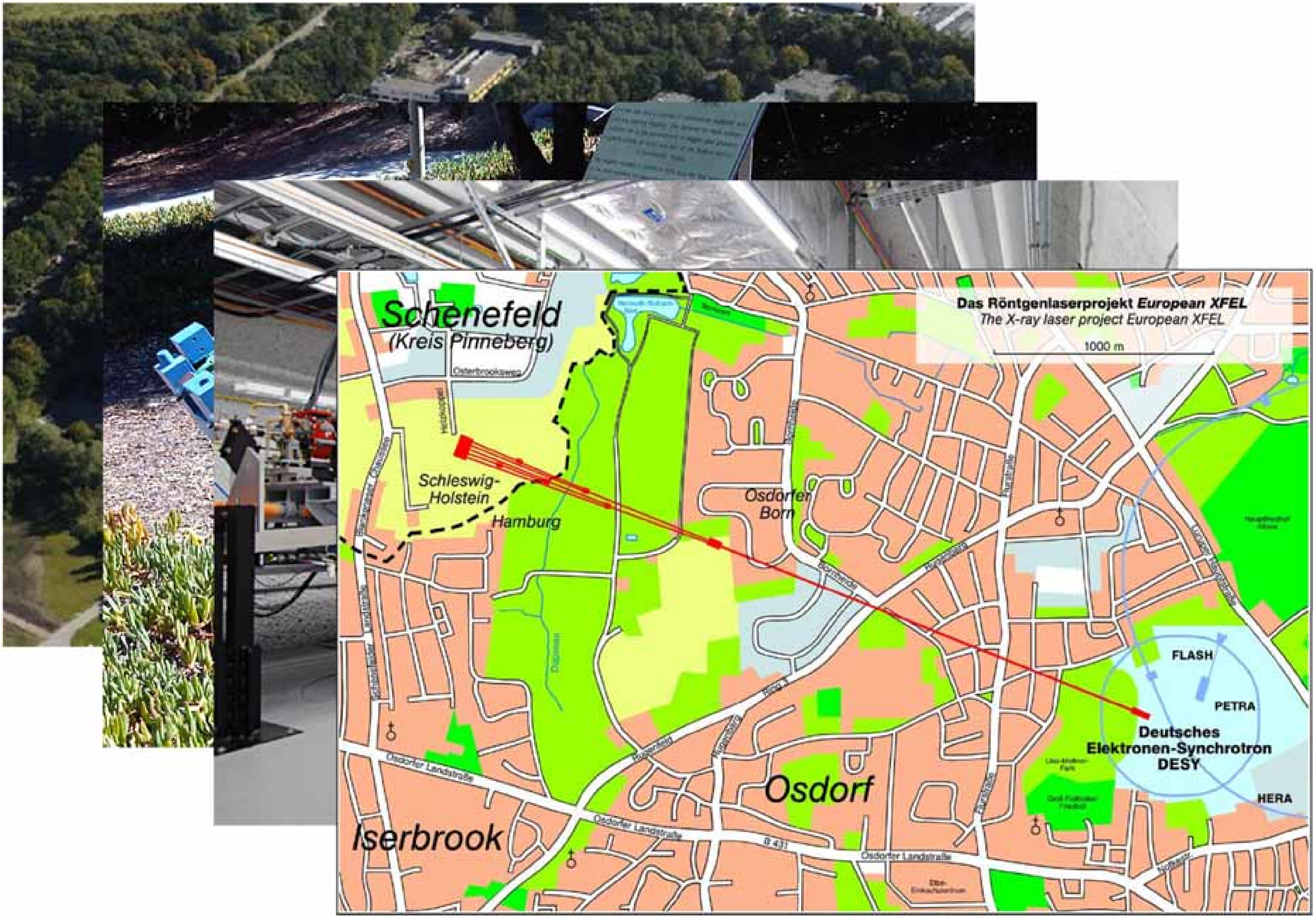
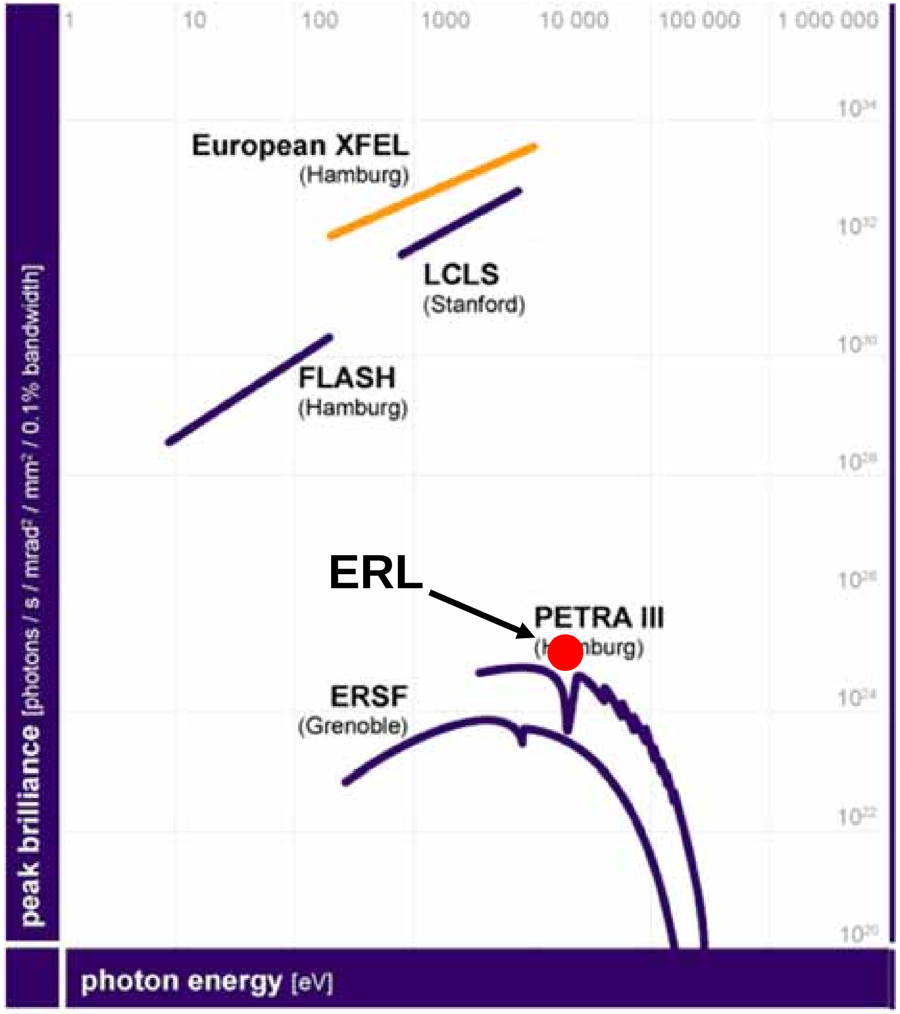
Single-Crystal XRD and IXS of Elements Under Extreme Pressure

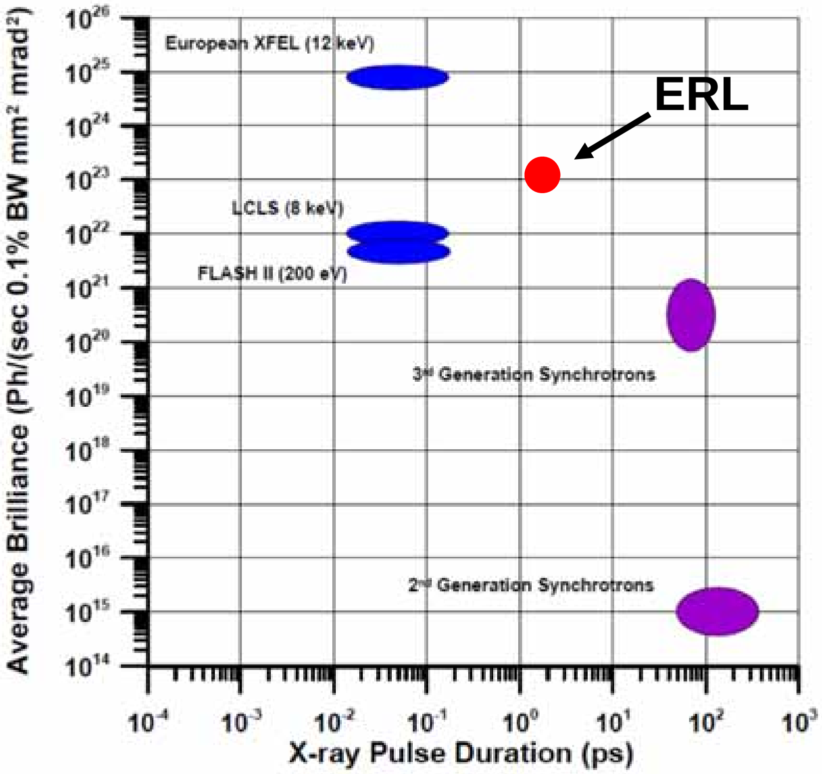
Malcolm McMahon, The University of Edinburgh

1997


Das Röntgenlaserprojekt *European XFEL*
The X-ray laser project European XFEL
1000 m
Schenefeld
(Kreis Pinneberg)
Schleswig-
Holstein
Hamburg
Osdorfer
Born
Osdorf
Iserbrook
FLASH
PETRA
HERA
Deutsches
Elektronen-Synchrotron
DESY
Osdorfer Landstraße
B 431



Peak Brilliance



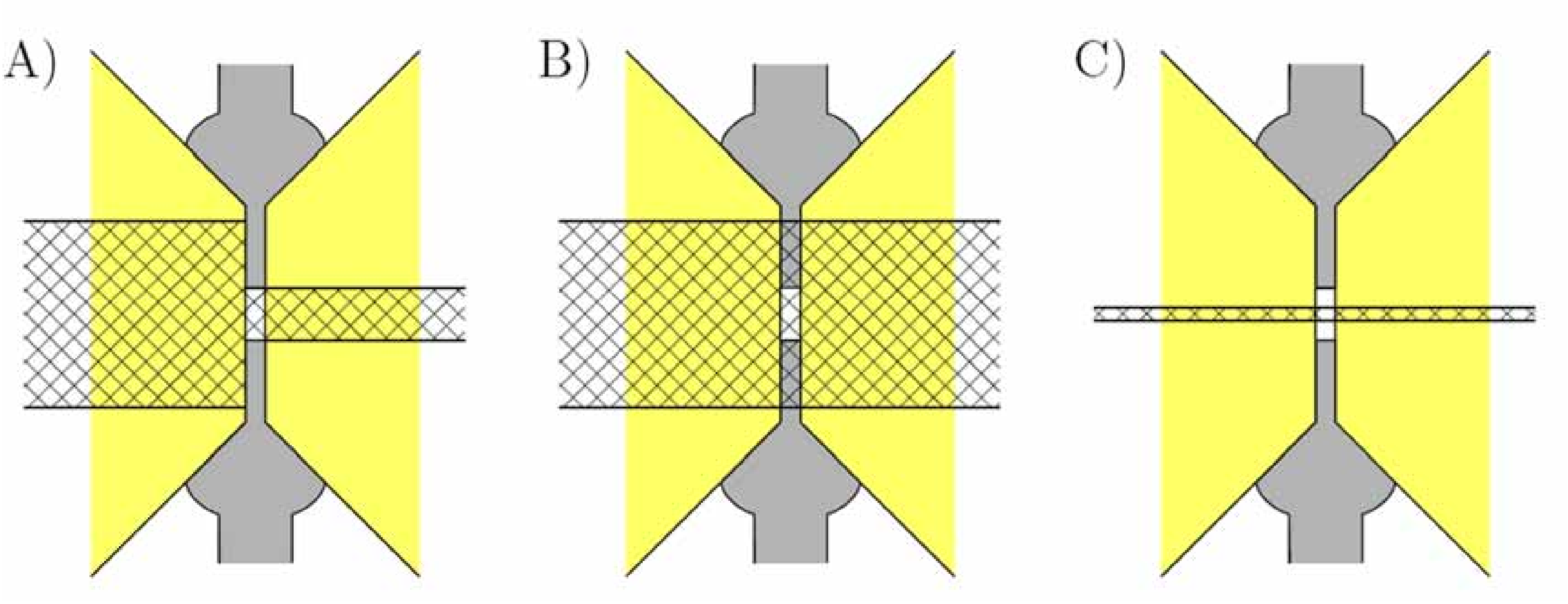
Average Brilliance

- 100 fs hard x-ray pulses at MHz+ pulse rates (XFEL)
- 2 ps hard x-ray pulses at GHz+ pulse rates (ERL)
- coherent x-ray radiation

