Transitions in Hydrogen Under Pressure

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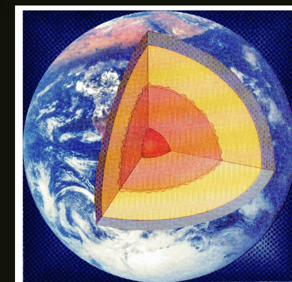
Pennsylvania State University, Apr. 7, 2016

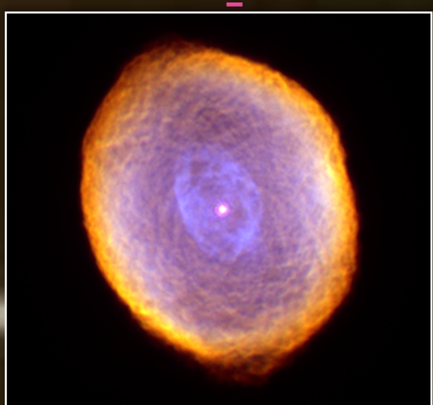
PRESSURES IN THE VISIBLE UNIVERSE

Hydrogen gas in intergalactic space 10^{-32} atm

Center of Jupiter - 8×10^{7} atm



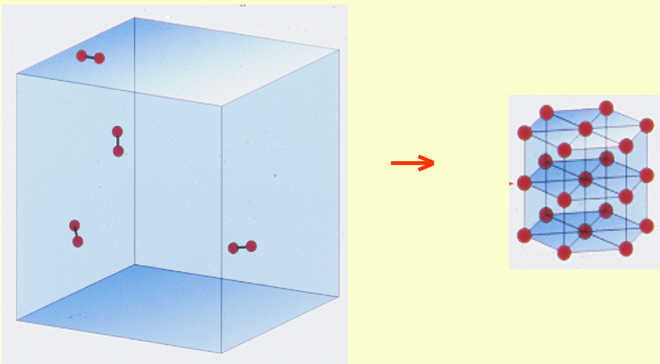
Center of Earth 3.6×10^{6} atm.

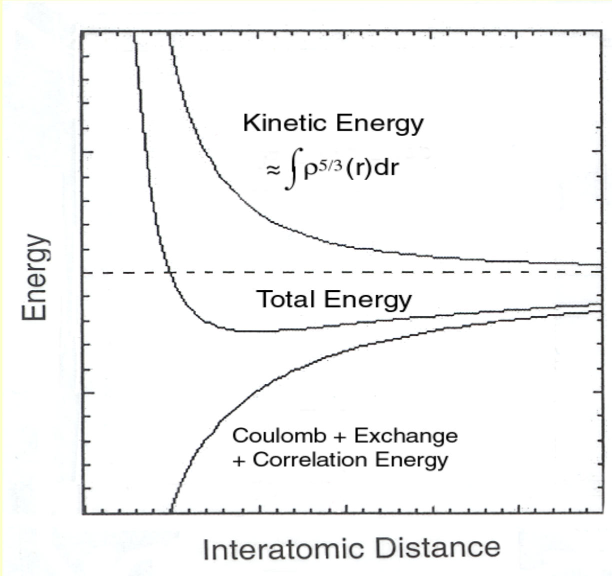


Center of Neutron Star - 10^{28} atm

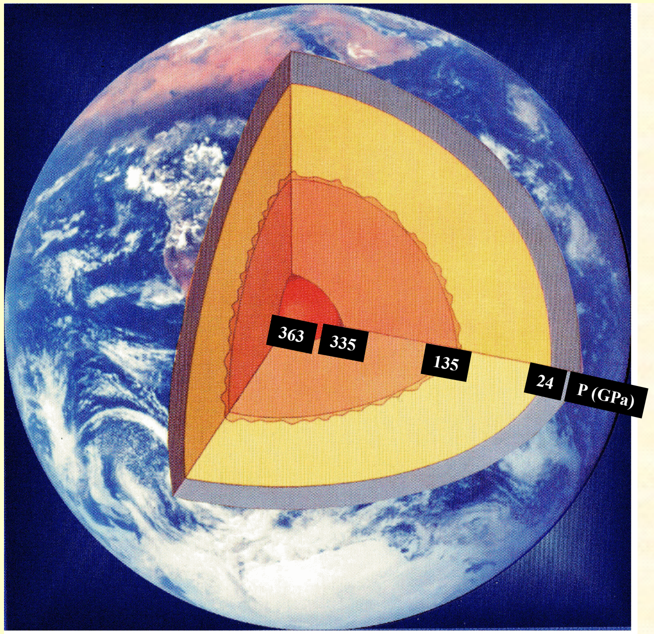
10^{3} atm ≈ kbar
10^{6} atm ≈ Mbar
10 kbar = 1 GPa
1 Mbar = 100 GPa

Effects of Extreme Pressures on Molecules



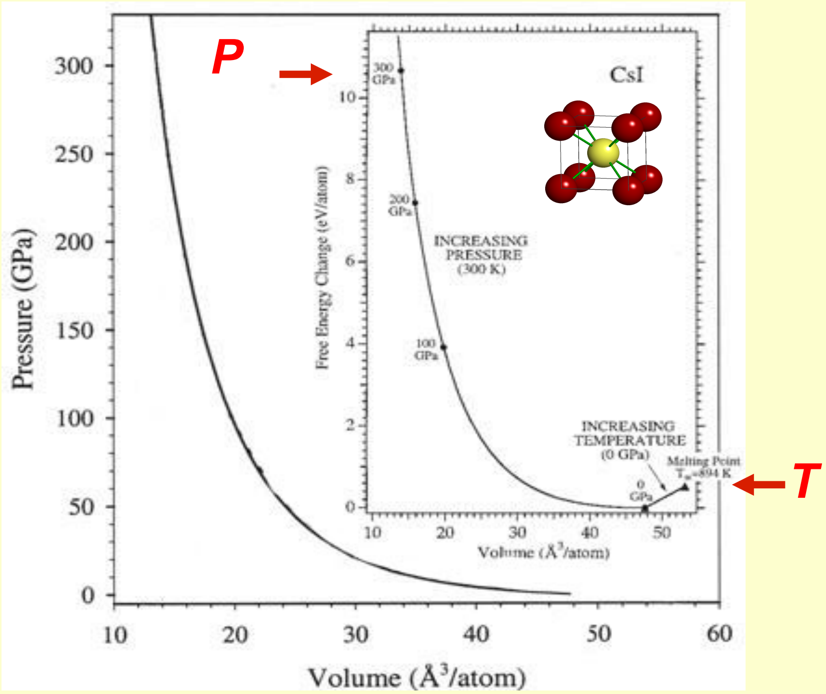


$$P = -\frac{\partial E}{\partial V}$$



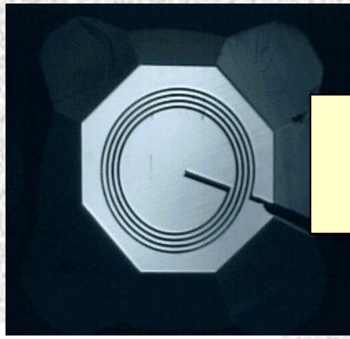
$P\Delta V \sim eV$

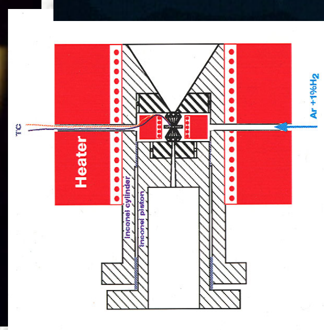
chemical bond strengths

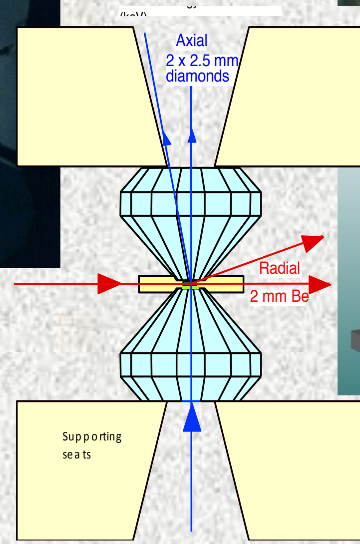


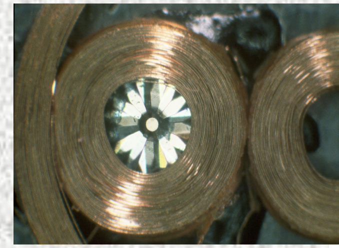
High-Pressure Technology:

