High Pressure Inelastic X-ray Scattering of Hydrogen

Opportunity

- Study of elementary electronic excitations at low momentum transfer
- Spectroscopy at Mbar pressures, down to 4K, laser heating

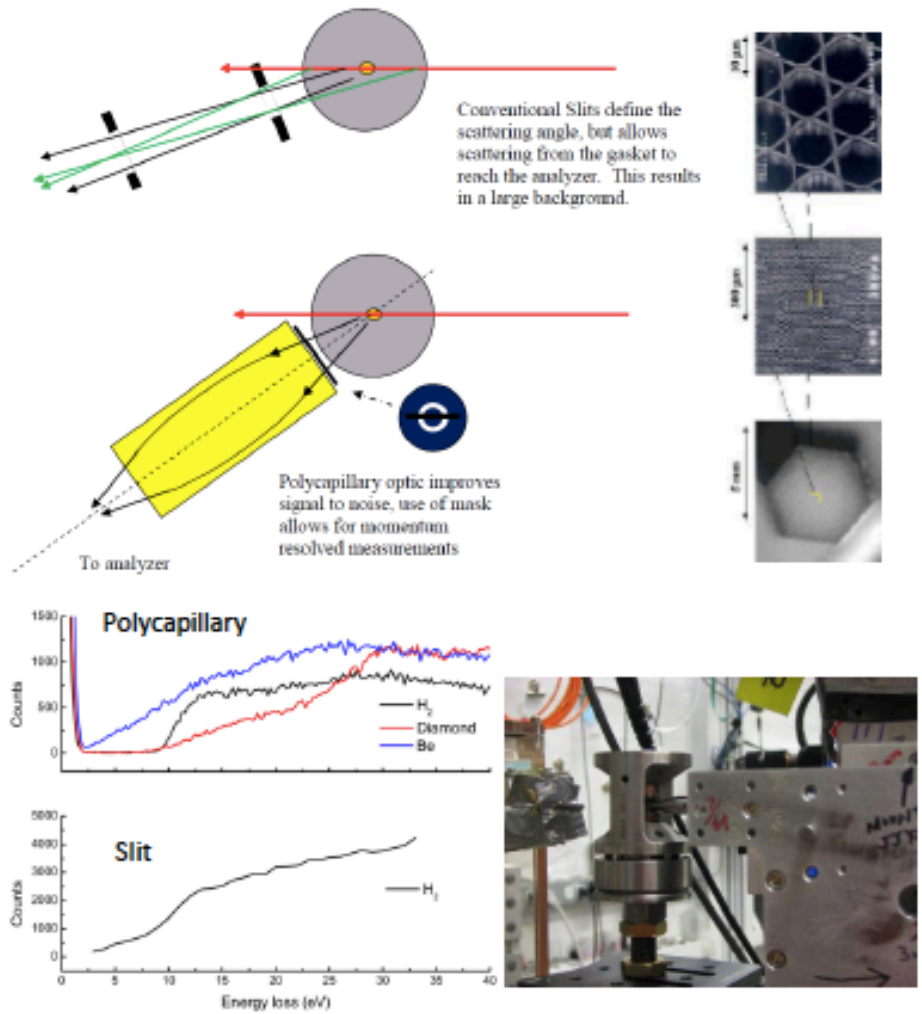
Challenge

- Diamond Anvil Cell development
- Minimize scattering from Diamond Avil Cell

4GSR Strength

- 10^{13} photons/sec at 10 keV in 1 µm Focused Beam

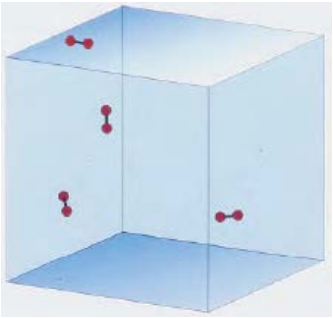
Excitation Spectrum of Hydrogen at High Pressure



D. Mao *et. al* unpublished

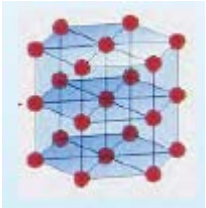
Paul Chow /HPCAT, Sector 16

Metallic Hydrogen
the holy grail of high pressure Science



25GPa?

Not yet



Reports

77k, 250 GPa nearly opaque

Optical Studies of Hydrogen Above 200 Gigapascals: Evidence for Metallization by Band Overlap

H. K. Mao and R. J. Hemley

Direct optical observations of solid hydrogen to pressures in the 250-gigapascal (2.5-megabar) range at 77 K are reported. Hydrogen samples appear nearly opaque at the maximum pressures. Measurements of absorption and Raman spectra provide evidence that electronic excitations in the visible region begin at ~200 gigapascals. The optical data are consistent with a band-overlap mechanism of metallization.

Solid hydrogen at 342 GPa: no evidence for an alkali metal

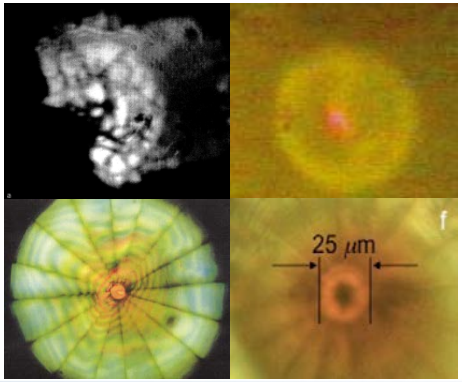
Chandrabhas Narayana*, Huan Luo*†, Jon Orloff‡ & Arthur L. Ruoff*

* Department of Materials Science and Engineering, Cornell University, Ithaca, New York 14853, USA
‡ Institute for Plasma Research, University of Maryland, College Park, Maryland 20742-3511, USA