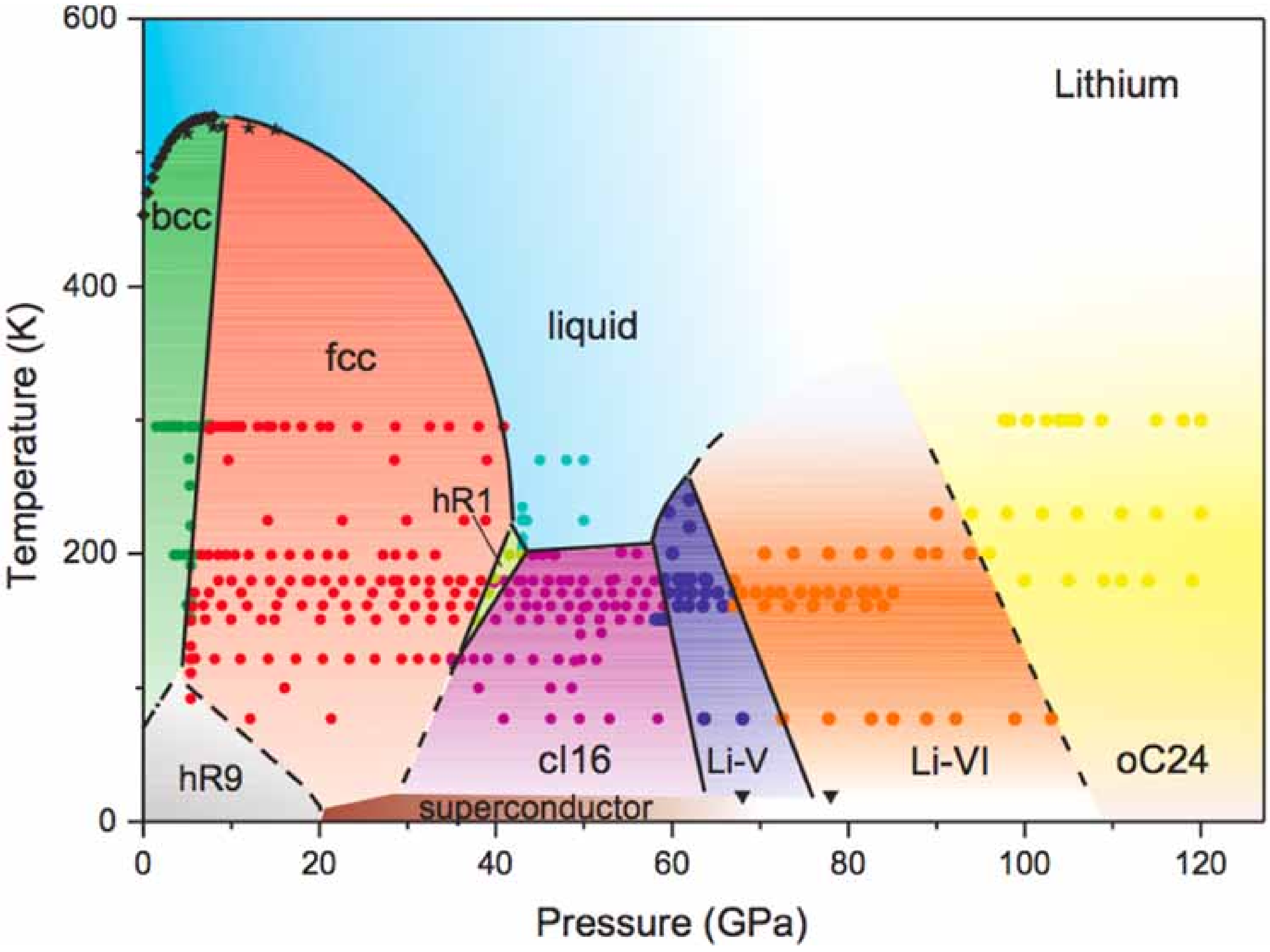
HS3090 - Long Term Project at the ESRF

Utilise extreme intensity, micro-focused beams and high energy of ESRF to:

- Push single-crystal diffraction to 100+ GPa
- Extend high-P high-T methods
- Develop high-P low-T methods

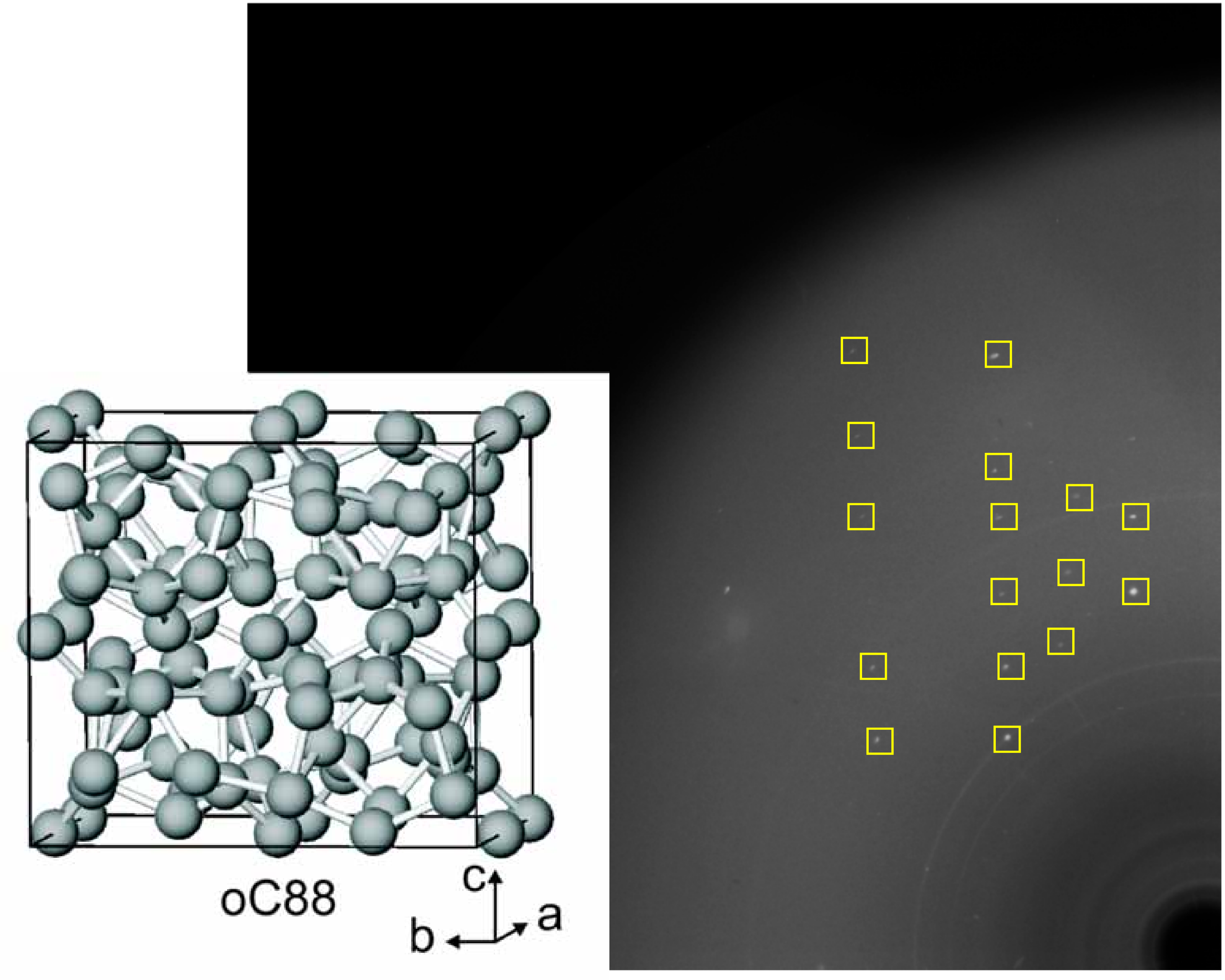


Sample 15-30 μm diam & 3 μm thick, 5-15 μm diam beam, λ~ 0.3-0.4 Å

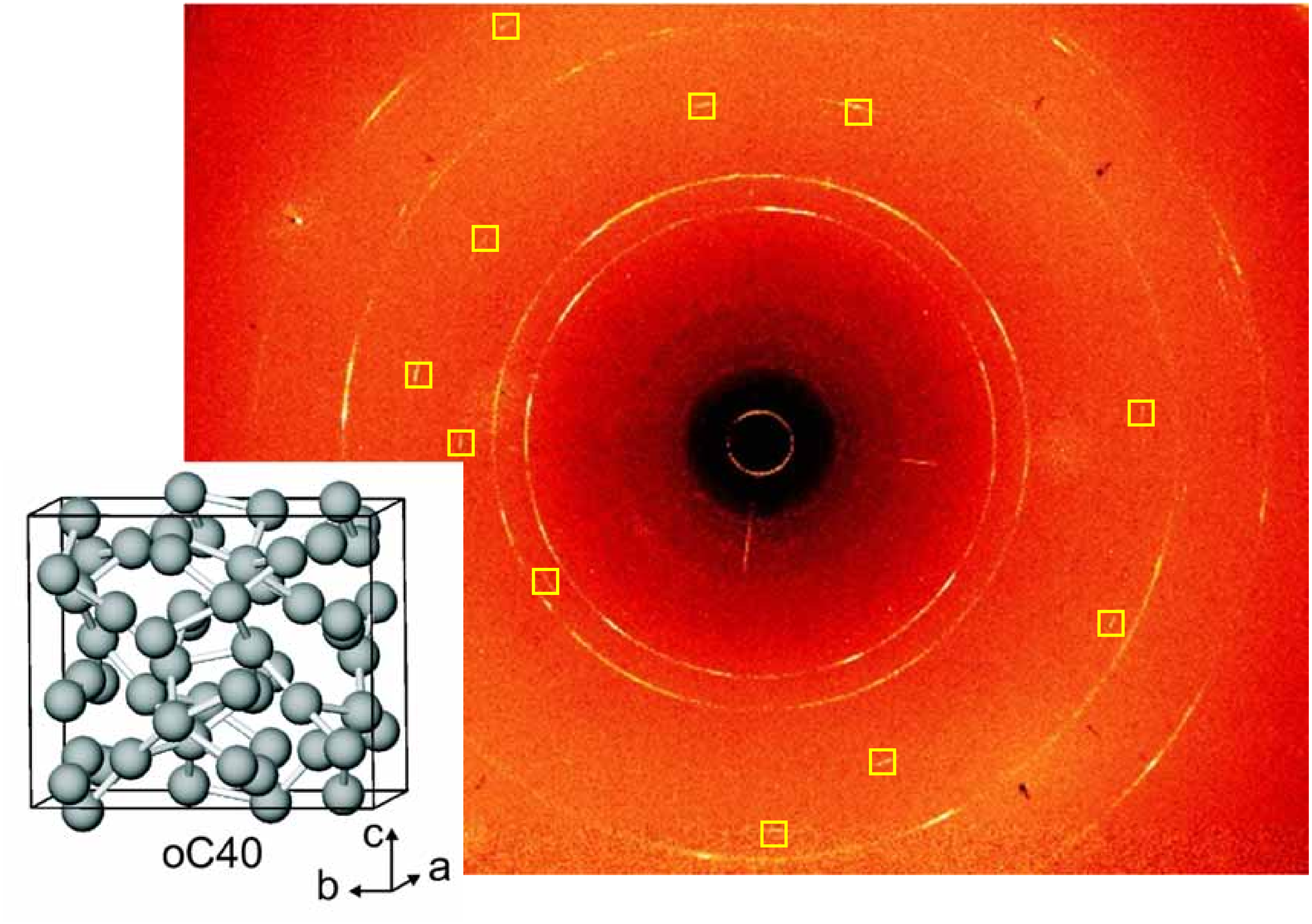


Gregoryanz *et al* Nature Mat (2011)
Marques *et al* PRL (2011)