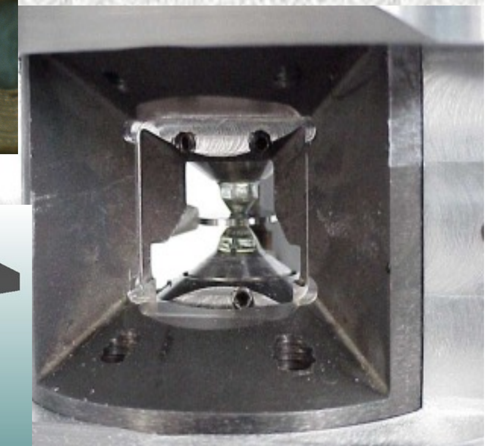
STATIC COMPRESSION

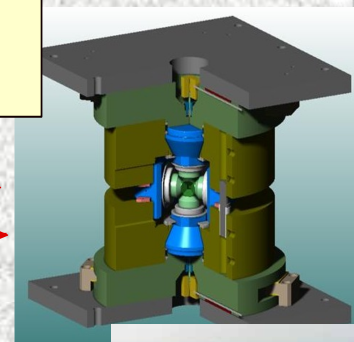


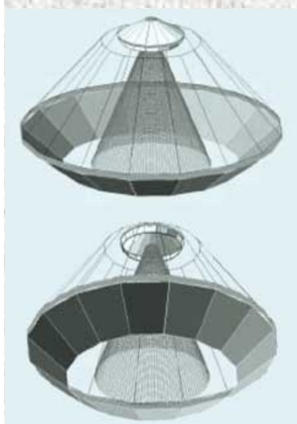




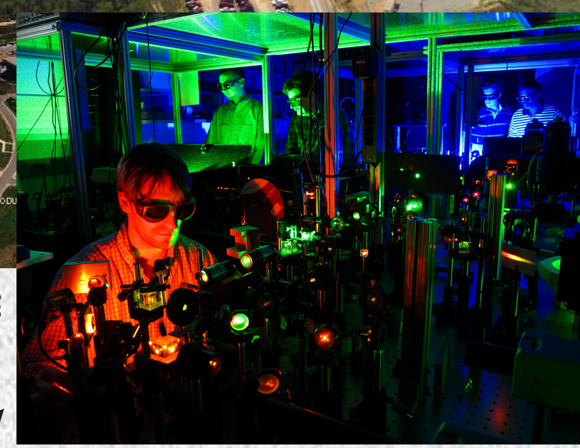






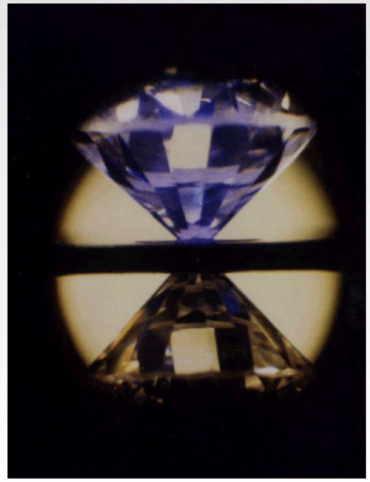


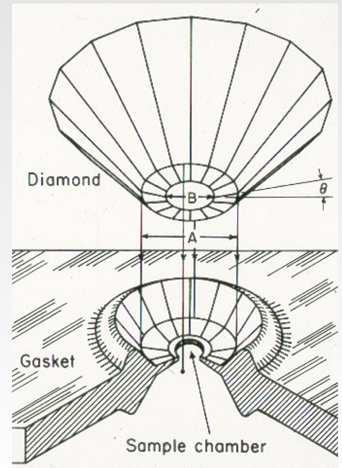
200mm

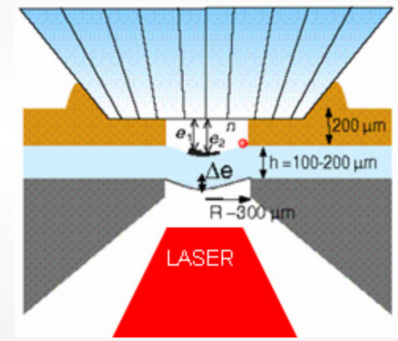


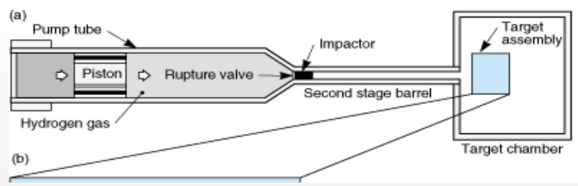
High-Pressure Technology:
STATIC AND DYNAMIC COMPRESSION

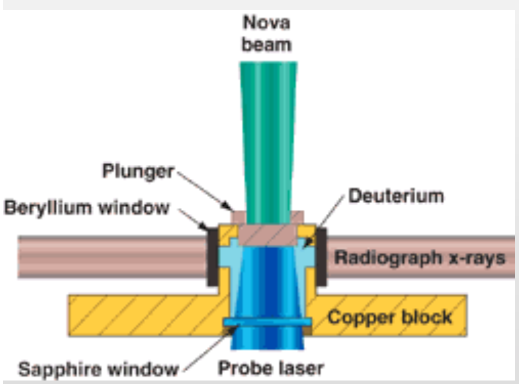
- **Static compression**
  - **Low temperature**
  - **High temperature**
- **Dynamic compression**
  - **Shock-wave**
  - **Isentropic**
- **Combined static/dynamic**

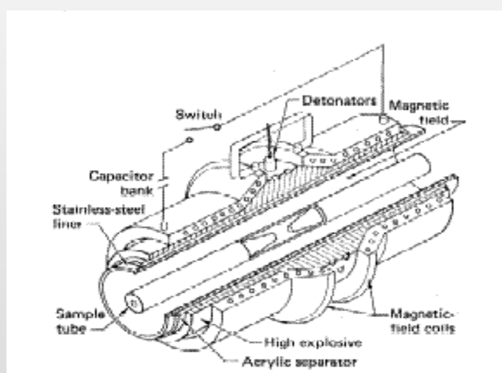


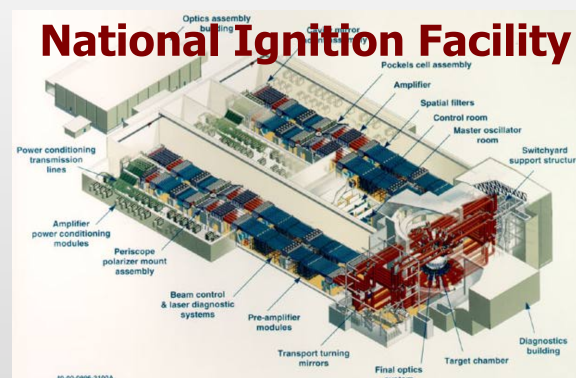












I. Introduction

II. Isolated Molecule/Zero Pressure

III. Hydrogen under Pressure

IV. New Phases

V. High Pressures and Temperatures

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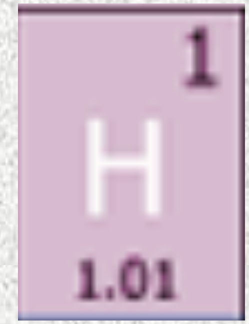
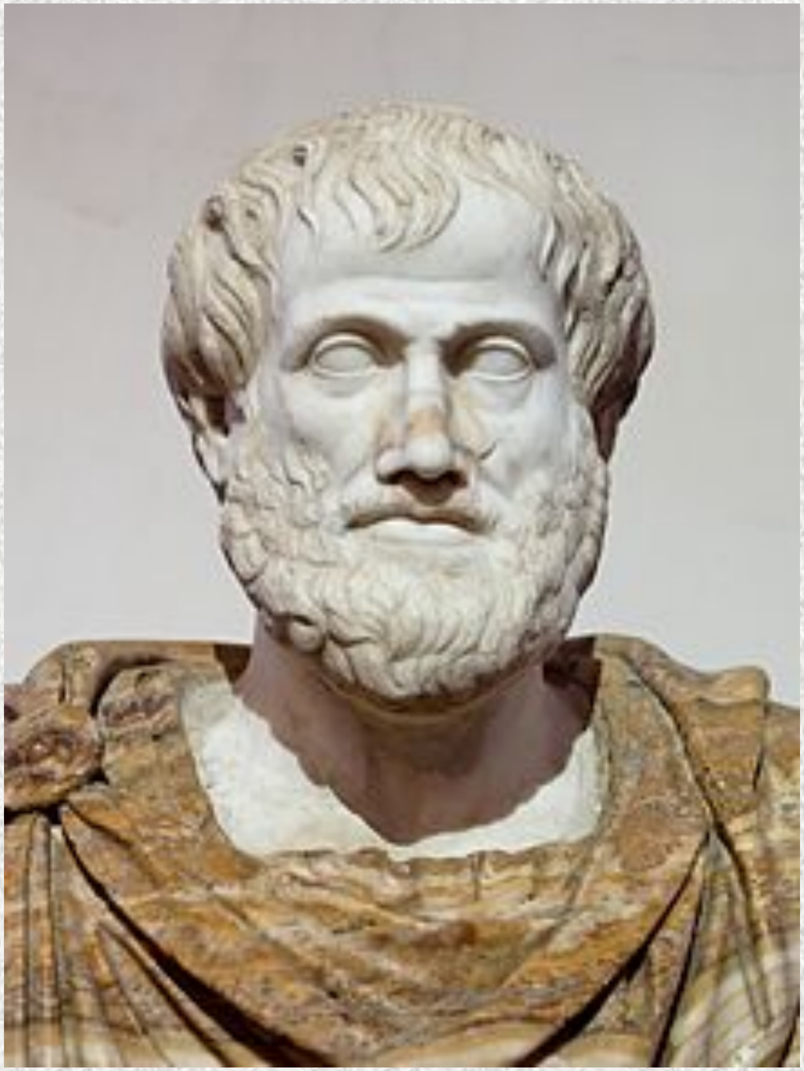
ELEMENT ONE





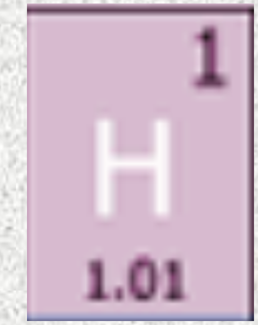
Aristotle (384-322 BC)

1. The aether

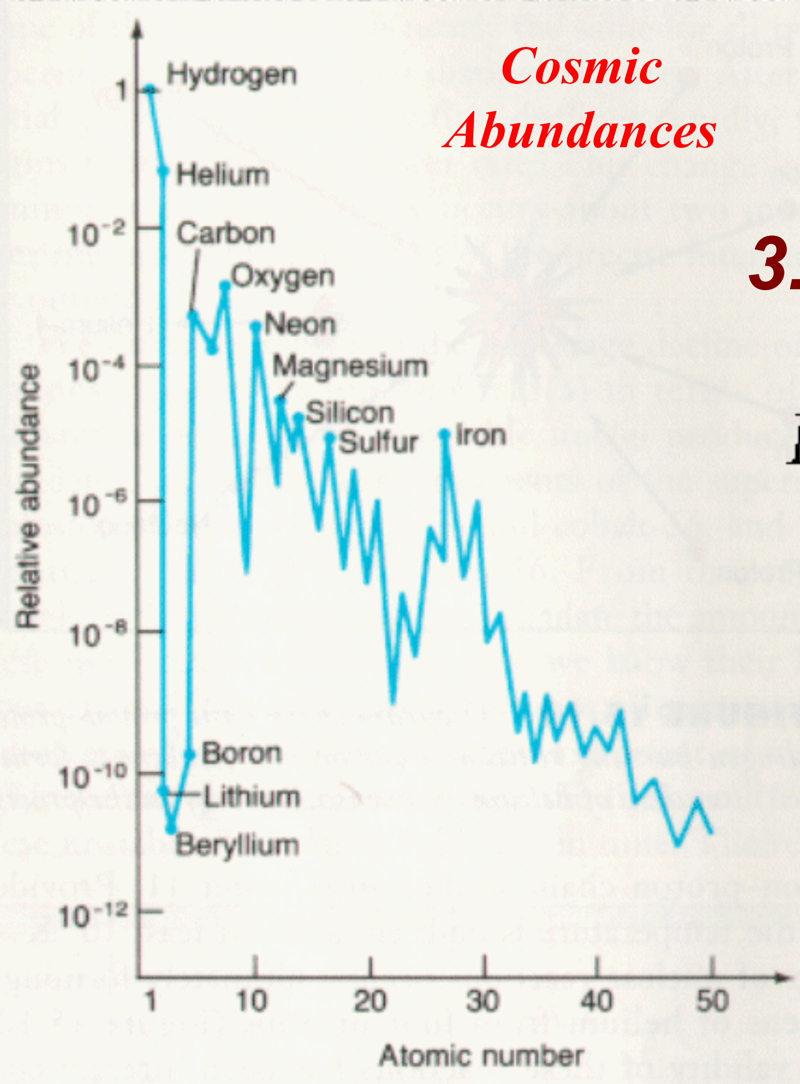
Adding to the four elements proposed by Empedocles: Earth, water, air, and fire

"Outside all the other spheres, the heavenly, fifth element, the aether is manifested in the stars and planets, moves in the perfection of circles."