Optical studies of solid hydrogen to 320 GPa and evidence for black hydrogen

Paul Loubeyre*, Florent Occelli* & René LeToullec*†

* Département Physique Théorique et Applications, Commissariat à l'Energie Atomique, 91680, Bruyères-le-Châtel, France
† Université Paris 6, PMC, 4 place Jussieu, 75252 Paris, France



5402 (2012) PHYSICAL REVIEW LETTERS week ending 6 APRIL 2012

Synchrotron Infrared Measurements of Dense Hydrogen to 360 GPa

Chang-Sheng Zha, Zhenxian Liu, and Russell J. Hemley

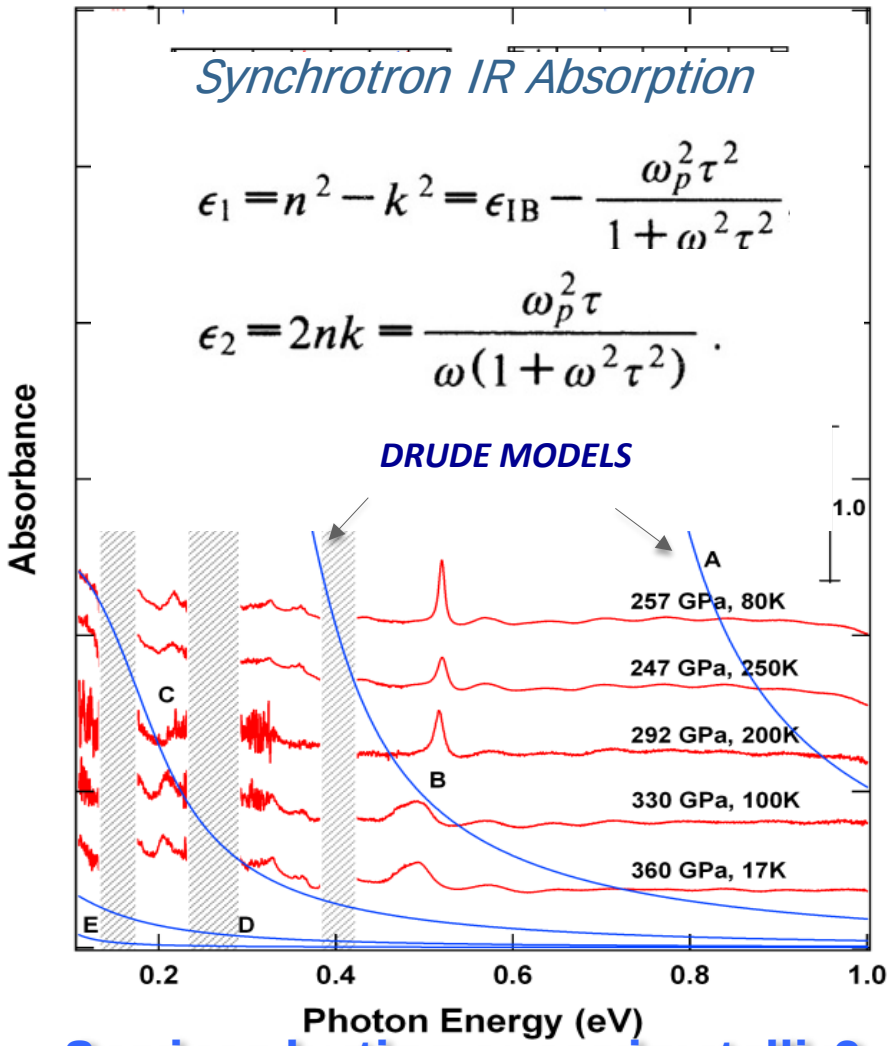
al Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Road NW, Washington D.C. 20015, USA

(Received 26 December 2011; published 3 April 2012)

Diamond-anvil-cell techniques have been developed to confine and measure hydrogen samples under static conditions to pressures above 300 GPa from 12 to 300 K using synchrotron infrared and optical absorption techniques. A decreasing absorption threshold in the visible spectrum is observed, but the material remains transparent at photon energies down to 0.1 eV at pressures to 360 GPa over a broad temperature range. The persistence of the strong infrared absorption of the vibron characteristic of phase III indicates the stability of the paired state of hydrogen. There is no evidence for the predicted metallic state over these conditions, in contrast to recent reports, but electronic properties consistent with semimetallic behavior are observed.

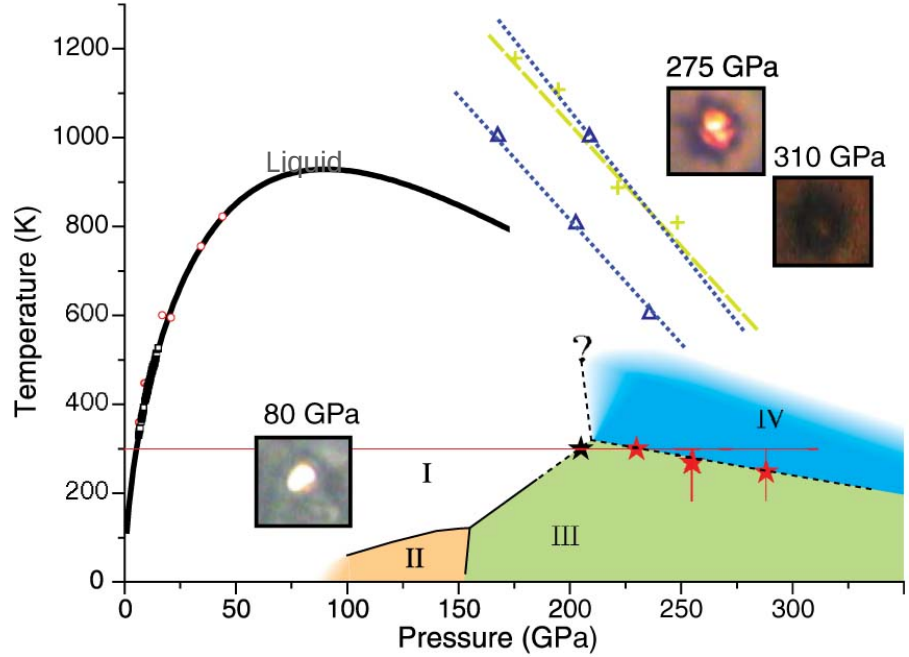
Metallization at 450 GPa? Electronic structures?