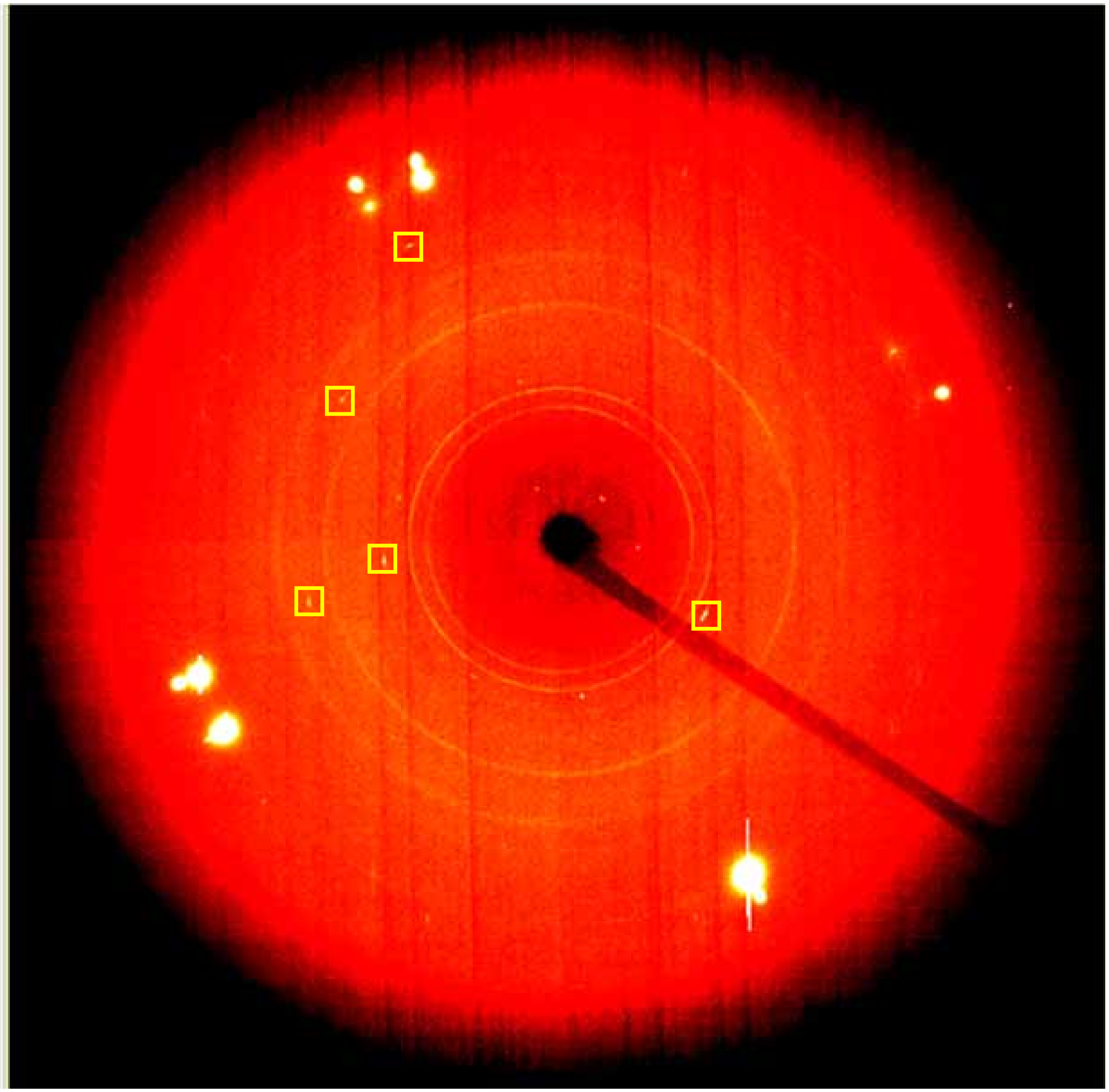
Single Crystal Diffraction from Li-V at ~65 GPa & 200 K

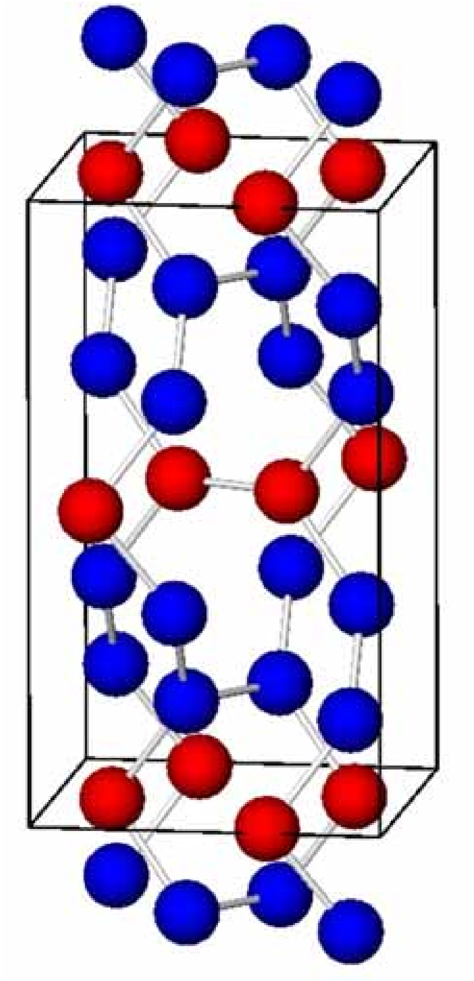


Single Crystal Diffraction from Li-VI at ~75 GPa & 200 K



Single Crystal Diffraction from Li-VII at ~125 GPa & 300 K



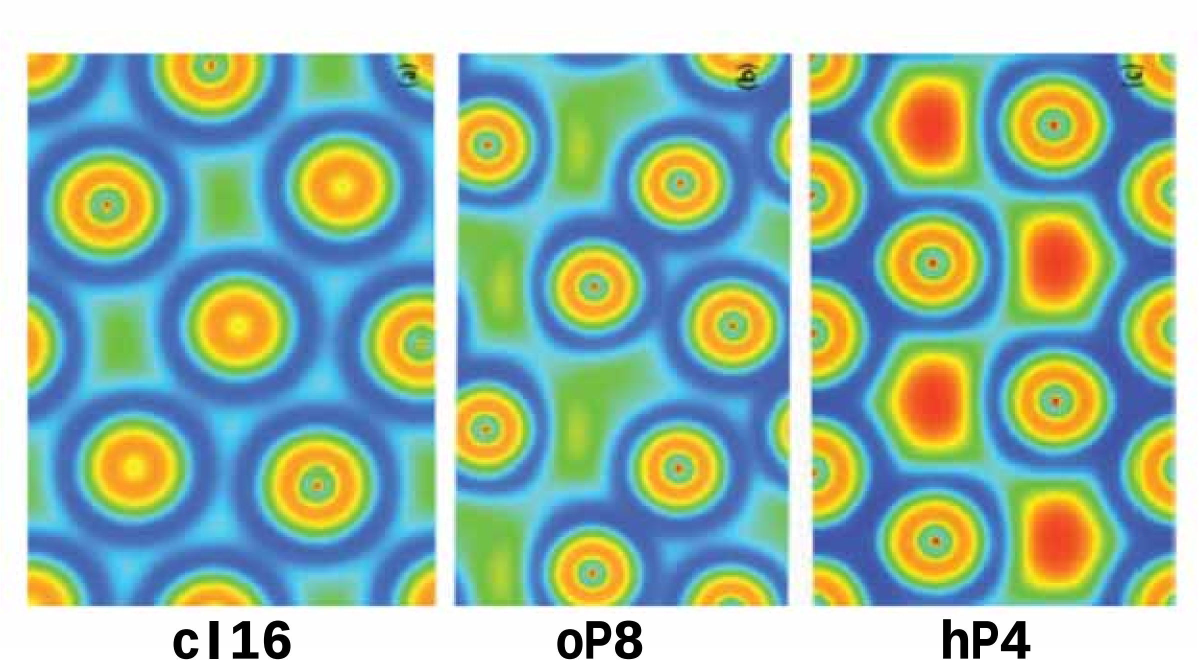


oC24

What similar experiments might one imagine doing on ERL and XFEL???

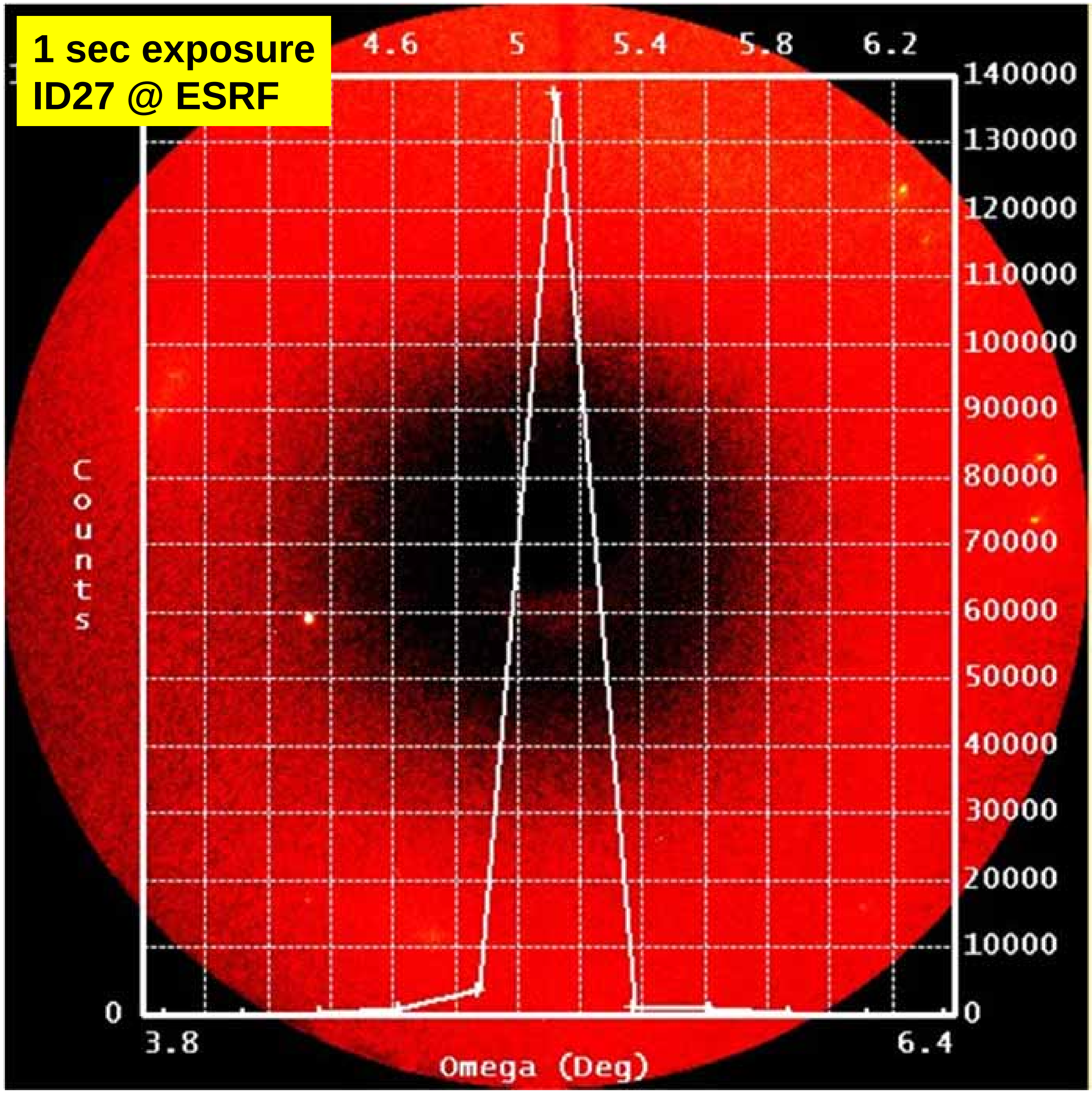
Megabar single-crystal diffn. (D_2-II/III, metallic O_2, Li-VIII, Ice-X)

- ~micron sized beam
- 60 keV x-rays
- use short pulse length to minimise DAC background (2 ps = 600 microns)
- direct determination of e– density