ELEMENT ONE



2. Most abundant element



3. Tests of fundamental theory

$$\hat{H} = \sum_{i}^{\text{nuclei}} \hat{T}(i) + \sum_{j}^{\text{electrons}} \hat{T}(j) + \sum_{k}^{n}\sum_{l}^{e} \hat{V}(k,l) + \sum_{m}^{e}\sum_{n>m}^{e} \hat{V}(m,n) + \sum_{o}^{n}\sum_{p>o}^{n} \hat{V}(o,p)$$

ELEMENT ONE

3. Simple transformation

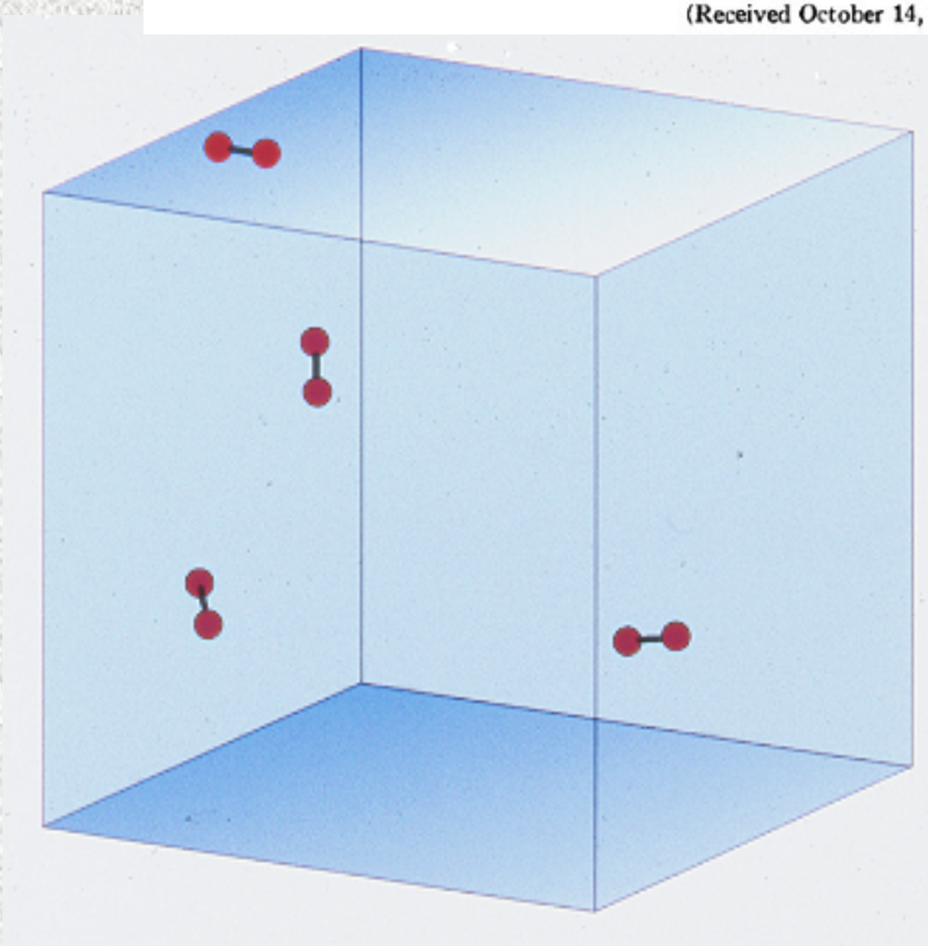
1
H
1.01

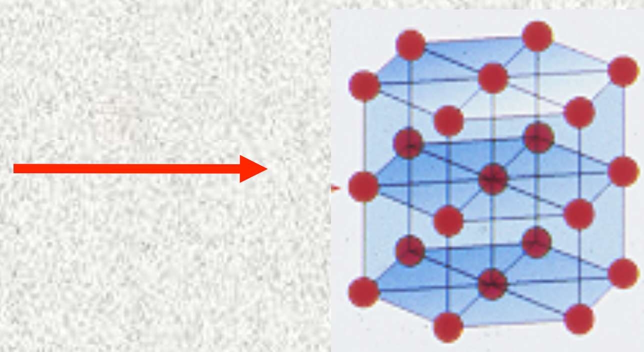
DECEMBER, 1935 JOURNAL OF CHEMICAL PHYSICS VOLUME 3

On the Possibility of a Metallic Modification of Hydrogen

E. Wigner and H. B. Huntington, *Princeton University*

(Received October 14, 1935)





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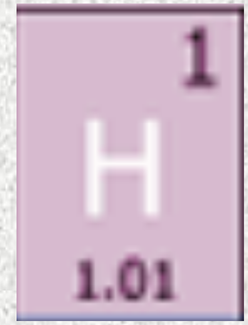
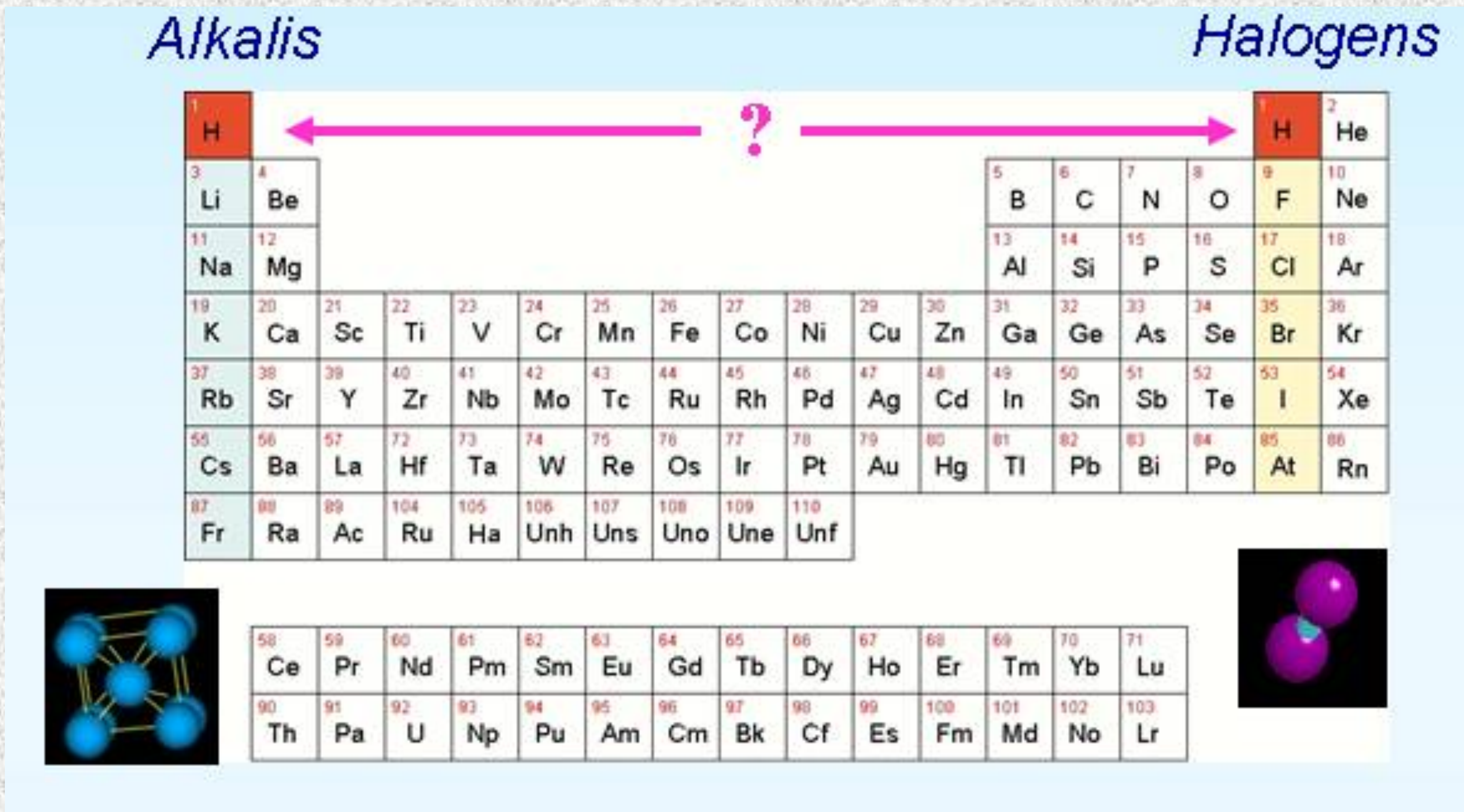
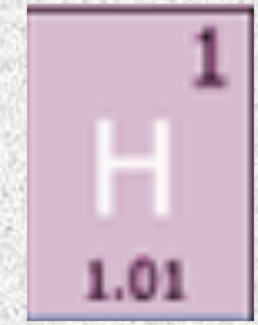


4. Chemical dichotomy



"Reluctant alkali" or "tenacious halogen" [Ashcroft, *Physics World* (1995)]

ELEMENT ONE

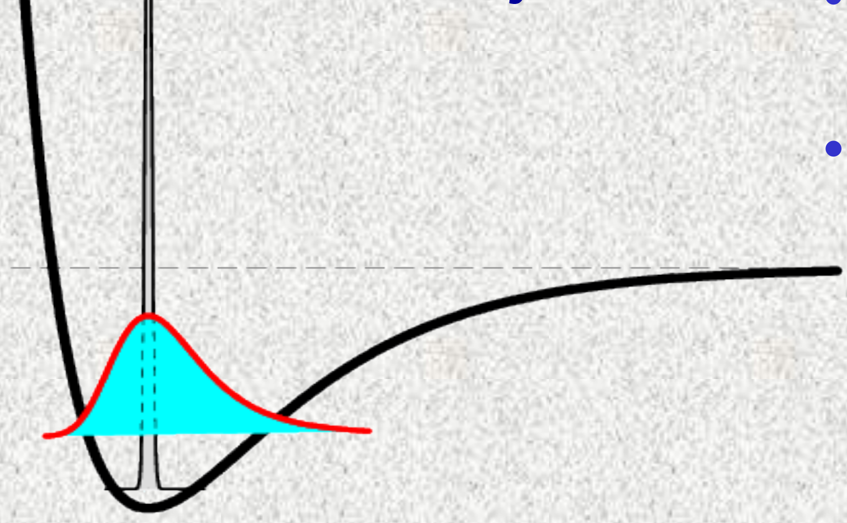


5. Quantum system

$$\hat{H} = \sum_{i}^{\text{nuclei}} \hat{T}(i) + \sum_{i}^{\text{electrons}} \hat{T}(j)$$

Fluid ground state
[Brovman et al., *JETP* (1974)]

'The Element of Uncertainty'