Persistence of Molecular States of Hydrogen



Semiconducting or semimetallic?

[Zha *et al., Phys. Rev. Lett.*, **108,** 125501 (2012)查长生



Evidence for a new phase

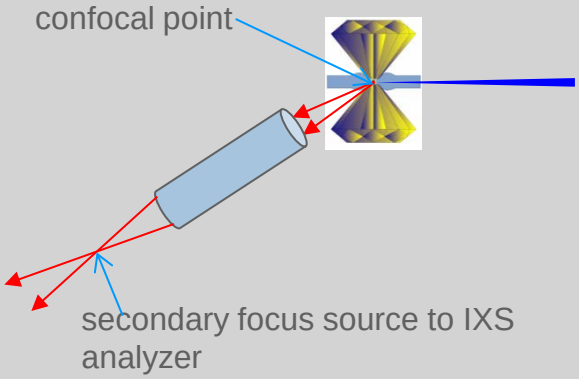
[Howie *et al., Phys. Rev. Lett.* **108,** 125501 (2012)]

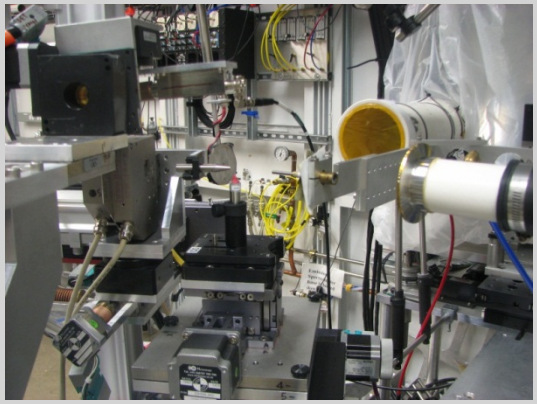
- **Continued challenge**
- **EFree experiment / theory synergy**

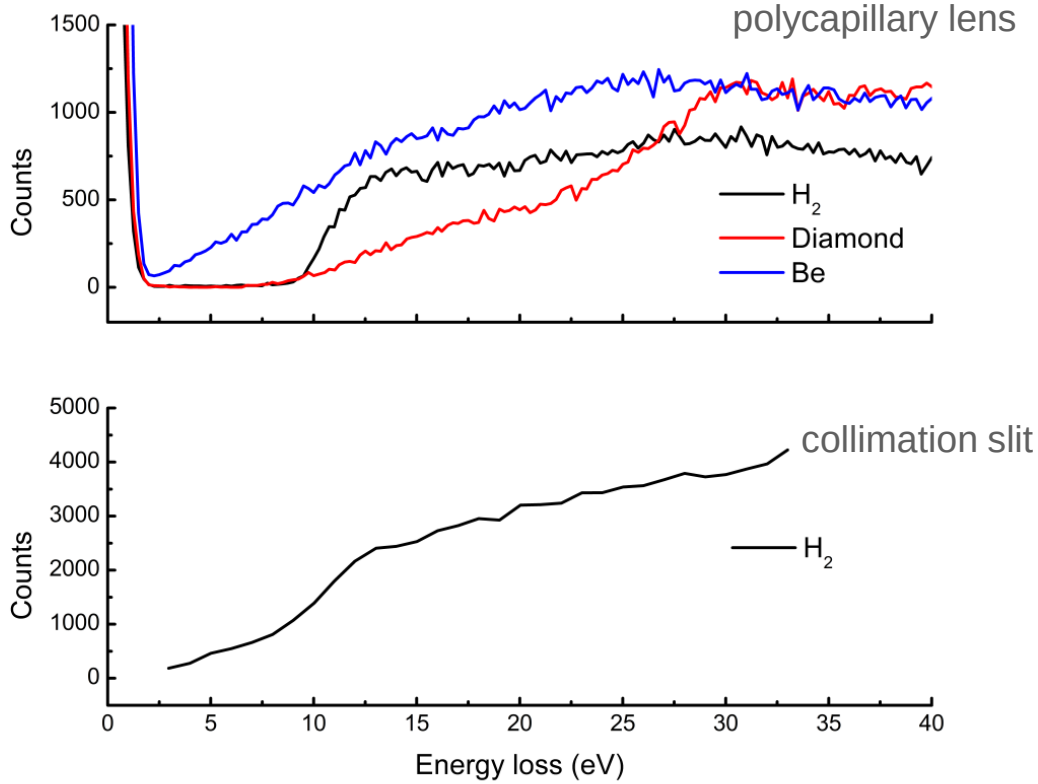
µm size sample at 300 GPa

Inelastic X-ray Scattering with Polycapillary Lens

- Enabled new measurements of band structures of H_2 which was not feasible with the conventional slit collimation
- Improved the collimation and suppressed the background signal by an order of magnitude
- Direct measurement of H2 ionization energy, excitons, and electronic band gap to 50 GPa.
- Higher pressures need sub-µm x-ray and intensity of MBA!!