Ma *et al*
Nature 2009

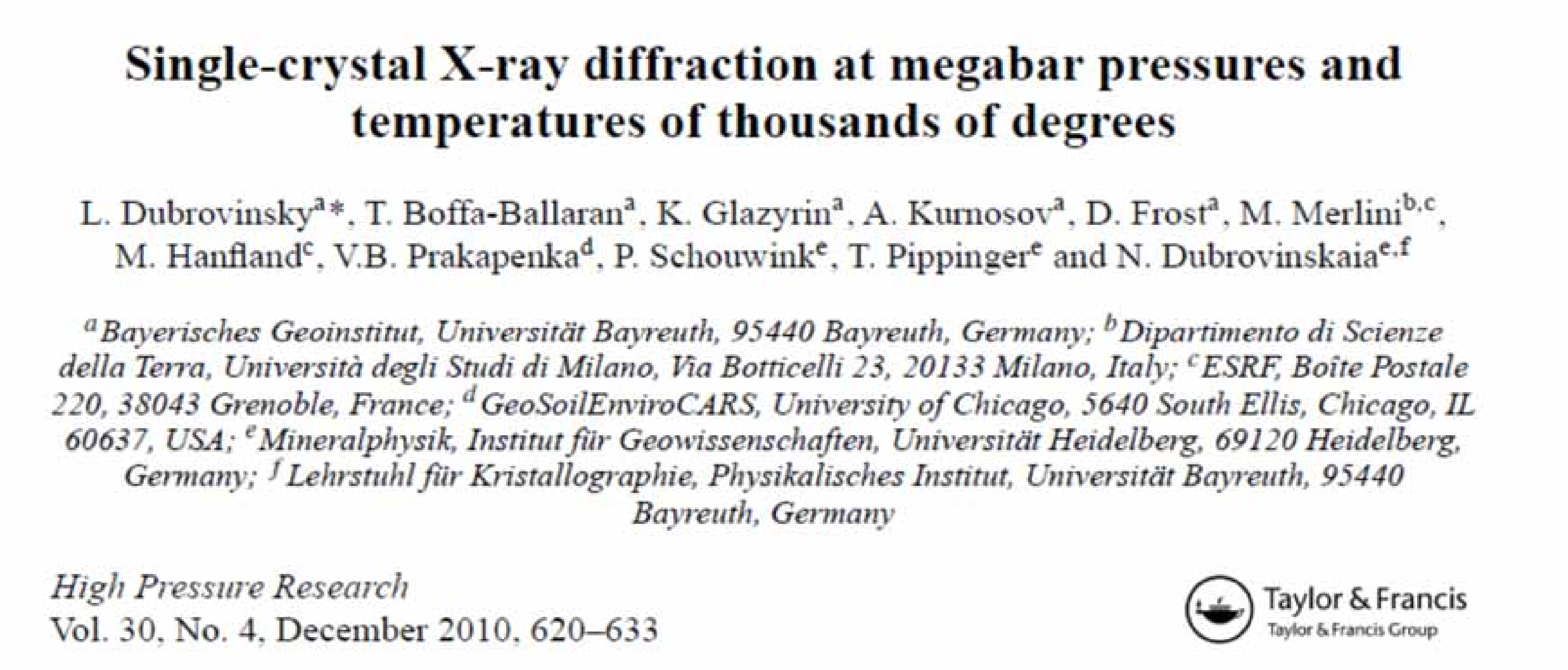
Marques *et al*
PRB 2011



1 sec exposure
ID27 @ ESRF

Diffraction Studies of Dynamically-Heated Single Crystals

- Using either x-ray beam itself to heat (XFEL)
- Or a separate IR laser



Single-crystal X-ray diffraction at megabar pressures and temperatures of thousands of degrees

L. Dubrovinsky[a]*, T. Boffa-Ballaran[a], K. Glazyrin[a], A. Kurnosov[a], D. Frost[a], M. Merlini[b,c], M. Hanfland[c], V.B. Prakapenka[d], P. Schouwink[e], T. Pippinger[e] and N. Dubrovinskaia[e,f]

[a]Bayerisches Geoinstitut, Universität Bayreuth, 95440 Bayreuth, Germany; [b]Dipartimento di Scienze della Terra, Università degli Studi di Milano, Via Botticelli 23, 20133 Milano, Italy; [c]ESRF, Boîte Postale 220, 38043 Grenoble, France; [d]GeoSoilEnviroCARS, University of Chicago, 5640 South Ellis, Chicago, IL 60637, USA; [e]Mineralphysik, Institut für Geowissenschaften, Universität Heidelberg, 69120 Heidelberg, Germany; [f]Lehrstuhl für Kristallographie, Physikalisches Institut, Universität Bayreuth, 95440 Bayreuth, Germany

High Pressure Research
Vol. 30, No. 4, December 2010, 620–633

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But We Can Also Imagine Doing Entirely New Things

- Time-resolved Studies of the Dynamics of Melting
- Time and Momentum Resolved Imaging of Non-equilibrium Phonons
- Electron-Lattice Coupling in CDW Phases of Elements
- Diffraction from Dynamically Compressed Matter

What would we need:

- Fast fs heating lasers synchronised to x-ray beam
- Ability to split x-ray pulses into two, with variable time delay
- Ability to bring together pulses of different wavelengths
- Largest possible ns laser for ramp compression

Dynamics of Melting/Crystallisation

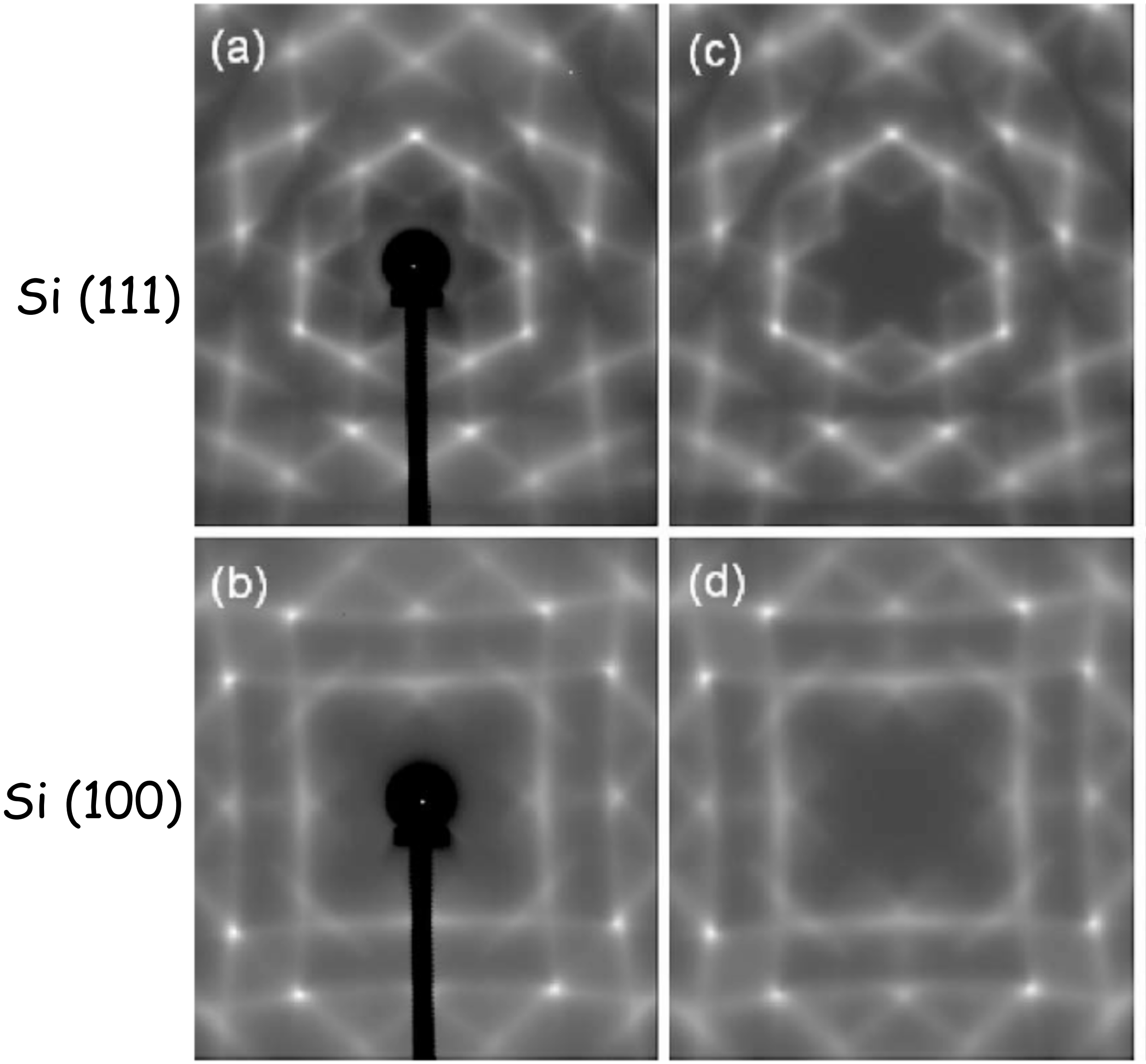
- How do materials melt/recrystallise – including non-thermal melting?
- How do they behave on the approach to melting?

PRL 95, 125701 (2005) PHYSICAL REVIEW LETTERS week ending 16 SEPTEMBER 2005

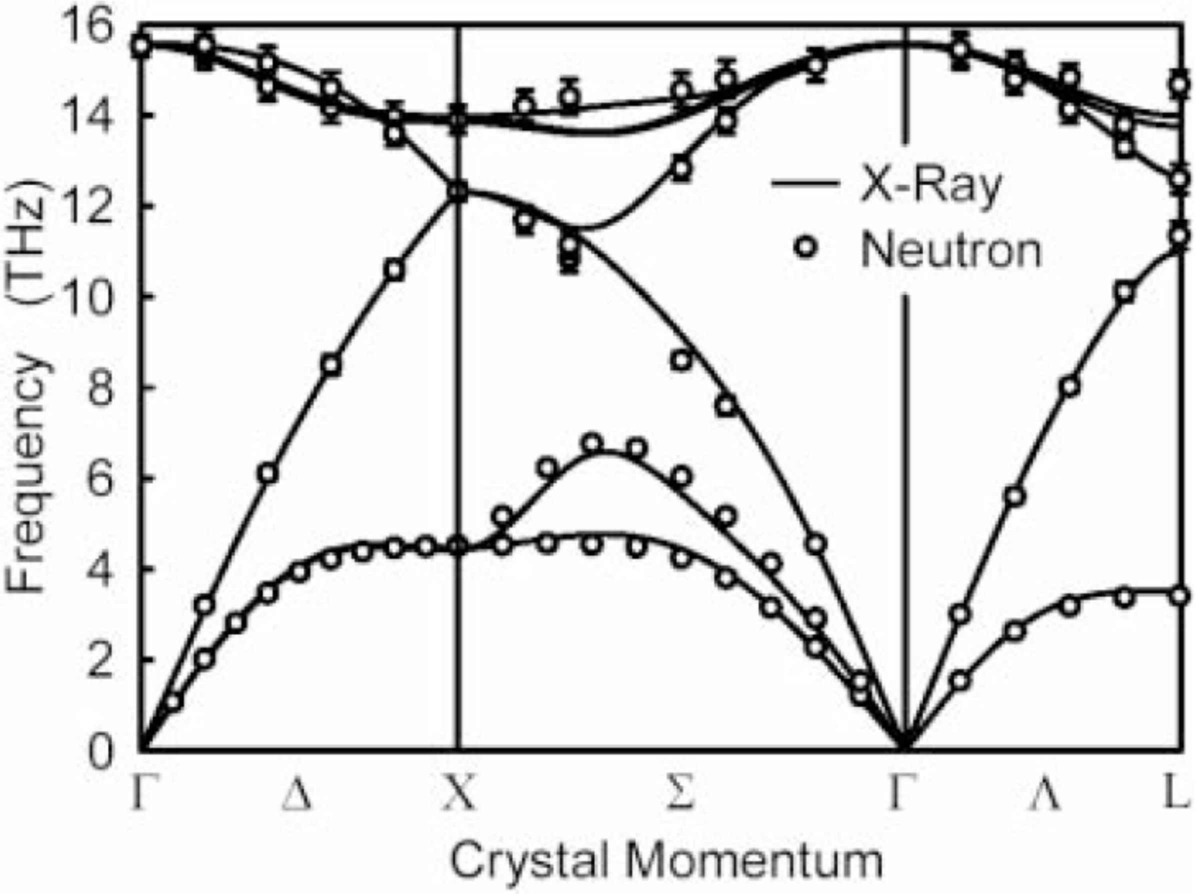
Observation of Structural Anisotropy and the Onset of Liquidlike Motion During the Nonthermal Melting of InSb