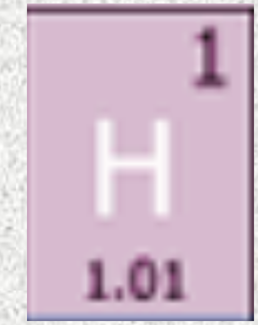


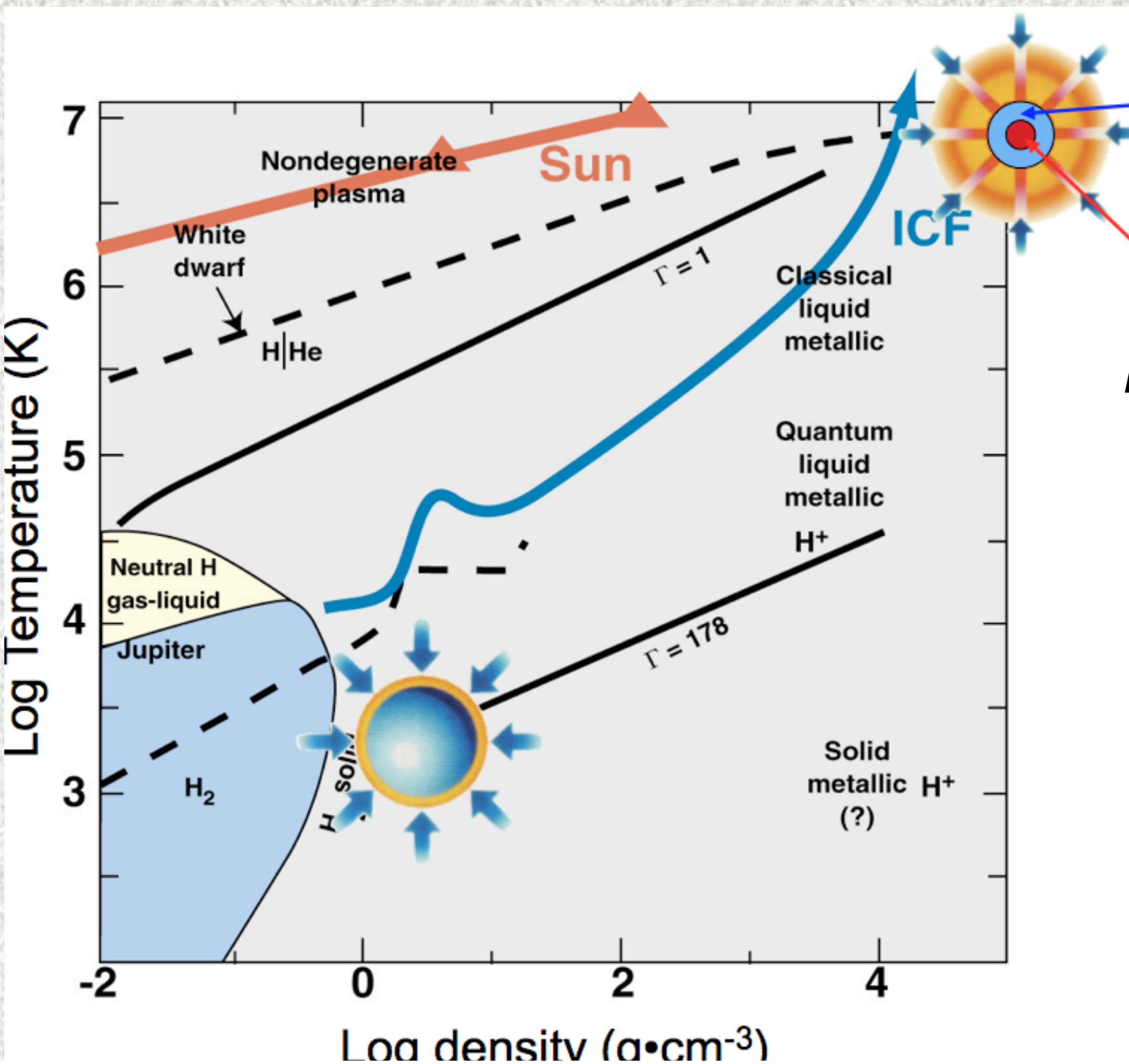
6. Potential energy material

- **High energy density material**
  (400 kJ/mole: 35 x TNT)
- **High-T_c superconductor?**
  [Ashcroft, *Phys. Rev. Lett.* (1968)]
- **Superconducting/superfluid?**
  [Babaev, Sudbo, & Ashcroft, *Phys. Rev. Lett.* (2005)]

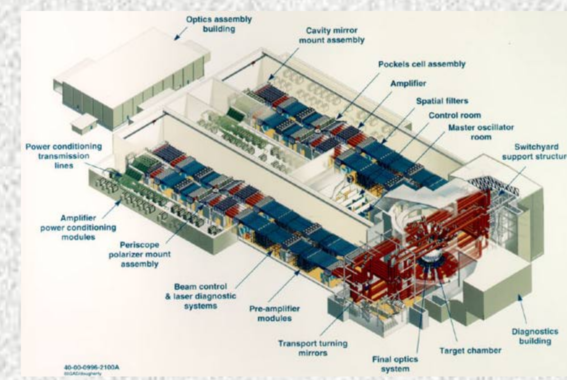
ELEMENT ONE

7. Path to Inertial Confinement Fusion





National Ignition Facility



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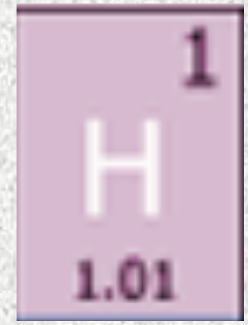
ELEMENT ONE



8. Driven the development of many high-pressure techniques

ρ/ρ_0 = 14
at 300 GPa

- **High compressibility (e.g., deformation of apparatus)**
- **Reactivity with metals (weaken apparatus, electrical leads)**
- **Weak x-ray scattering power**
- **Strong Raman cross-section but variable infrared absorption**
- **Large neutron cross-section (coherent, incoherent)**

<u>Techniques Used Over Different P-T ranges</u>

Optical spectroscopy
Raman, infrared spectroscopy
Brillouin scattering
X-ray diffraction (single xtal, polyxtal)
X-ray inelastic scattering
Neutron diffraction
Neutron inelastic scattering

Non-linear spectroscopy (e.g., CARS)
NMR
Electrical transport
Ultrasonic
Shock compression (Hugoniot)
Isentropic compression

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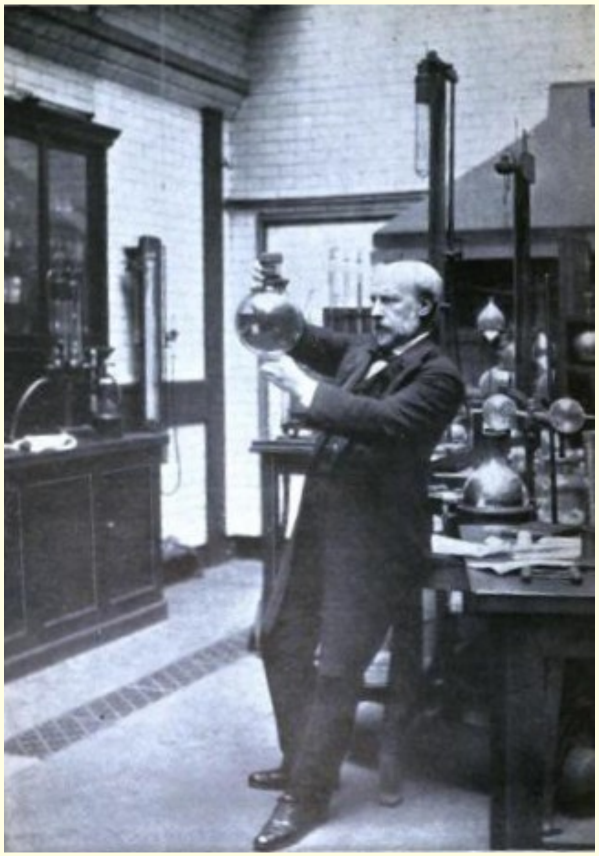
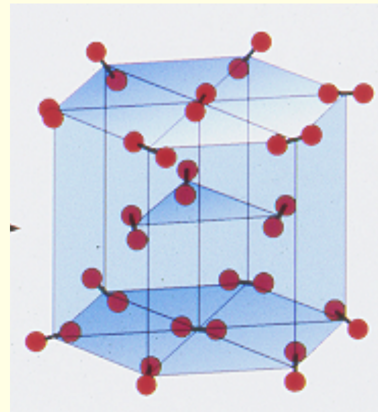
Hydrogen in condensed phase

1898 – First liquified – 20 K
1899 – First solidified – 14 K



Sir James Dewar

X-ray diffraction



J=0
p-H_2 (hcp)

V_0 = 23.2 cm^3/mol
R(H_2-H_2) = 3.0 Å
R(H-H) = 0.74 Å

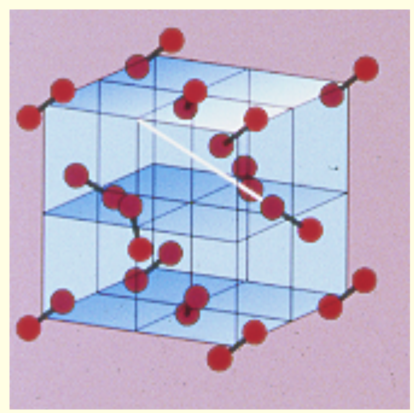
[Keesom et al., *Comm. Kamerlingh Onnes Lab* (1930)]

THE ROTATIONAL MOTION OF MOLECULES IN CRYSTALS

By Linus Pauling

Gates Chemical Laboratory, California Institute of Technology

(Received May 7, 1930) [*Phys. Rev.* (1930)]

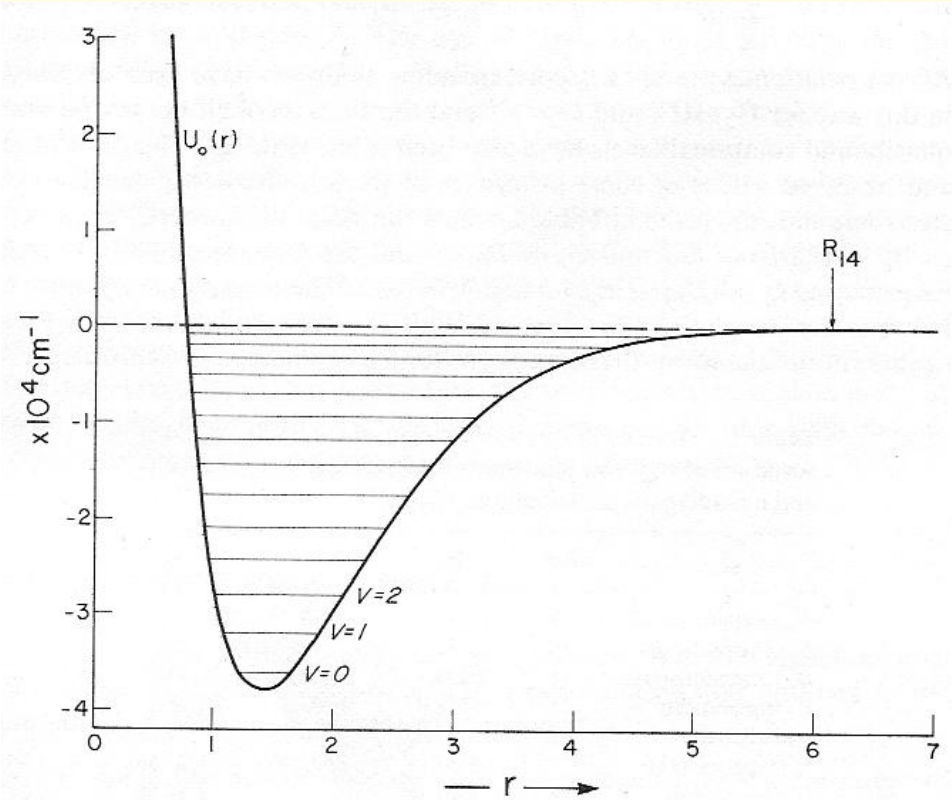


J=1 ordered state
o-H_2 (Pa3)