Yuming Xiao, Paul Chow, Yang Ding, Junyue Wang, Dave Mao, to be published

Electronic IXS of Light Elements at High Pressures

Opportunity

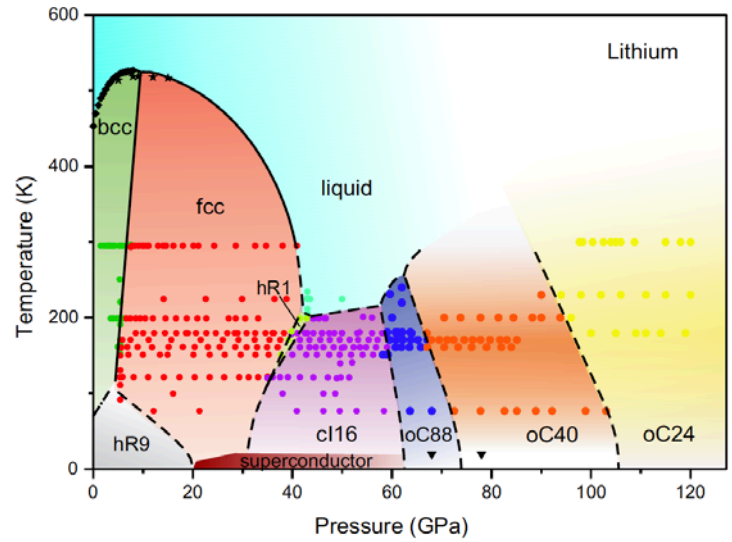
- Plasmons of high-*P* insulating Li and Na.
- Pressure-induced π to σ bonding transitions in B, C, and N.
- Oxygen bonding change in high-*P* symmetric ice and superconducting oxygen.

Challenge

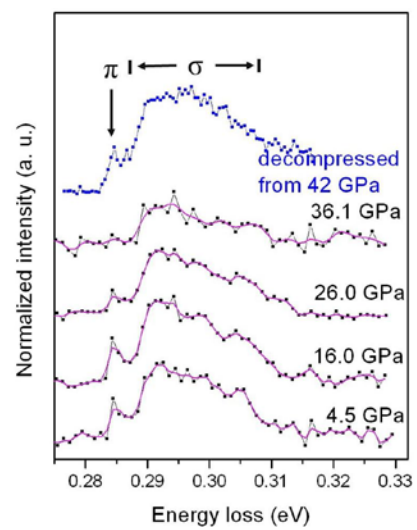
- Weak IXS signals from µm-size samples at multimegabar pressures.
- High background in DAC.

4GSR Strength:

- The two of magnitude higher brilliance at MBA APS will be essential for improving the signal. The submicron focusing capability allows probing the sample while avoiding the DAC backgrounds.