K. J. Gaffney,[1] A. M. Lindenberg,[1] J. Larsson,[2] K. Sokolowski-Tinten,[3,4] C. Blome,[5] O. Synnergren,[2] J. Sheppard,[6] C. Caleman,[7] A. G. MacPhee,[8] D. Weinstein,[8] D. P. Lowney,[8] T. Allison,[8] T. Matthews,[8] R. W. Falcone,[8] A. L. Cavalieri,[9,10] D. M. Fritz,[9] S. H. Lee,[9] P. H. Bucksbaum,[9] D. A. Reis,[9] J. Rudati,[11] A. T. Macrander,[11] P. H. Fuoss,[12] C. C. Kao,[13] D. P. Siddons,[13] R. Pahl,[14] K. Moffat,[14] J. Als-Nielsen,[15] S. Duesterer,[5] R. Ischebeck,[5] H. Schlarb,[5] H. Schulte-Schrepping,[5] J. Schneider,[5] D. von der Linde,[4] O. Hignette,[16] F. Sette,[16] H. N. Chapman,[17] R. W. Lee,[17] T. N. Hansen,[2] J. S. Wark,[6] M. Bergh,[7] G. Huldt,[7] D. van der Spoel,[7] N. Timneanu,[7] J. Hajdu,[7] R. A. Akre,[18] E. Bong,[18] P. Krejcik,[18] J. Arthur,[1] S. Brennan,[1] K. Luening,[1] and J. B. Hastings[1]

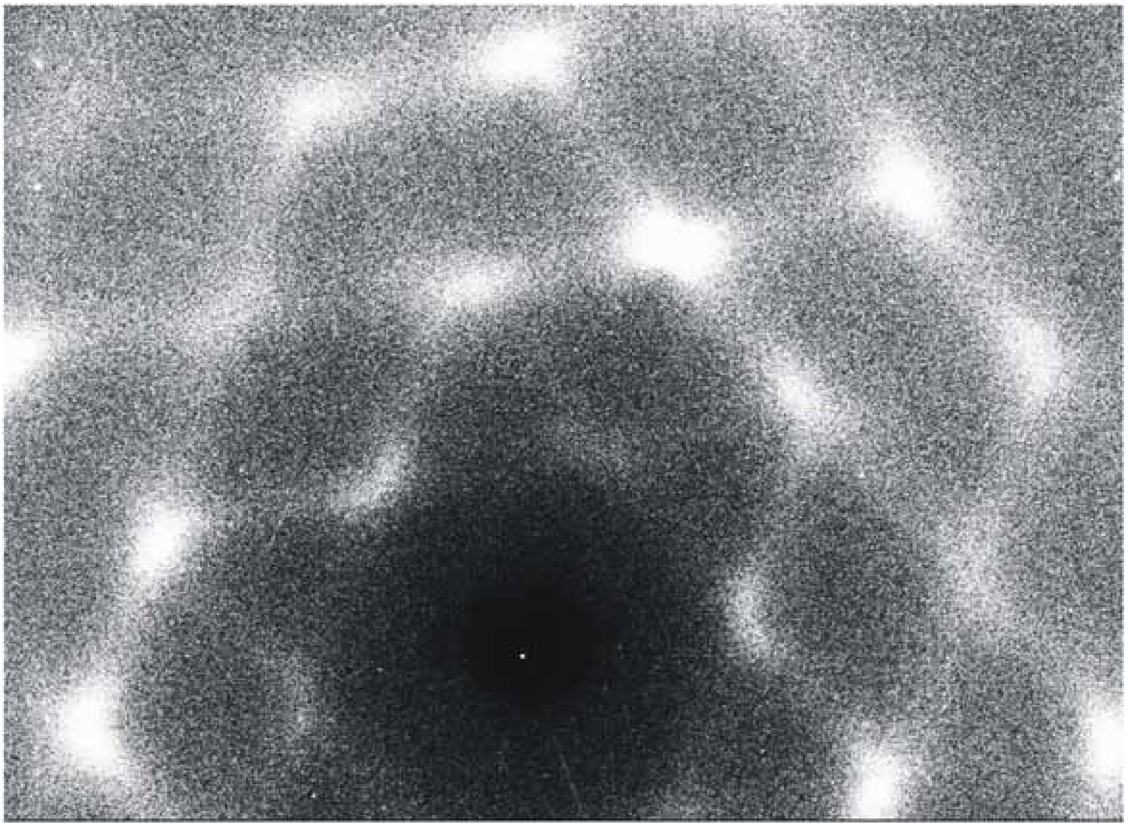
Classical approach to measuring phonons and dispersion – inelastic neutron and x-ray scattering



Holt *et al*, PRL **83**, 3317 (1999).



Agreement with neutron experiments is excellent

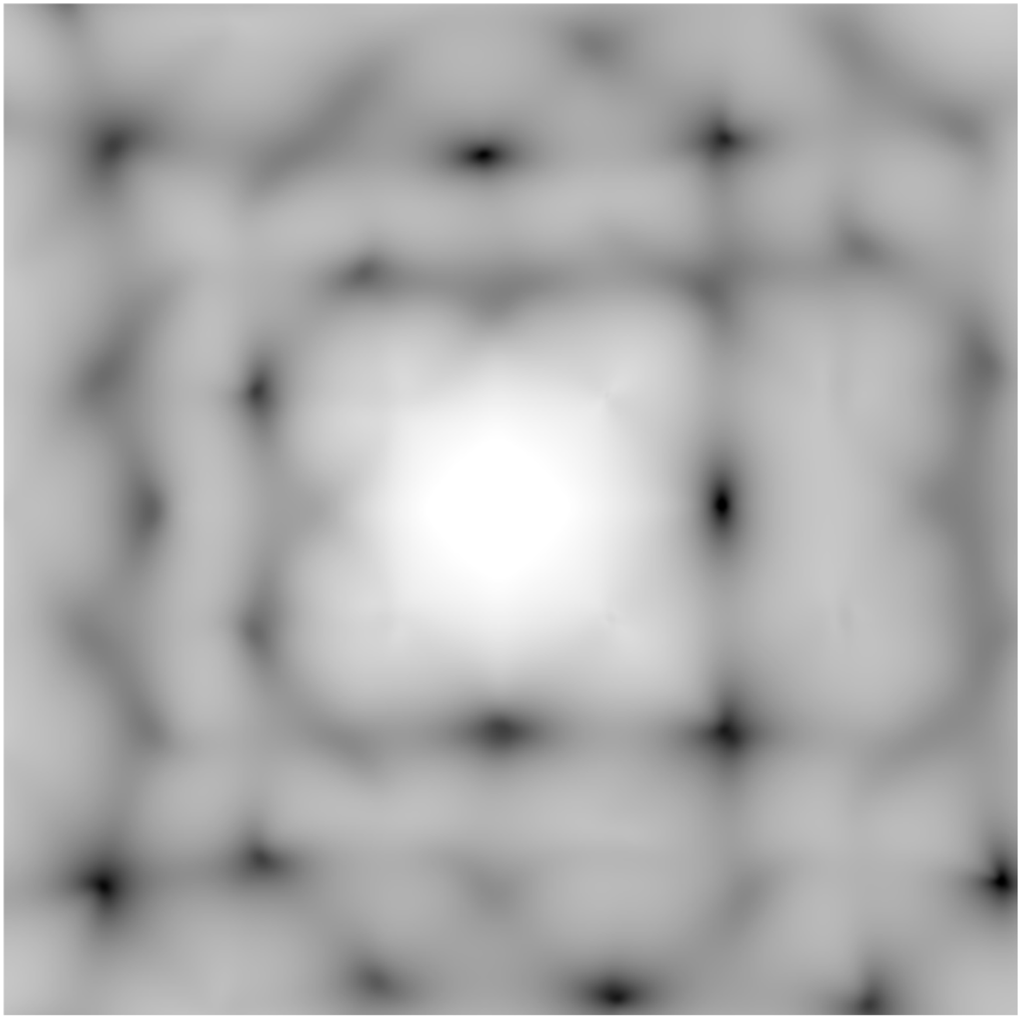


TDS from Cs-II at 3 GPa

Slow (ns to μs) **l**aser-induced melting – IR lasers well known to the HP community

Fast (fs) laser-induced melting – heats only electrons and not the lattice *or* excite a particular optical phonon to give displacements > Lindemann limit.

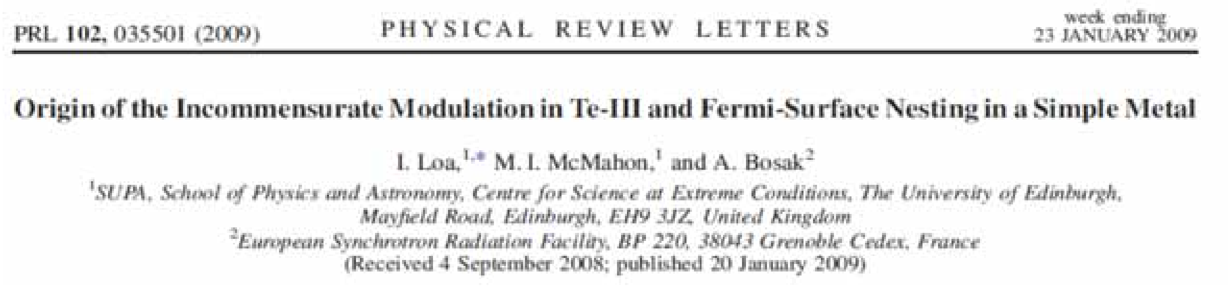
Determine phonon dispersion on approach to melting. Which modes go unstable, and where.



Calculated TDS from Si at AP

Can determine TDS pattern from known phonon spectrum – and are close to be able to do the reverse problem.