[Mills and Schuch, *Phys. Rev. Lett.* (1964)]

Molecular structure and bonding

INTRAMOLECULAR POTENTIAL
(pure; i.e., isolated molecule)

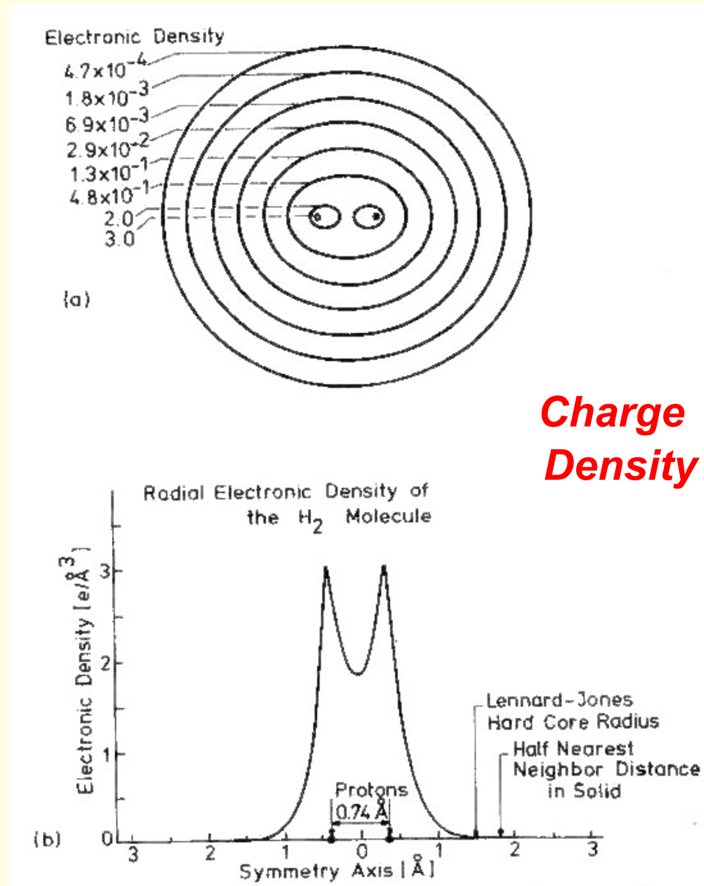


Origin of the chemical bond
- ***Valence bond (Heitler-London)***
- ***Molecular orbital (Mulliken)***

- **Strong covalent bond: 4.53 eV**
- **14 bound vibrational states**

[Kolos & Wolniewicz, *J. Chem. Phys.* (1964-1974)]

ISOLATED MOLECULE
Nearly spherical electron density

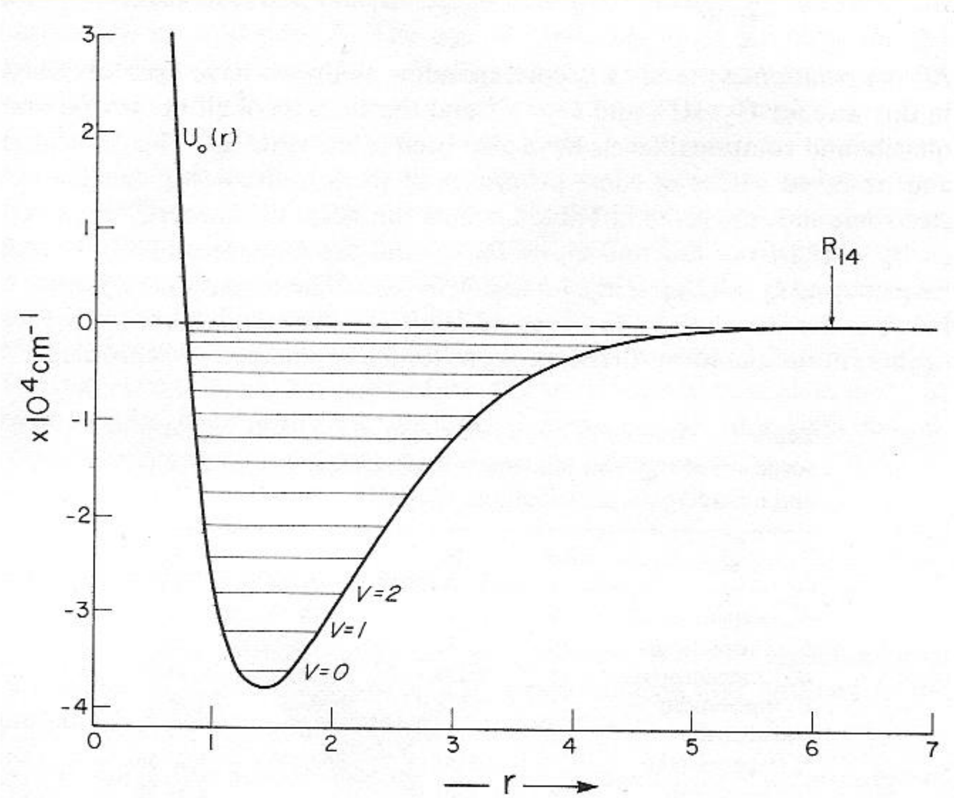


Charge Density

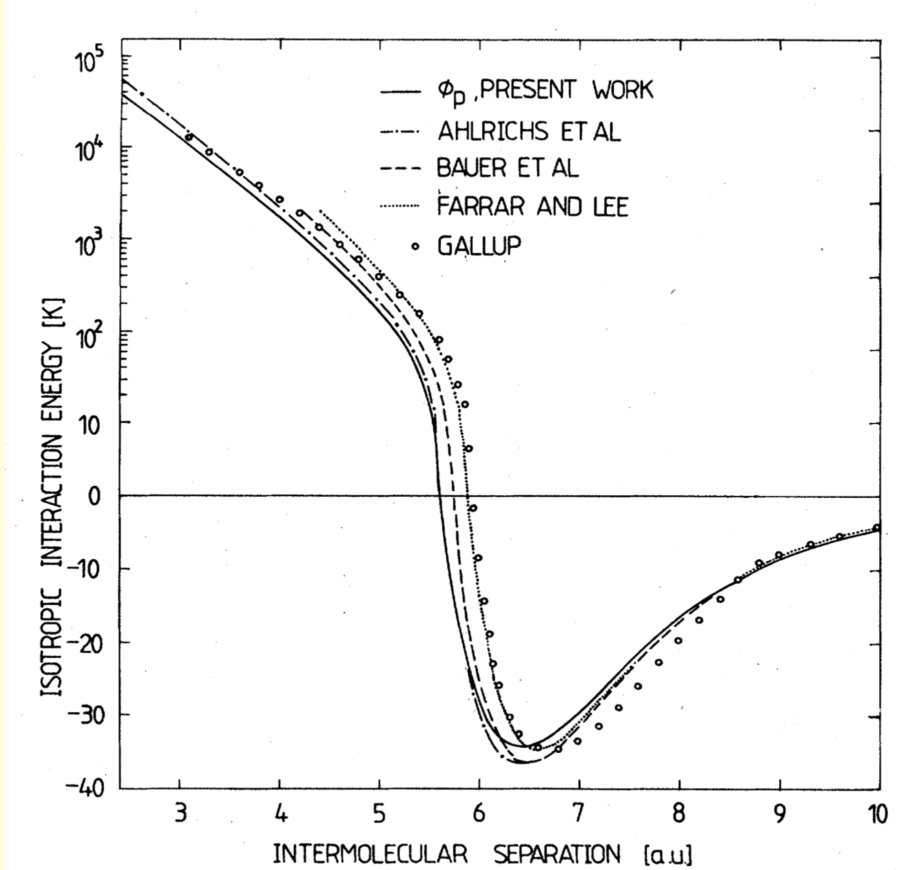
[Silvera, *Rev. Mod. Phys.* (1980)]

Intramolecular and intermolecular interactions

INTRAMOLECULAR POTENTIAL
(pure; i.e., isolated molecule)



INTERMOLECULAR POTENTIAL
(effective potential in condensed phase)



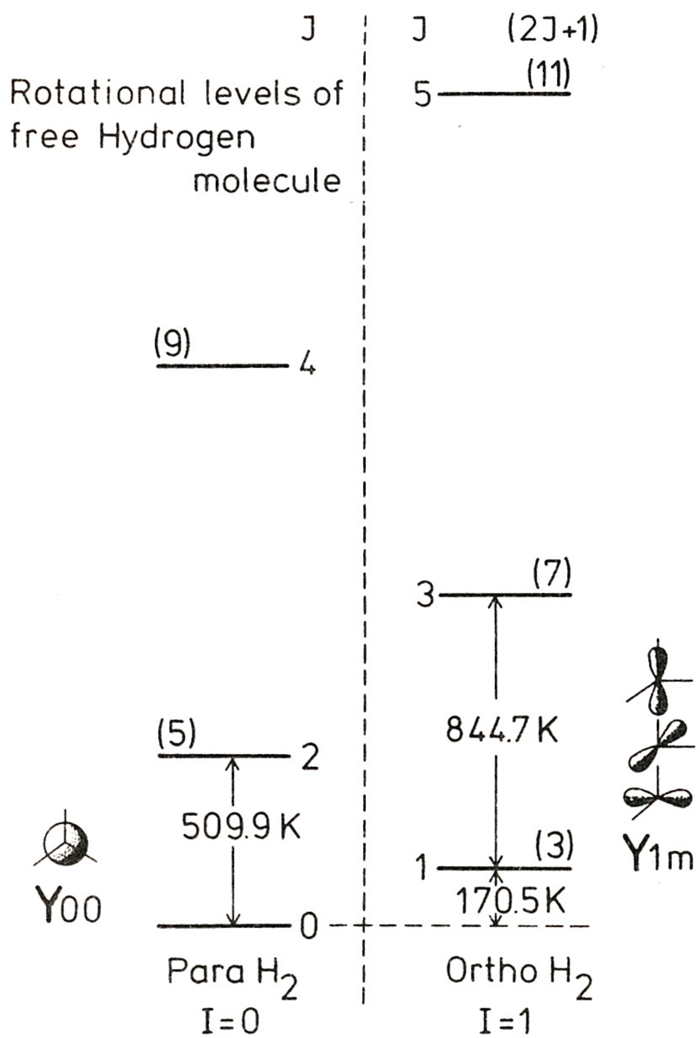
- Strong Raman cross-section
- No dipole-allowed IR absorption

- Binding energy 3.0 meV (35 K)
- Interactions: *Isotopic* + *Anisotropic*
  - *Leading anisotropic term is electric quadrupole-quadrupole (EQQ) interaction (odd J)*