Phase diagram of Li. *Guilaume et al. Nature Physics 2011*



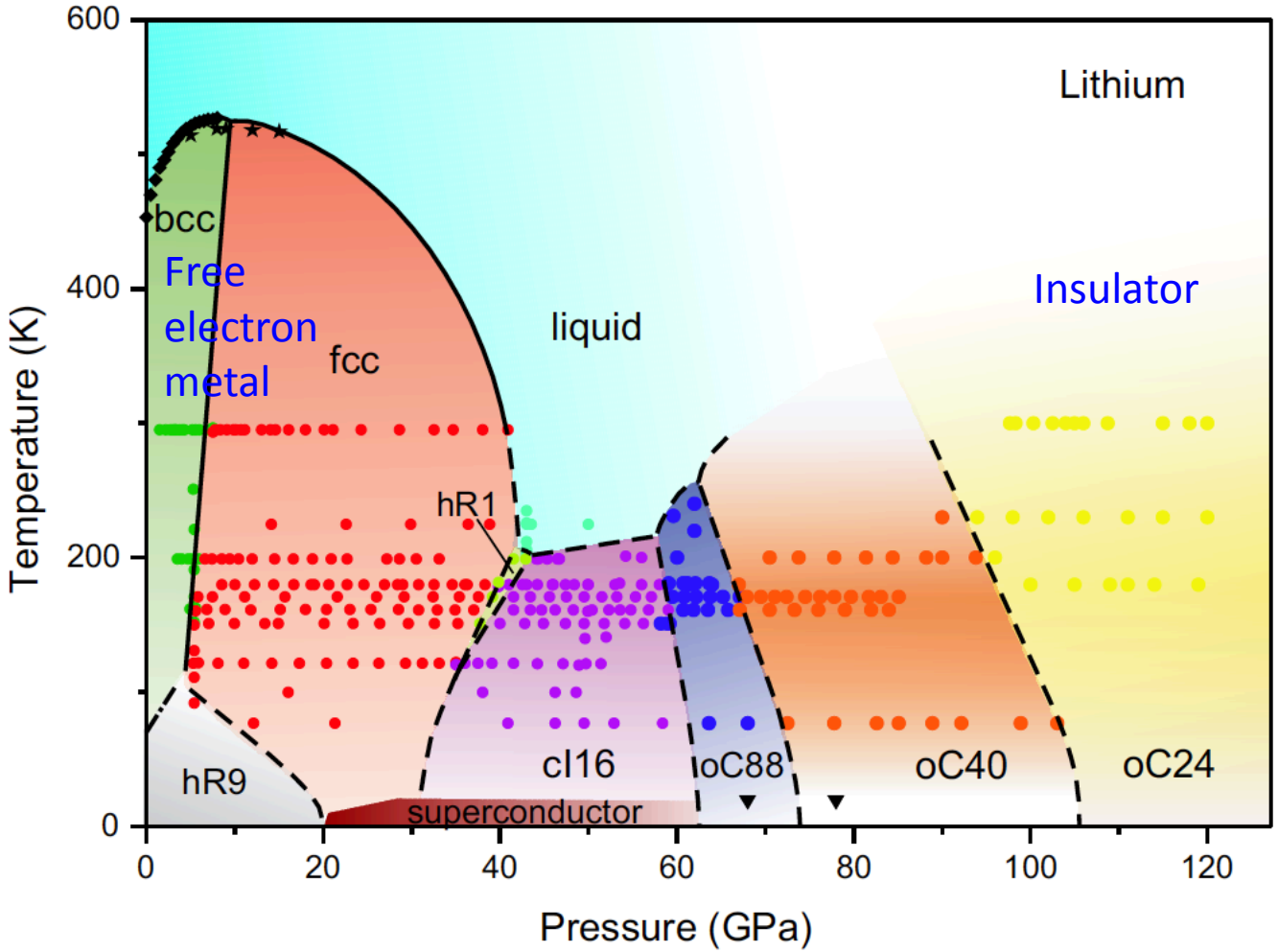
Carbon K-edge X-ray Raman spectra

Long-range ordered with amorphous carbon building blocks.

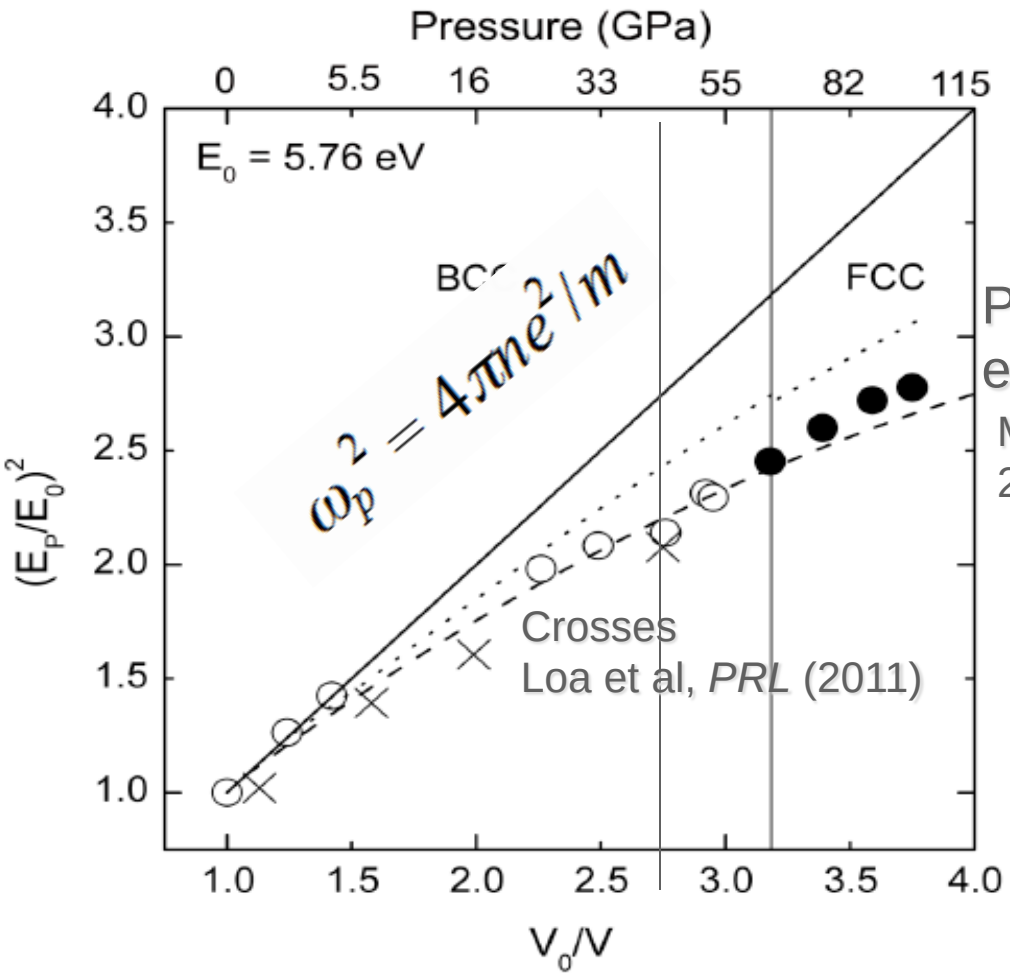
Wang et al. Science 2012

Cold melting and multiple phases of dense lithium

Christophe L. Guillaume, Eugene Gregoryanz, Olga Degtyareva, Malcolm I. McMahon, Shaun Evans, Michael Hanfland, Malcolm Guthrie, Stas V. Sinogeikin, & H-K. Mao, *Nature Physics 7, 211* (2011)



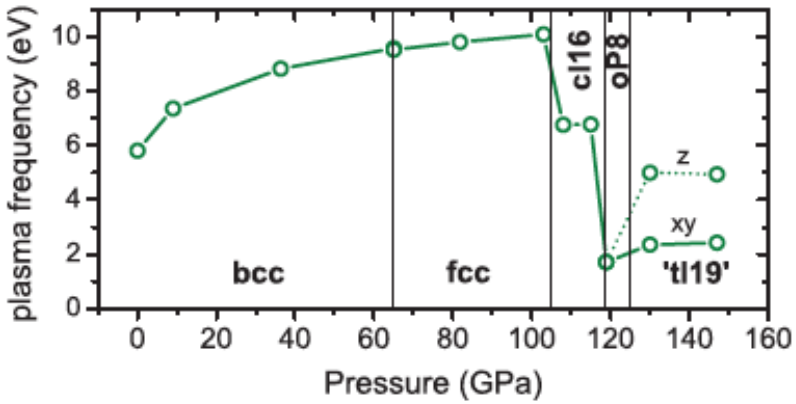
Studying plasmon and Li K-edge IXS for understanding the metal-insulator transition