Lattice Dynamics

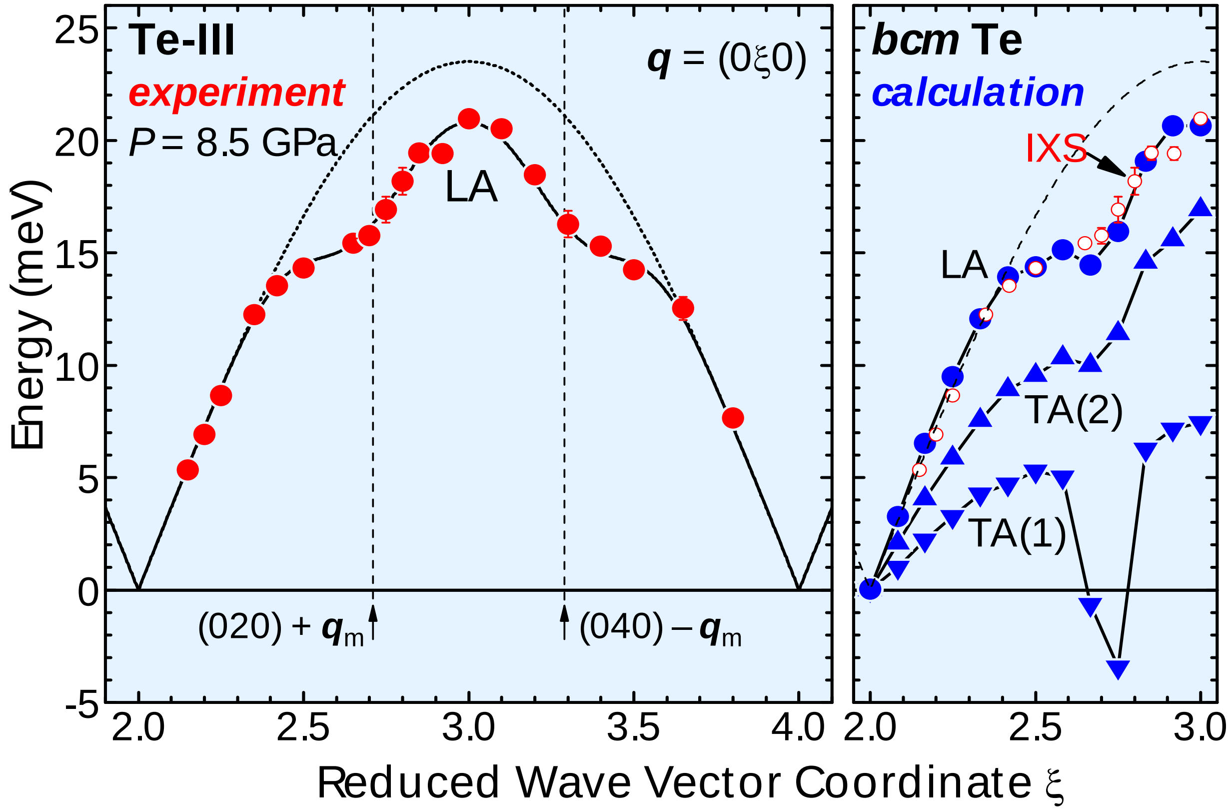


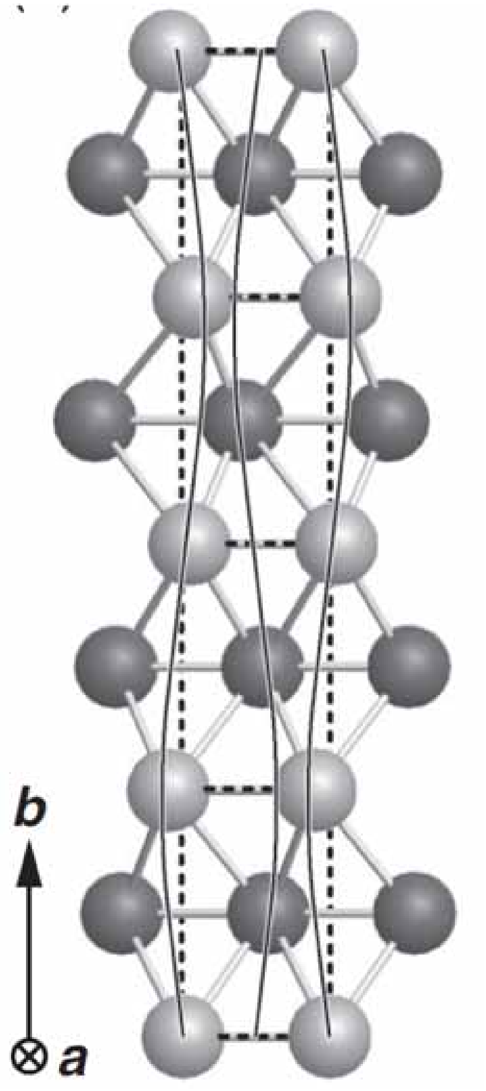
PRL 102, 035501 (2009) PHYSICAL REVIEW LETTERS week ending 23 JANUARY 2009

Origin of the Incommensurate Modulation in Te-III and Fermi-Surface Nesting in a Simple Metal

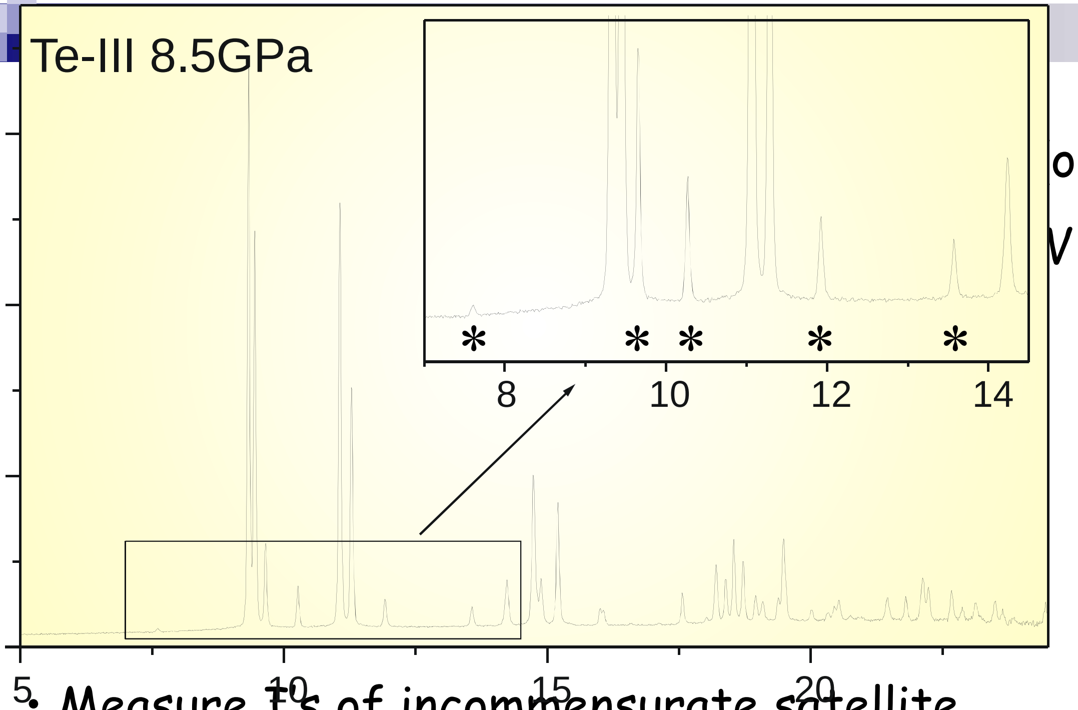
I. Loa,[1,*] M. I. McMahon,[1] and A. Bosak[2]
[1]SUPA, School of Physics and Astronomy, Centre for Science at Extreme Conditions, The University of Edinburgh, Mayfield Road, Edinburgh, EH9 3JZ, United Kingdom
[2]European Synchrotron Radiation Facility, BP 220, 38043 Grenoble Cedex, France

(Received 4 September 2008; published 20 January 2009)





Each data point takes ~hours



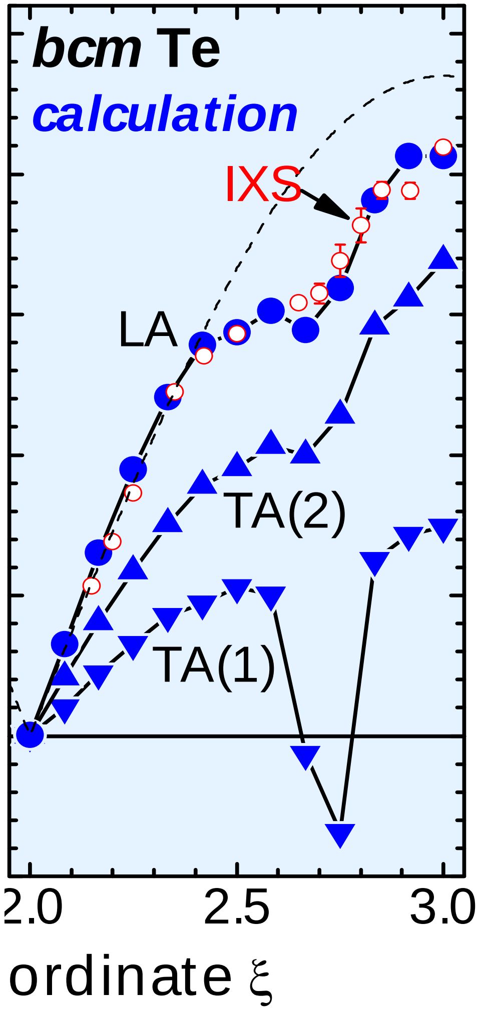
only) and “melt” the CDW

V “melts” and then

- Measure I’s of incommensurate satellite peaks as function of delay time between laser pulse and x-ray probe

- Might expect I’s → zero over hundred fs, and then I’s recover over few ps as soft-mode re-softens to zero, and CDW is re-established

- Can follow softening of the CDW mode by determining phonon branches as a function of time via time-resolved TDS measurements

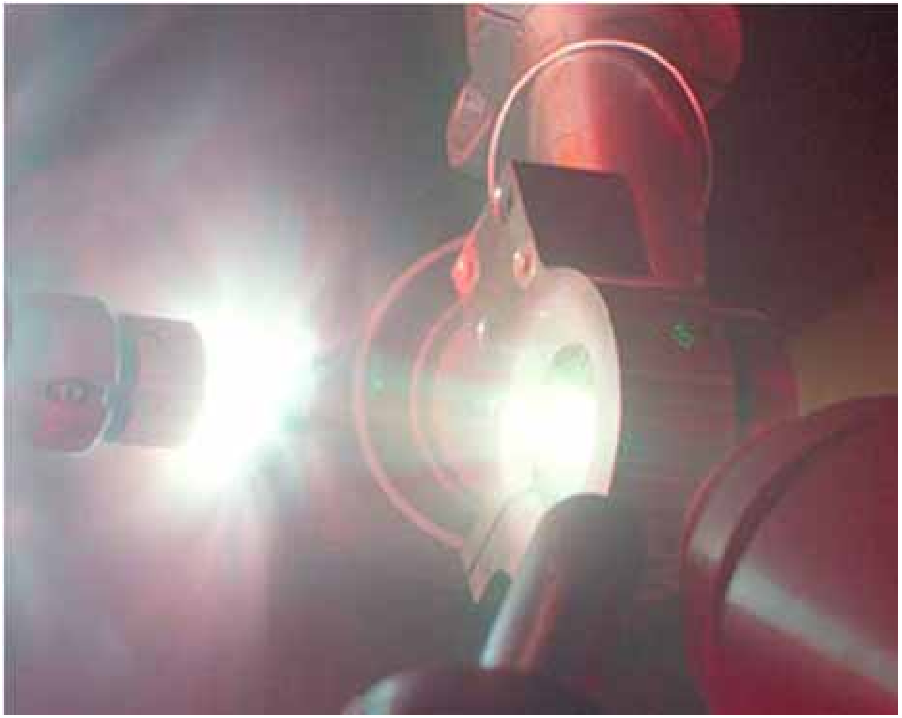


Diffraction from Dynamically-Compressed Matter

Upper P limit of ~400 GPa with DAC has not increased significantly in the last decade

The upper-T range available with laser heating has also remained relatively unchanged.

Dynamic compression can access truly extremes of P-Ts for nanoseconds, but structural studies impossible (to date).



Whole new areas of condensed matter physics, and new physical phenomena, lie tantalisingly out of reach, inaccessible to current techniques.

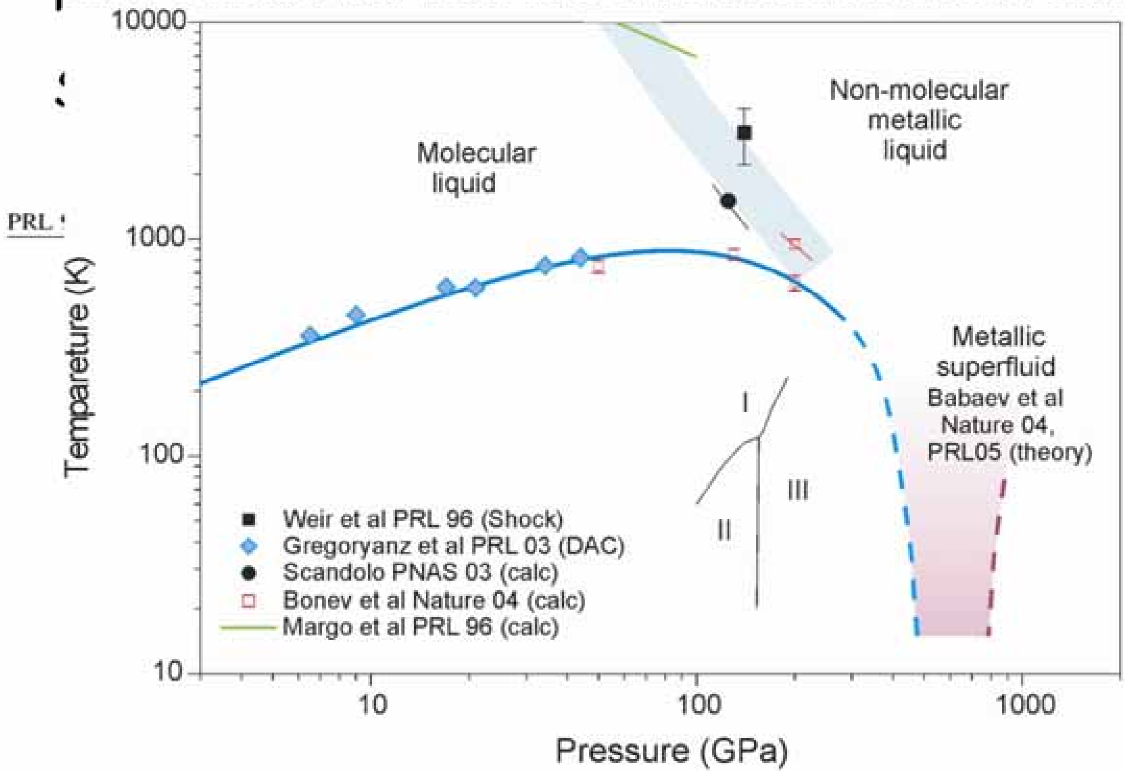
What Might We Want to Study?