[Silvera, *Rev. Mod. Phys.* (1980)] *Carnegie Institution*

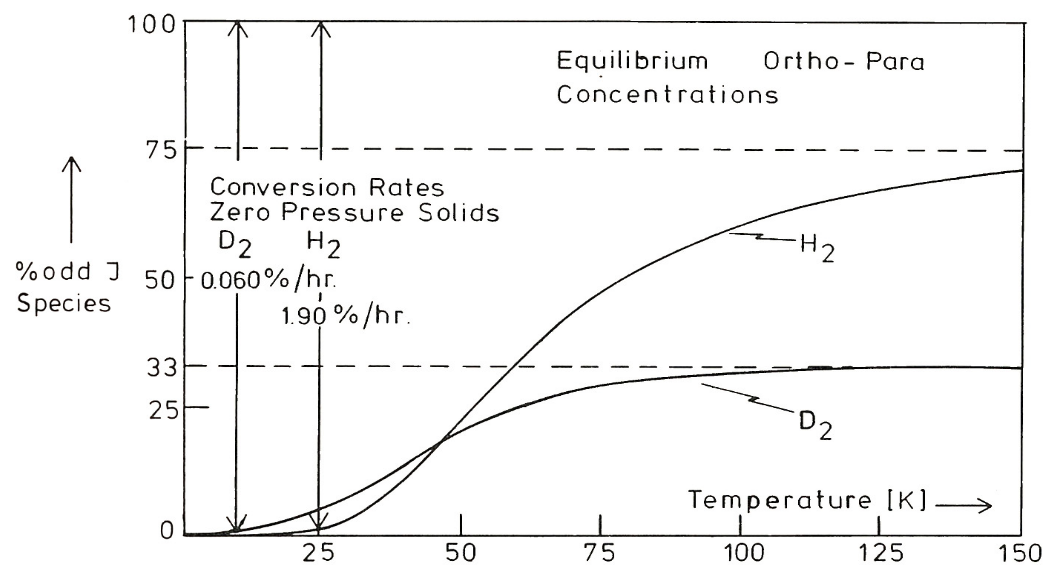
Ortho-para distinction

BJ (J+1)



Coupling of nuclear spin I_N and rotational states J in total molecular spin I_{mol}

	I_N	I_{mol}	J	Species
H_2	½	0	even	p-H_2
		1	odd	o-H_2
D_2	1	1	odd	p-D_2
		0,2	even	o-D_2



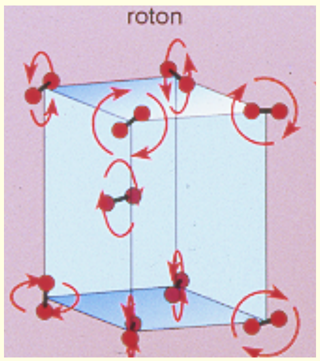
[Silvera, *Rev. Mod. Phys.* (1980)]

Carnegie Institution

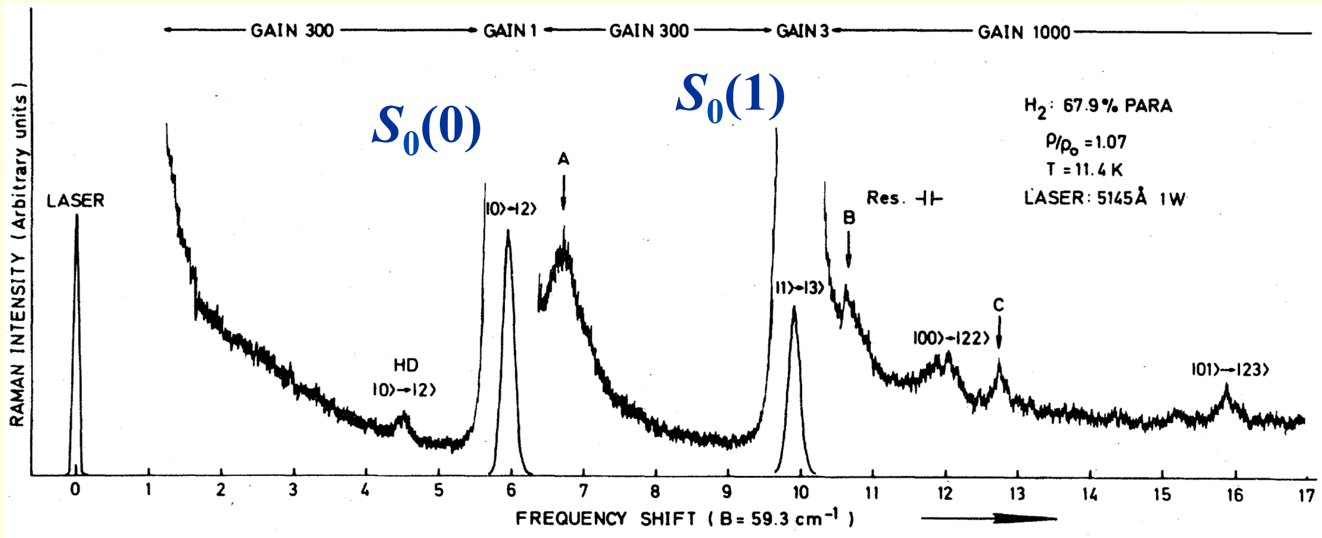
Vibrational excitations

O,P,Q,R,S … ΔJ = -2, -1, 0, -1, 0, -2

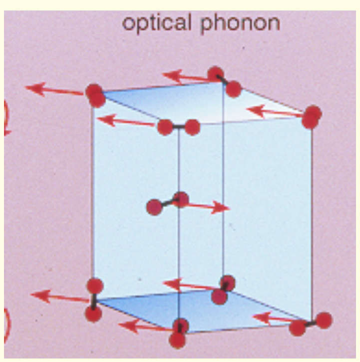
Rotons



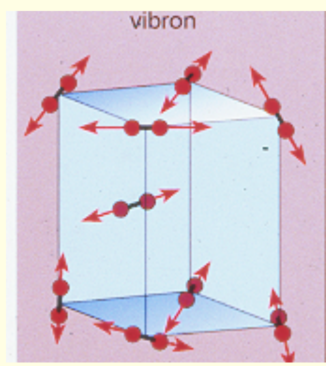
$S_0(J)$



Lattice Modes



Vibrons



$Q_{\Delta v}(J)$; e.g., $Q_1(1)$

PRESSURE EFFECTS

- **Rotational ordering**
  - *breakdown of J*
- **Molecular stability**
  - *lattice mode = vibron?*
- **Molecular interactions**
  - *molecular coupling?*

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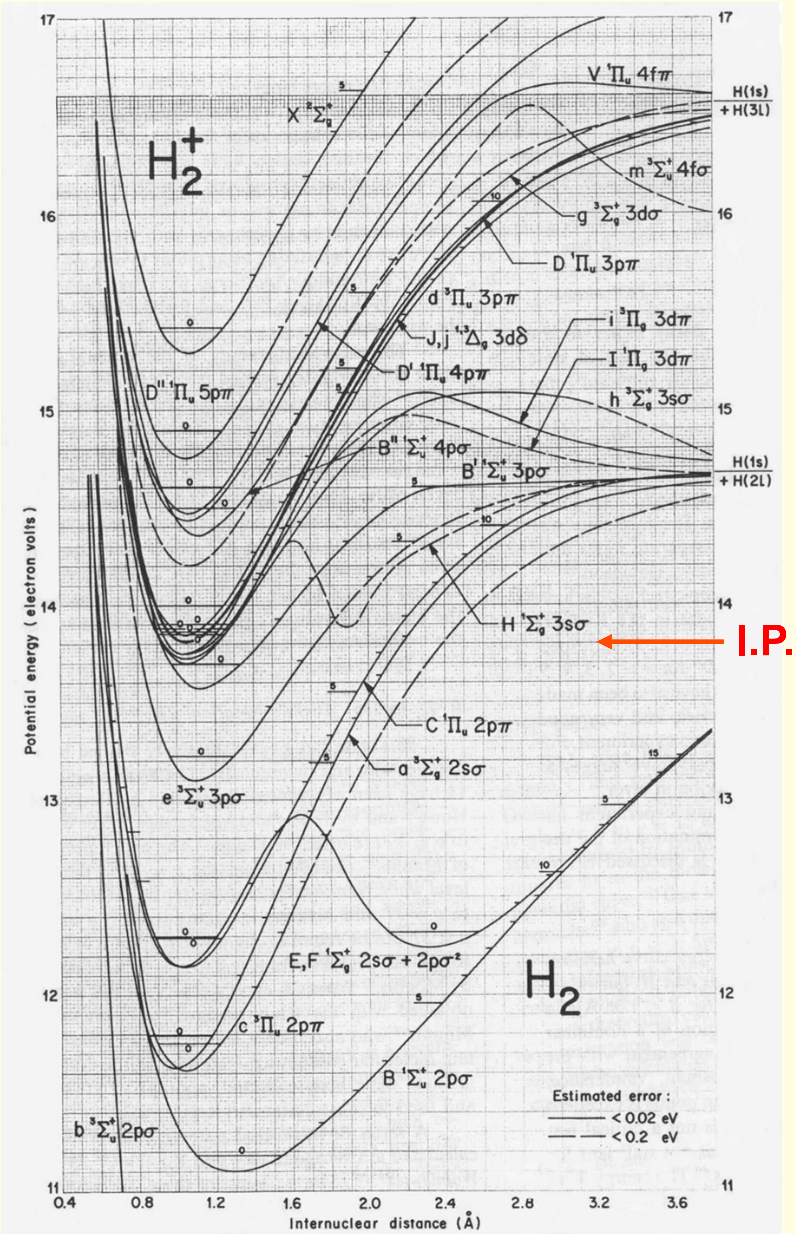
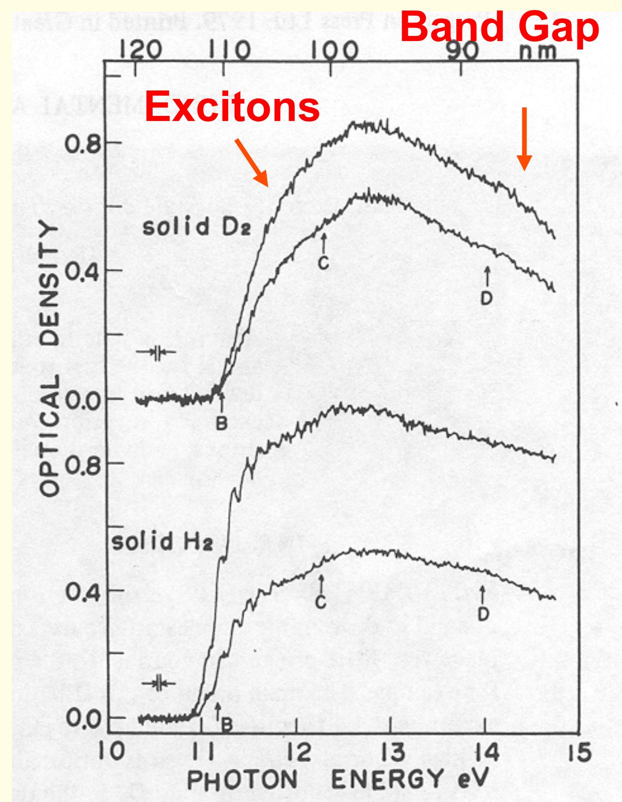
H_2 excited electronic states



[Sharp, *J. Phys. Chem. Ref. Data* (1979)]

UV Absorption (zero pressure)



[Inoue *et al. Solid State Comm.* (1979)]