Present experiments

Mao et al, *PNAS 108*, 20434 (2011)

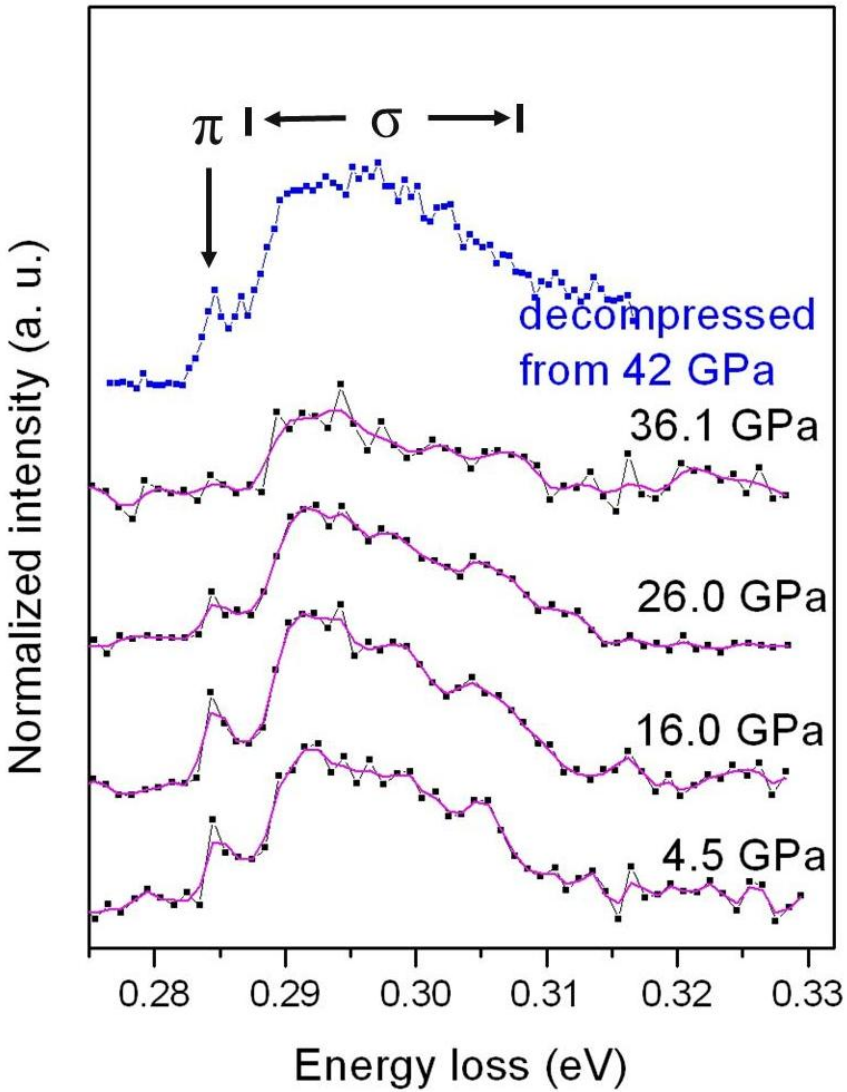
Na plasmon



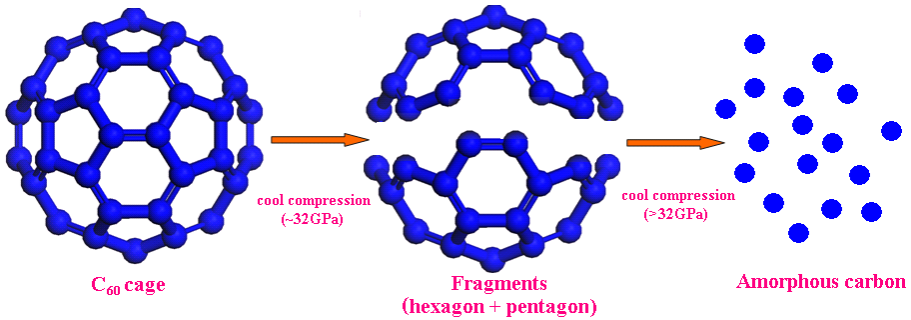
Calculated Na plasmon

Lazacki *et al*. *PNAS* ***106*** 6525 (2009)

Pressure crushed individual C_{60} but preserved the arrangement



X-ray Raman



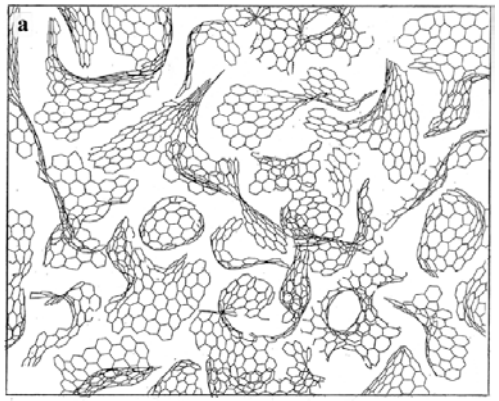
Wang et al., *Science* ***337***, 825 2012

Understanding the pressure-induced π–σ bonding change by studying X-ray Raman of carbon K-edge