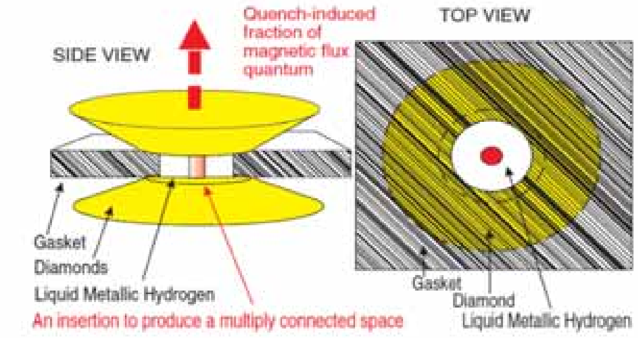
(Multi-)Terapascal behaviour of simple matter at ~300K (completely unknown)

(Multi-)Terapascal behaviour of simple matter to 11,000K (*almost* completely unknown)

New states of matter in hydrogen (and the study thereof)

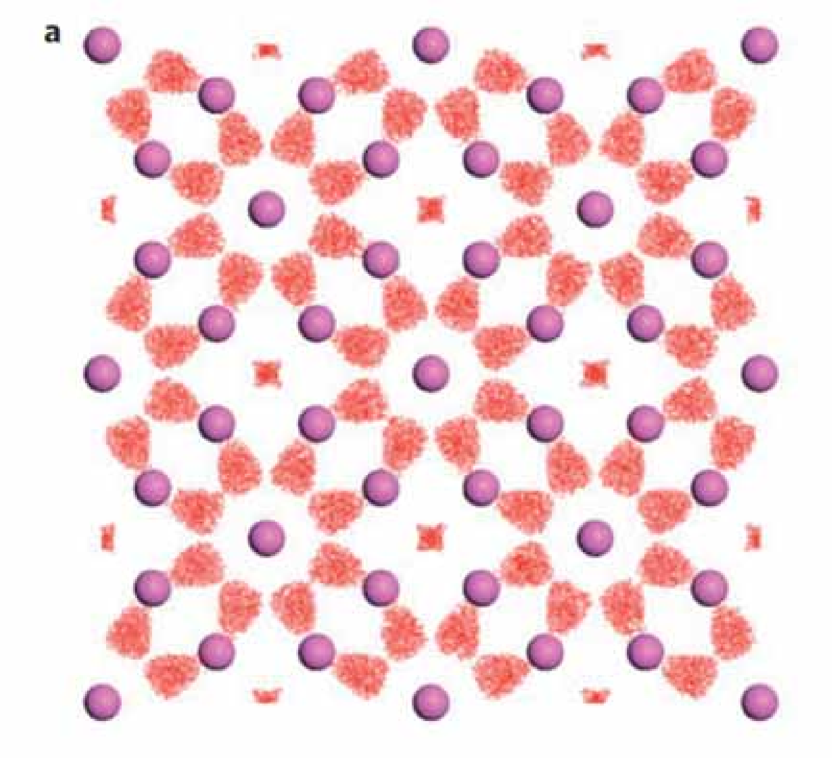
(~35,000K and 3 - 4.5 TPa)

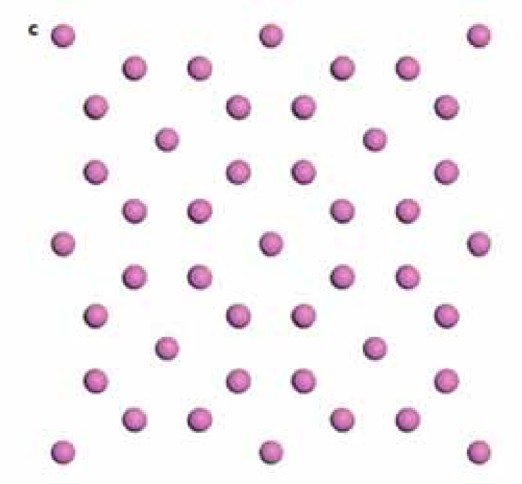




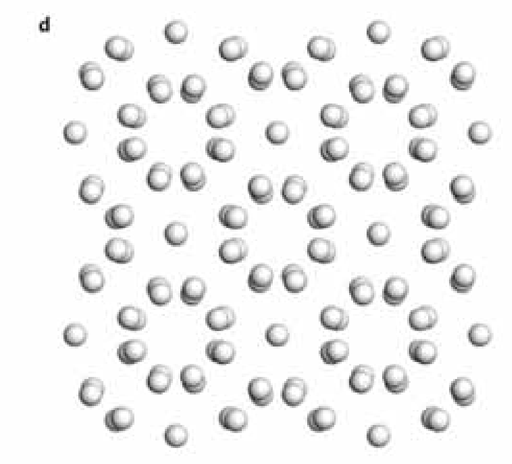
Further Complexity at Extreme Density
Al: A Simple Metal no More (Pickard & Needs, NatMat Jul2010)

Between 3.2 and 8.8 TPa (32-88 Mbars) Al is predicted to have an incommensurate H-G structure



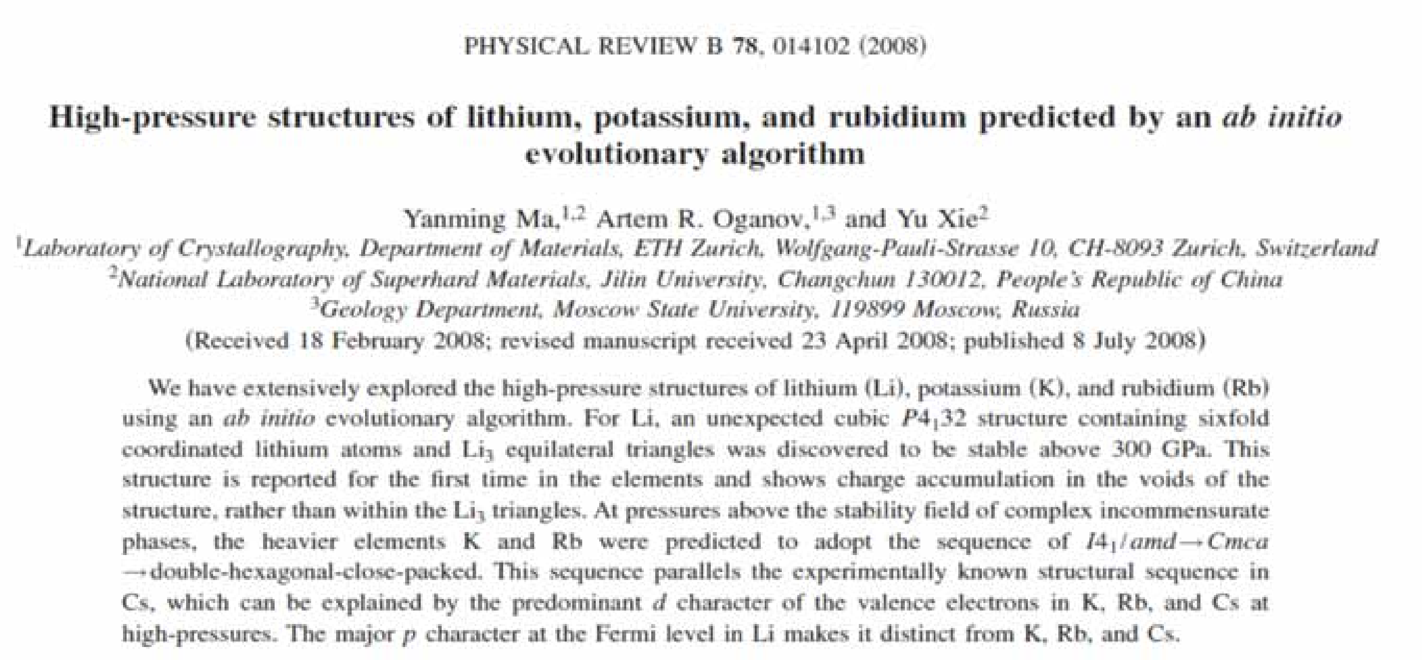


8-atom
H-G structure
(Ba-IV -type)



16-atom
H-G structure
(Rb-IV -type)

Further Complexity at Extreme Density
Li_3 units in Li stable 300-1000 GPa (Ma *et al*, PRB 2008)



PHYSICAL REVIEW B 78, 014102 (2008)

High-pressure structures of lithium, potassium, and rubidium predicted by an *ab initio* evolutionary algorithm

Yanming Ma,[1,2] Artem R. Oganov,[1,3] and Yu Xie[2]
[1] *Laboratory of Crystallography, Department of Materials, ETH Zurich, Wolfgang-Pauli-Strasse 10, CH-8093 Zurich, Switzerland*
[2] *National Laboratory of Superhard Materials, Jilin University, Changchun 130012, People's Republic of China*
[3] *Geology Department, Moscow State University, 119899 Moscow, Russia*

(Received 18 February 2008; revised manuscript received 23 April 2008; published 8 July 2008)

We have extensively explored the high-pressure structures of lithium (Li), potassium (K), and rubidium (Rb) using an *ab initio* evolutionary algorithm. For Li, an unexpected cubic $P4_132$ structure containing sixfold coordinated lithium atoms and Li_3 equilateral triangles was discovered to be stable above 300 GPa. This structure is reported for the first time in the elements and shows charge accumulation in the voids of the structure, rather than within the Li_3 triangles. At pressures above the stability field of complex incommensurate phases, the heavier elements K and Rb were predicted to adopt the sequence of $I4_1/amd \rightarrow Cmca \rightarrow$ double-hexagonal-close-packed. This sequence parallels the experimentally known structural sequence in Cs, which can be explained by the predominant d character of the valence electrons in K, Rb, and Cs at high-pressures. The major p character at the Fermi level in Li makes it distinct from K, Rb, and Cs.