Refractive Index of the Hydrogen Molecule

M. KARPLUS
Department of Chemistry and Watson Laboratory, Columbia University, New York, New York*

(Received 13 March 1964)

$$\epsilon_2(\omega) = \frac{e^2}{\pi m^2 \omega^2} \sum_{v,c} \int_{BZ} d\mathbf{k} |\mathbf{e}\cdot\mathbf{M}_{cv}(\mathbf{k})|^2 \delta(E_c(\mathbf{k}) - E_v(\mathbf{k}) - \hbar\omega),$$

$$\mathbf{e}\cdot\mathbf{M}_{cv}(\mathbf{k}) = \langle \psi_{c\mathbf{k}} | \mathbf{e}\cdot\mathbf{p} | \psi_{v\mathbf{k}} \rangle$$

[*J. Chem. Phys. Rev.* (1964)]

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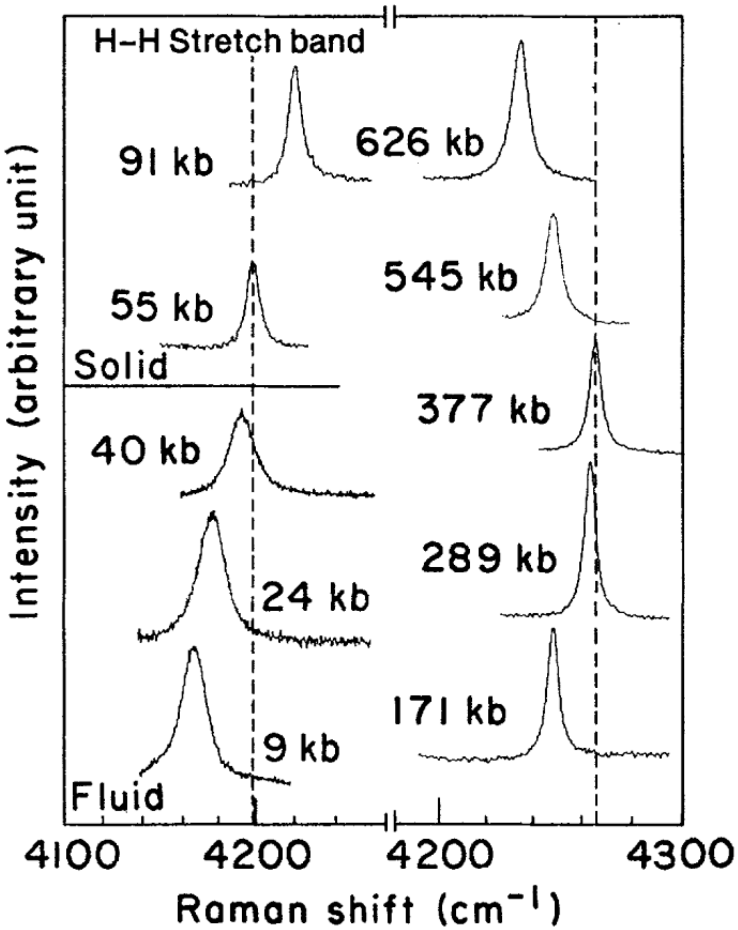
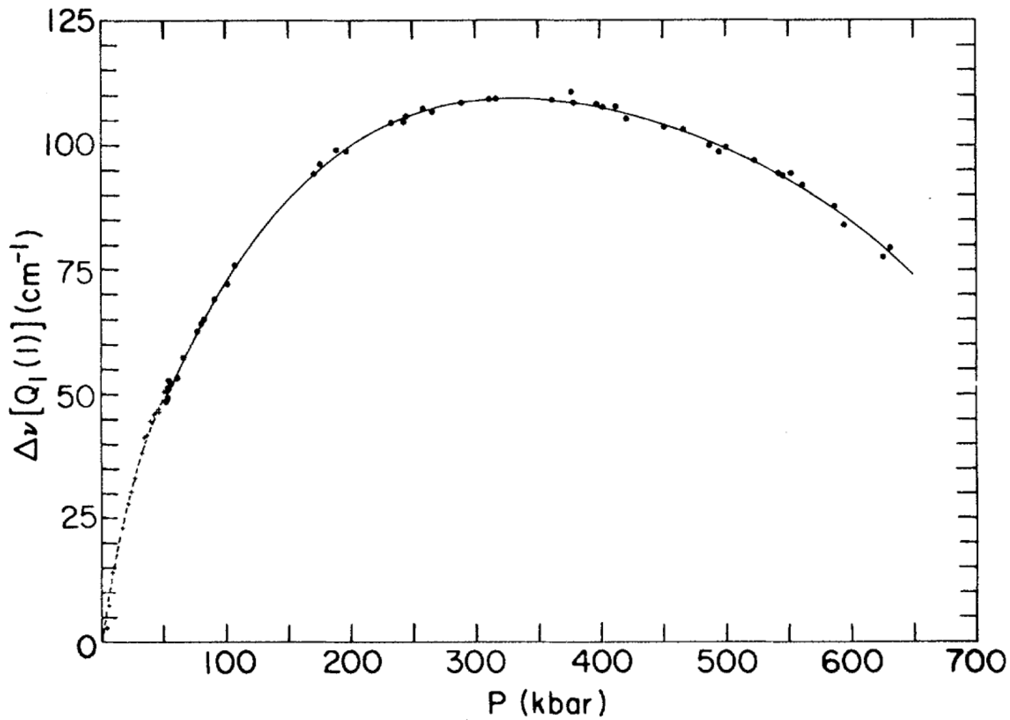
Room-Temperature Compression of Hydrogen Gas

Freezing at
Room Temperature
5.4 GPa

[Mao and Bell, *Science* (1979)]

High-pressure measurements of the Raman vibron

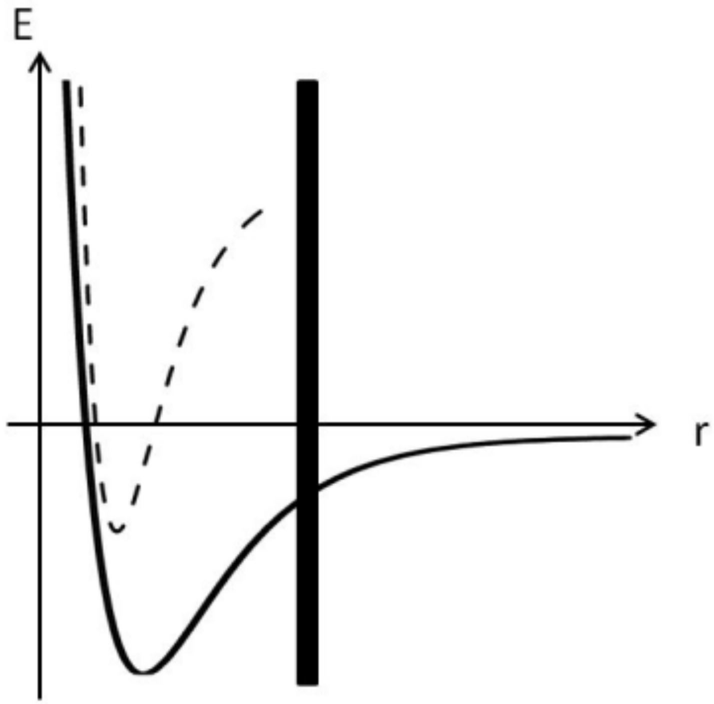


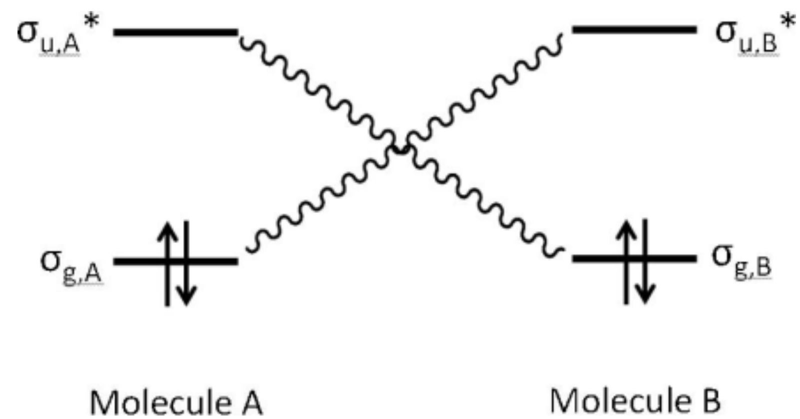


[Sharma *et al., Phys. Rev. Lett* (1978)]

Evidence for bond weakening?

Effect of compression on vibron frequencies



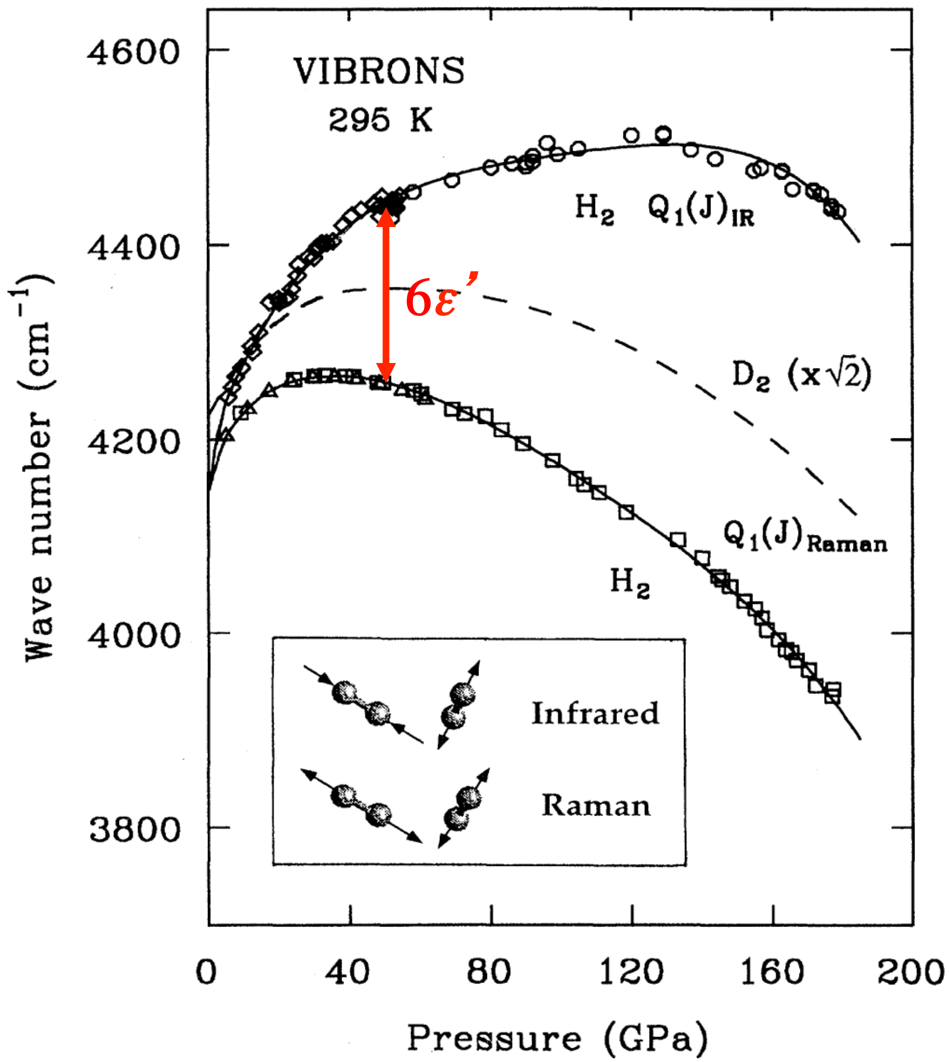


Compression
Frequency Increase

Orbital interactions
Frequency Decrease

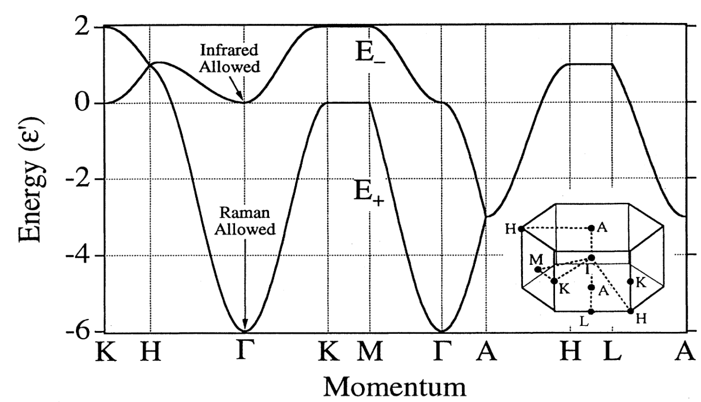
[Labet, Hoffmann, & Ashcroft, *J. Chem. Phys.* (2012)]