PRL 107, 175504 (2011) PHYSICAL REVIEW LETTERS week ending 21 OCTOBER 2011

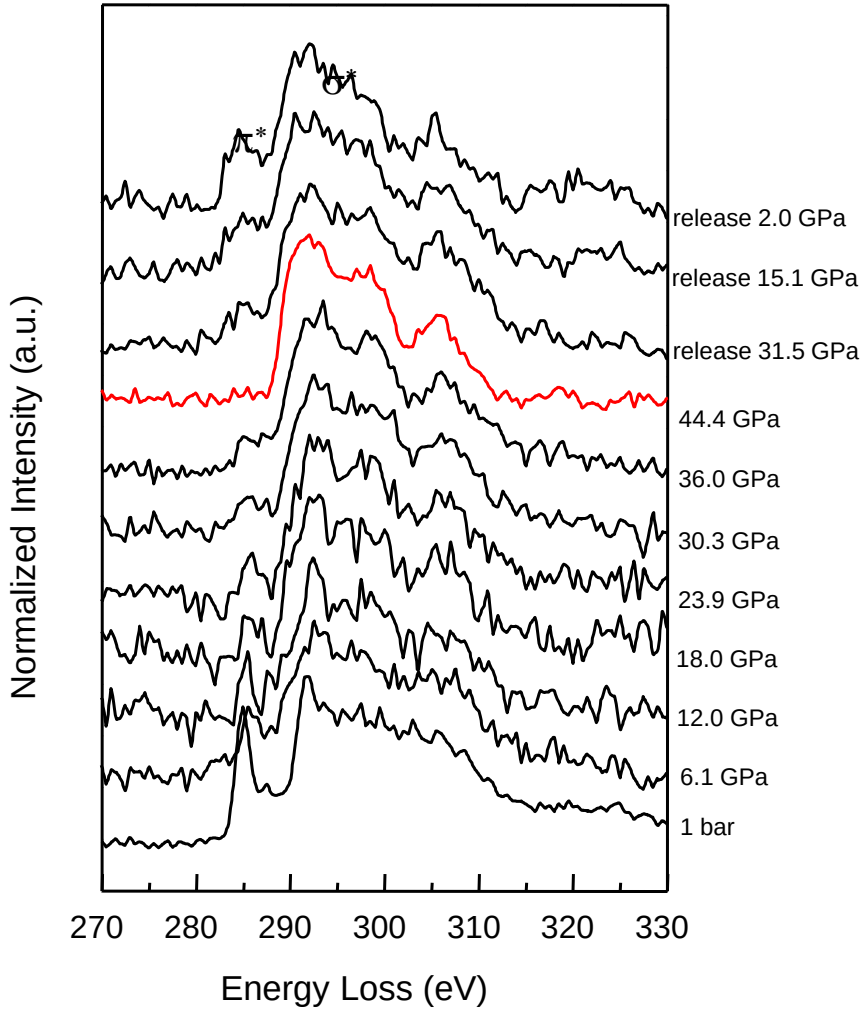
Amorphous Diamond: A High-Pressure Superhard Carbon Allotrope

Yu Lin,[1,*] Li Zhang,[2] Ho-kwang Mao,[2] Paul Chow,[3] Yuming Xiao,[3] Maria Baldini,[4] Jinfu Shu,[2] and Wendy L. Mao[1,5]



Harris, *Philos. Mag. 2004*

Xray Raman spectroscopy of the carbon K-edge showing the transition of sp^2-sp^3 bonding



U.S. DEPARTMENT OF ENERGY

High-Resolution IXS of Iron at Core *P-T* Conditions

Opportunity

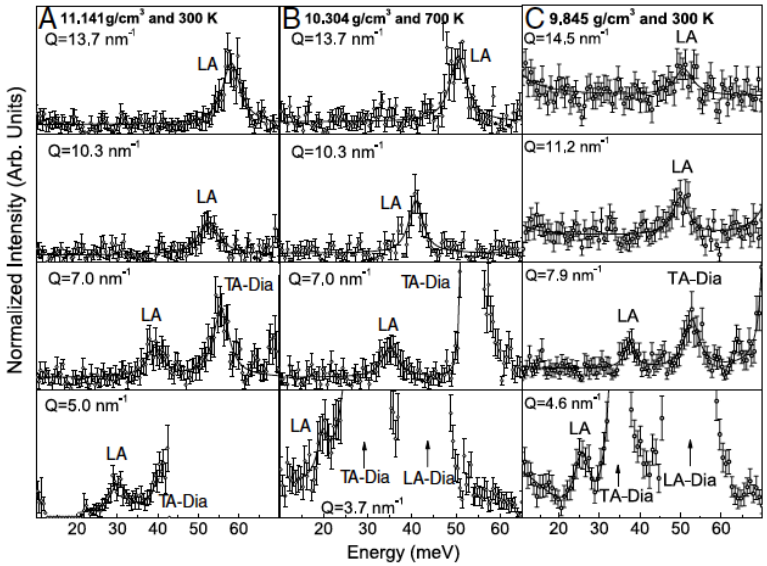
- Experimental measurements of acoustic velocity of iron at the high *P-T* conditions of the Earth's core.
- Direct constraints for deep Earth models

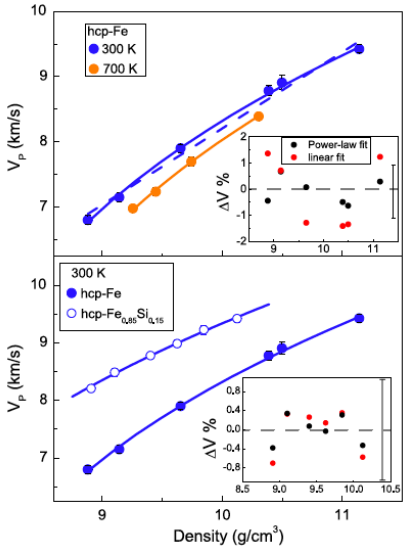
Challenge

- High-resolution IXS is count-limited.
- Simultaneous high P-T experiments require excellent coupling of the heating-laser and IXS probe.