Still metallic at these pressures

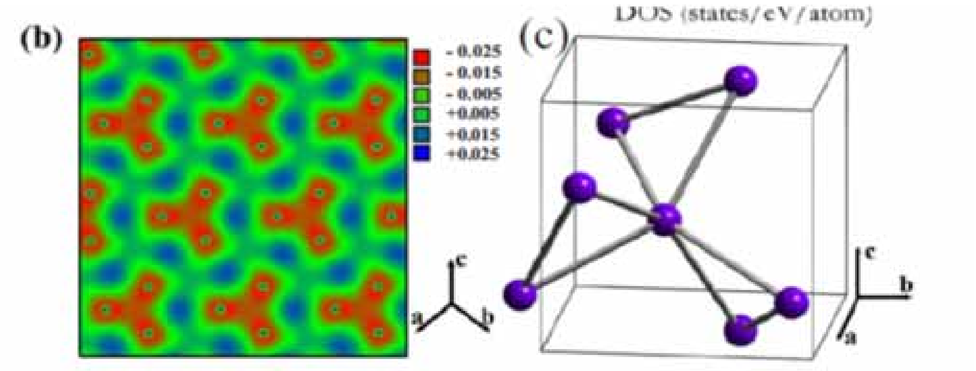
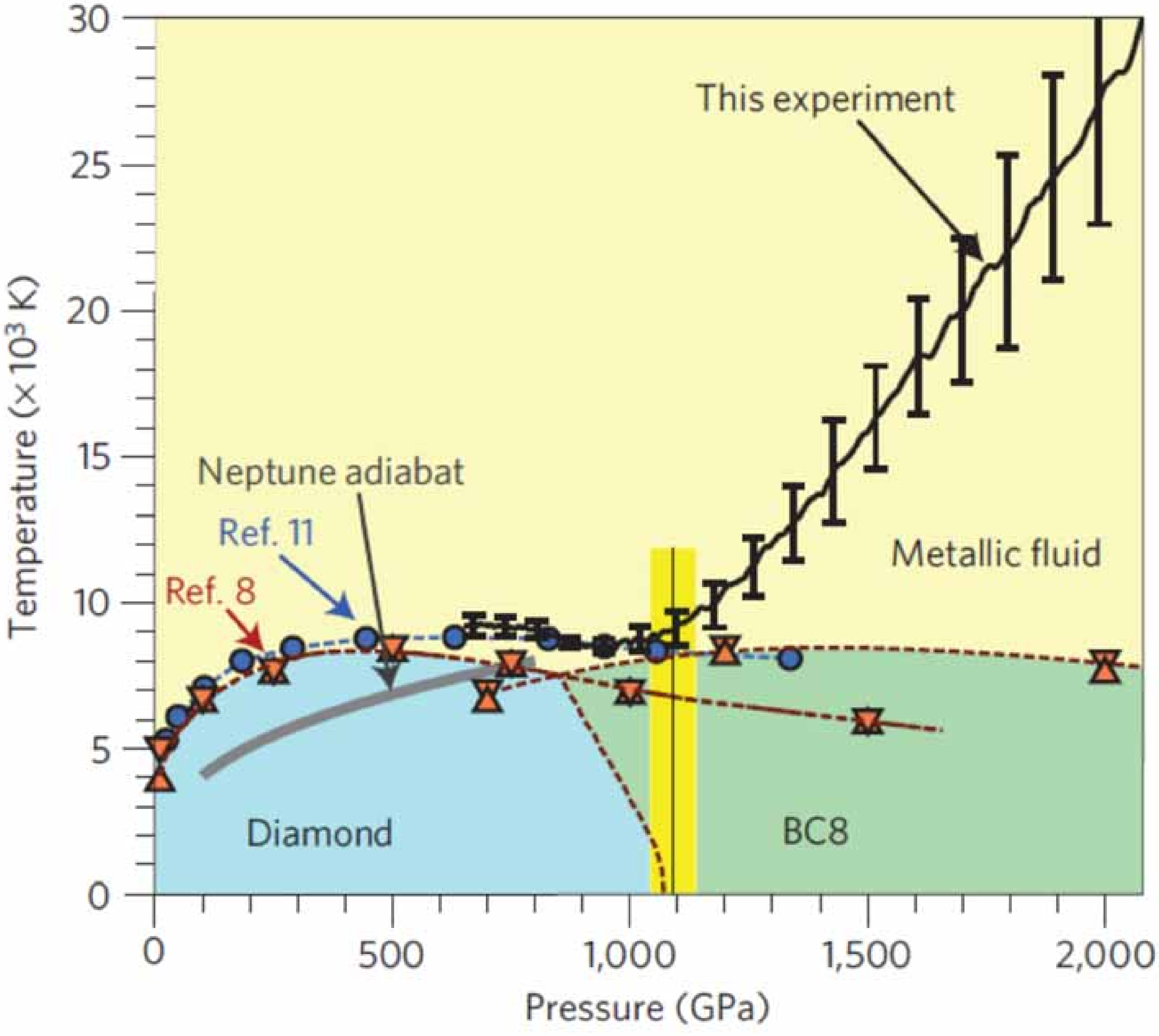
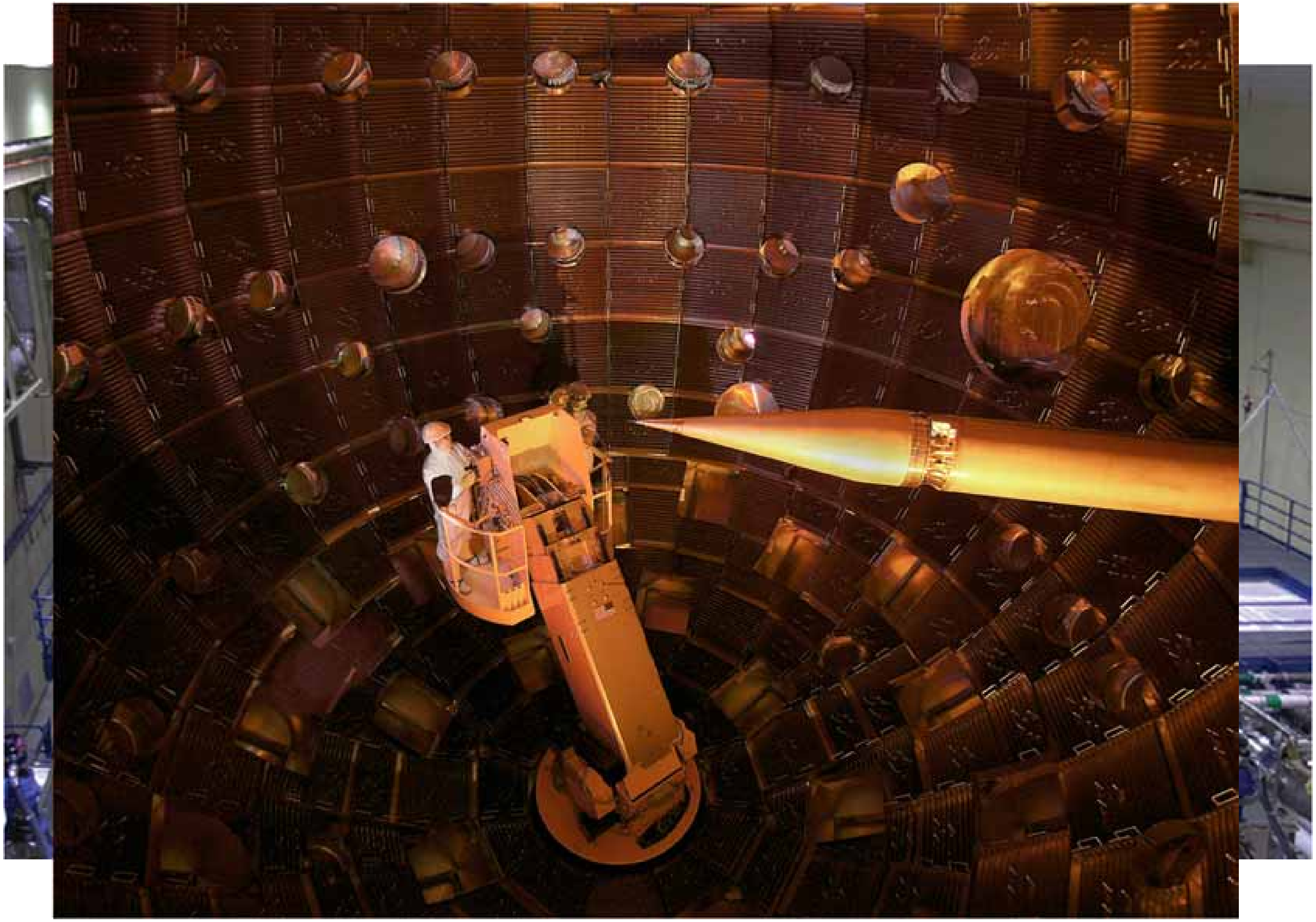


FIG. 2. (Color online) (a) The calculated band structure (left

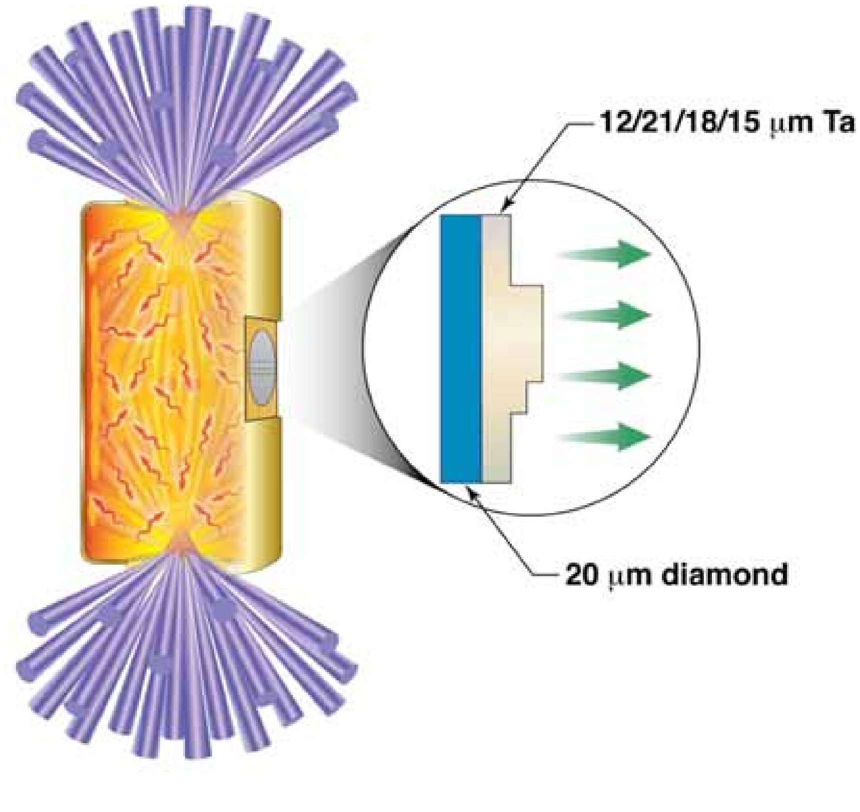

This experiment
Neptune adiabat
Ref. 11
Ref. 8
Metallic fluid
Diamond
BC8
Temperature (×10³ K)
30
25
20
15
10
5
0
500
1,000
1,500
2,000
Pressure (GPa)

NIF – the National Ignition Facility

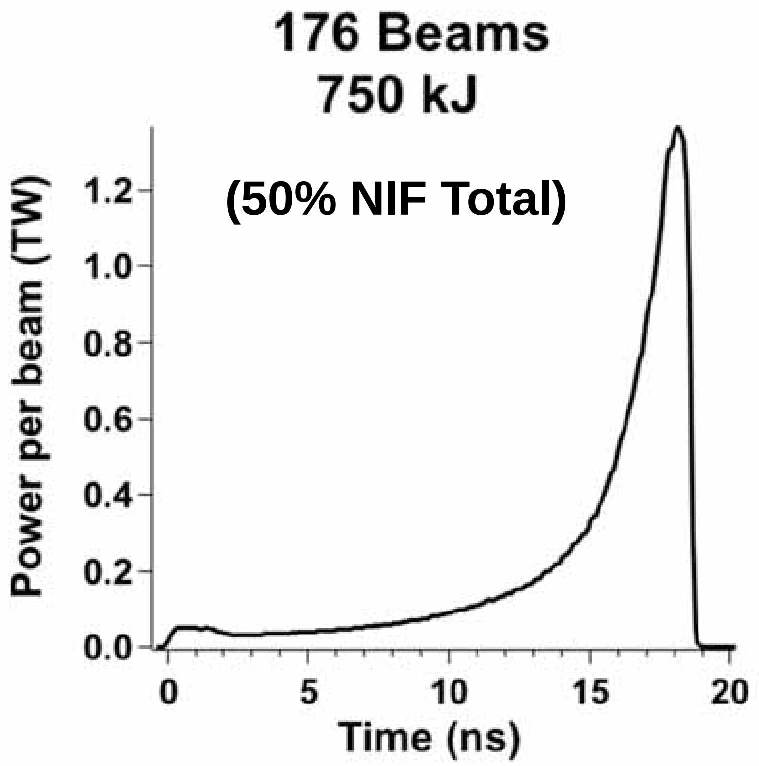


Aspiration: Ramp compression of hydrogen to ~50 $\mathrm{gcm^{-3}}$ (>4× density of lead)

Recent Results from NIF – courtesy Ray Smith (LLNL)



NIF-0310-18630


176 Beams
750 kJ

(50% NIF Total)

Conclusions

New Light Sources such as ERL and XFEL offer the possibility of entirely new extreme-conditions science

- Ability to study structure on a fs-ps timescale – and to higher P
- Ability to study dynamics on a fs-ps timescale – and to higher P
- Able to pump individual phonons of particular interest.
- Ability to study structures of dynamically compressed matter with same quality of data currently available from DACs

- BUT, comparatively little fast/ultrafast laser science at high-P done to date - so probably lots of interesting high-P phenomena still to be discovered.

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