4GSR Strength:

- The two of magnitude higher brilliance and submicron focusing capability at MBA APS will be essential for meeting the above challenges.





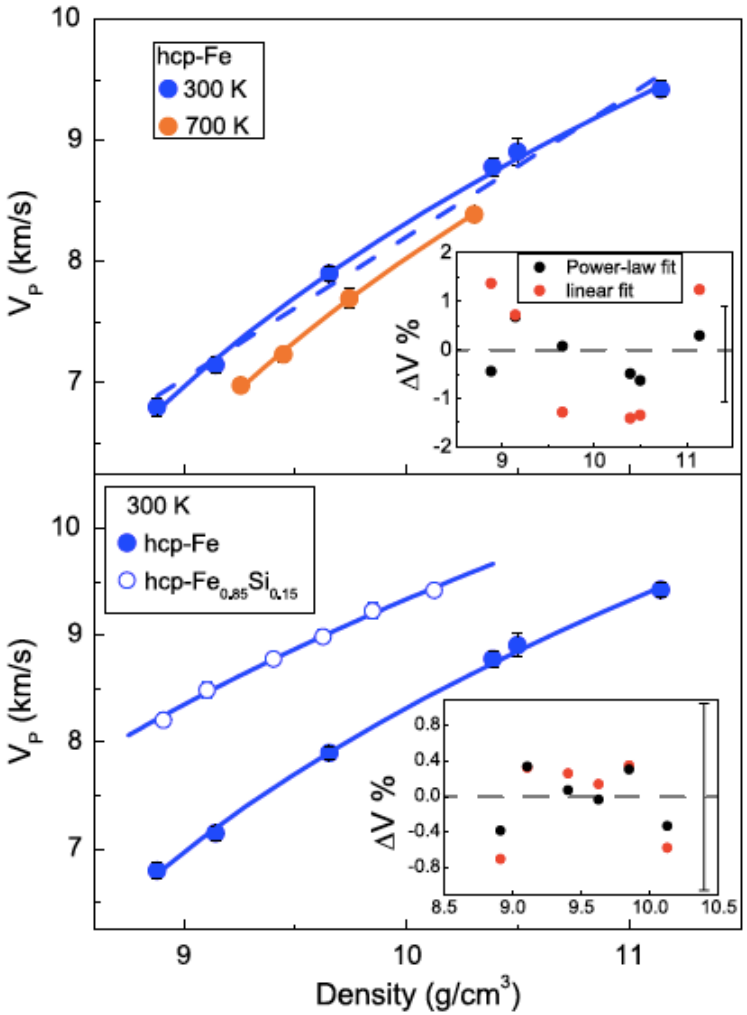
Preliminary sound velocity data of Fe and Fe-Si alloy have been obtained by HERIX measurements to 98 GPa and 700 K, and extrapolated to 360 GPa and 6000 K for interpretation of the inner core seismicity

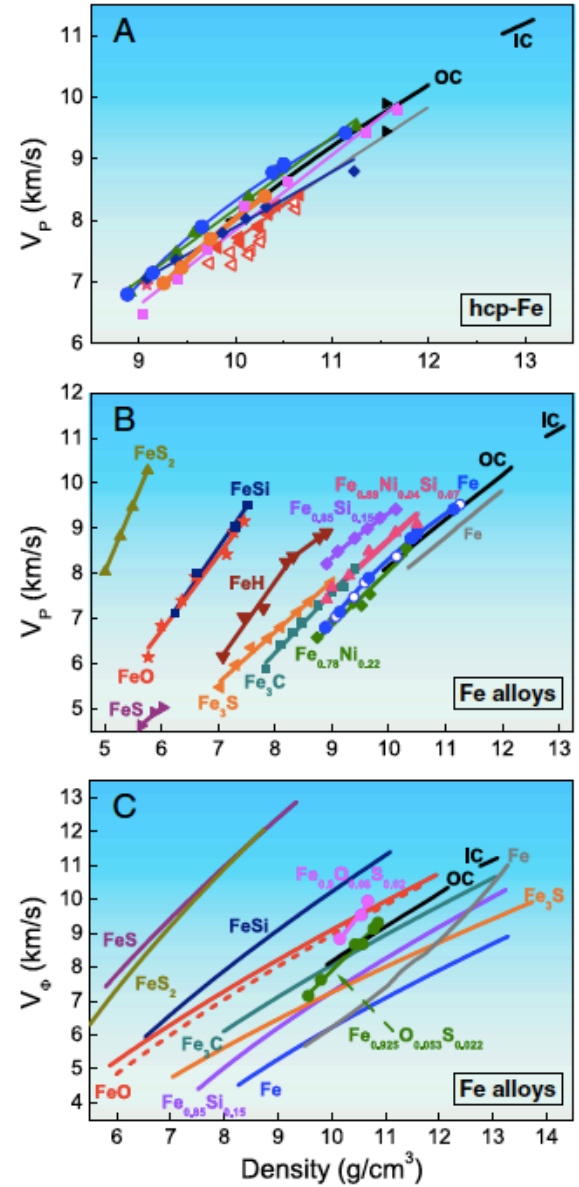
Z Mao et al. PNAS 2012

Sound velocities of Fe and Fe-Si alloy in the Earth's core

Zhu Mao[a,1], Jung-Fu Lin[a], Jin Liu[a], Ahmet Alatas[b], Lili Gao[b], Jiyong Zhao[b], and Ho-Kwang Mao[a]

PNAS | June 26, 2012 | vol. 109 | no. 26 | 10239–10244





High-pressure breakthroughs need MBA

1. Metallization of hydrogen.
2. Metal-insulator transition of Na and Li.
3. Pressure-induced π–σ bonding transition of B, C, N, O
4. Phonon and elasticity of high *P-T* Fe.