Enhancement of vibrational coupling: combining Raman and IR spectroscopy



[Hanfland *et al.*, *Phys. Rev. Lett* (1992)]

$$H_{JK} = \sum_m W'_m \,|m\rangle\langle m| - \frac{1}{2}\sum_{m,n\neq m} \epsilon'_{m,n}\,|m\rangle\langle n|$$



$6\varepsilon'$

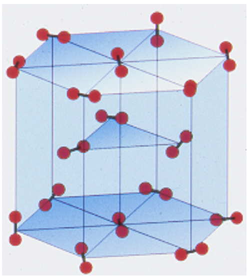
$6\varepsilon'$ = 3 cm^{-1} *(zero pressure)*

520 cm^{-1} *(180 GPa)*

Carnegie Institution

Crystal structure by high-pressure diffraction

hexagonal close packed

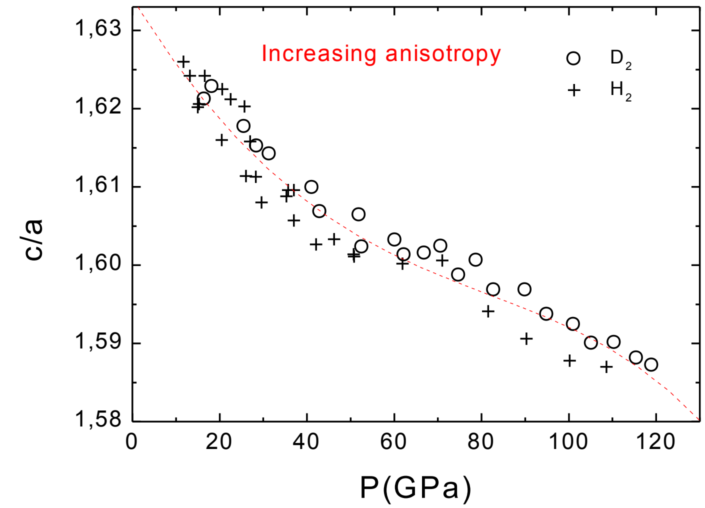


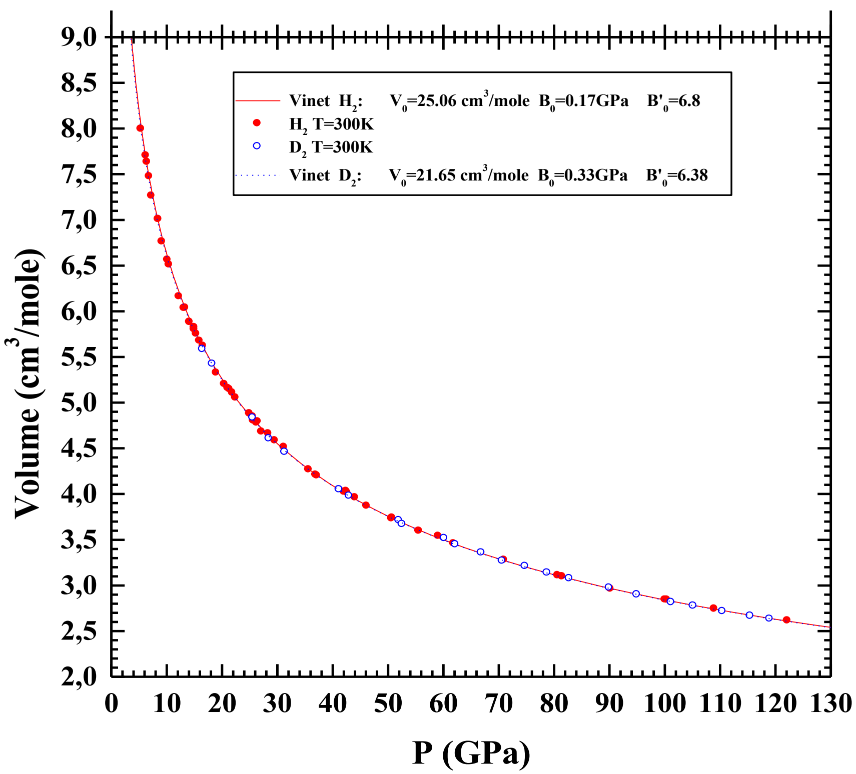
Neutron diffraction (to 30 GPa)

[Glazkov *et al.*, *JETP Lett.* (1986)]

X-ray diffraction (to 200 GPa)

[Mao et al. *Science* (1988); Loubeyre *et al.* *Nature* (1996); Kawamura *et al.*, *Solid State Comm.* (2001)]

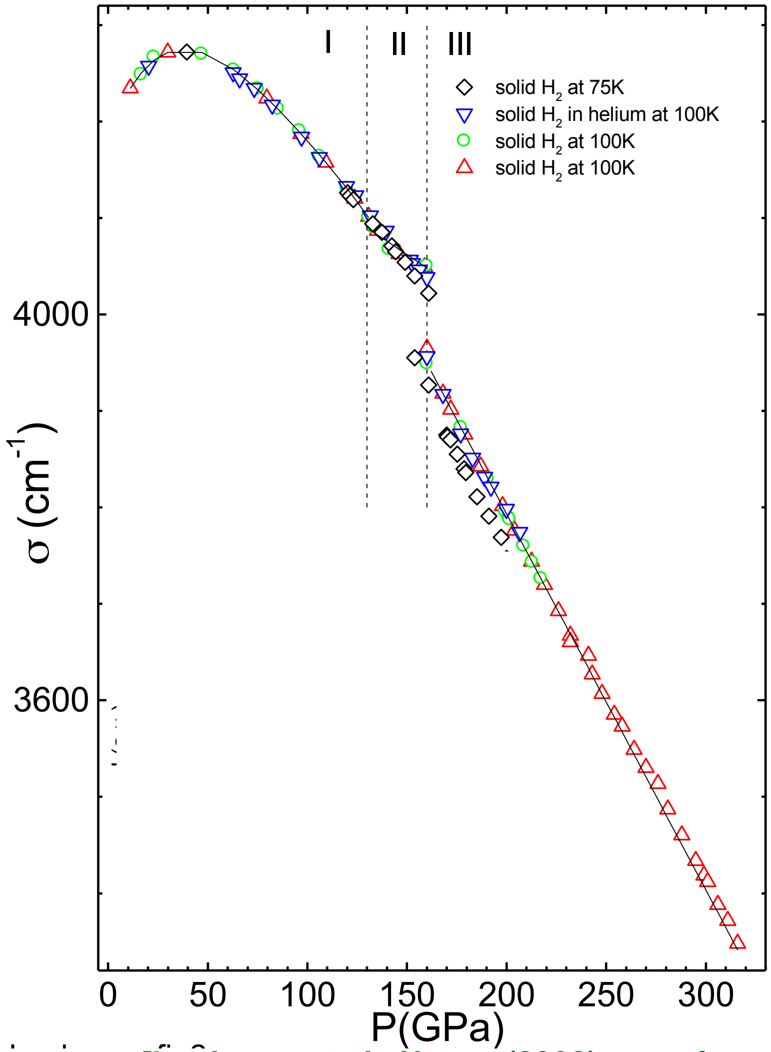




- **Monotonic decline in c/a**
- **No measurable isotope effect**
- **High compressibility**
  - **shifts P_{met} > 400 GPa**
  - **$\int$VdP compared to theory**

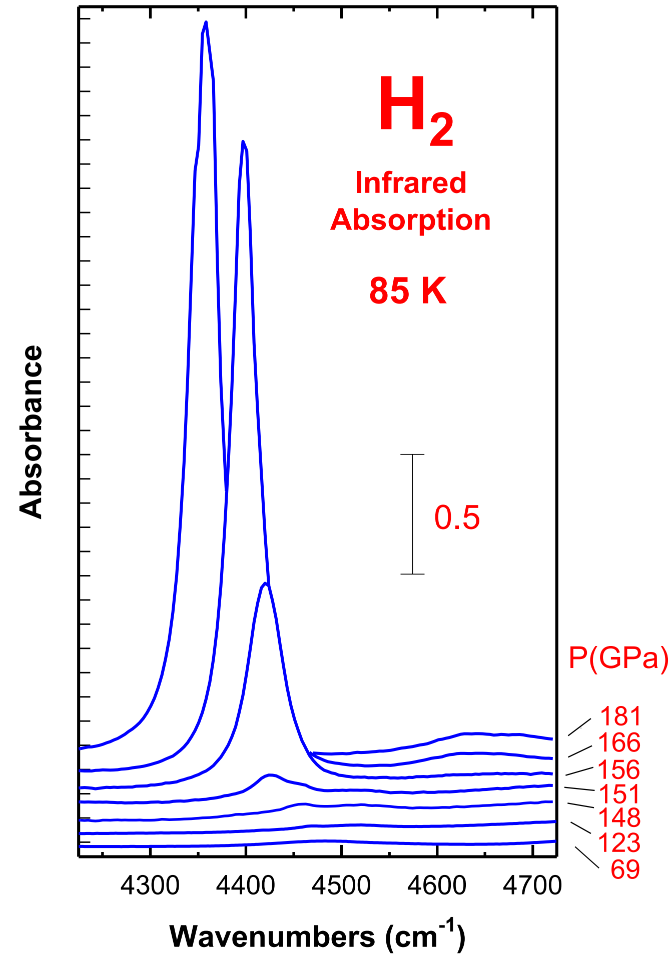
Higher pressure transition: phase III

Raman Vibron



[Loubeyre *et al.*, *Nature* (2002); see also Hemley & Mao, Phys. Rev. Lett. (1988)]]

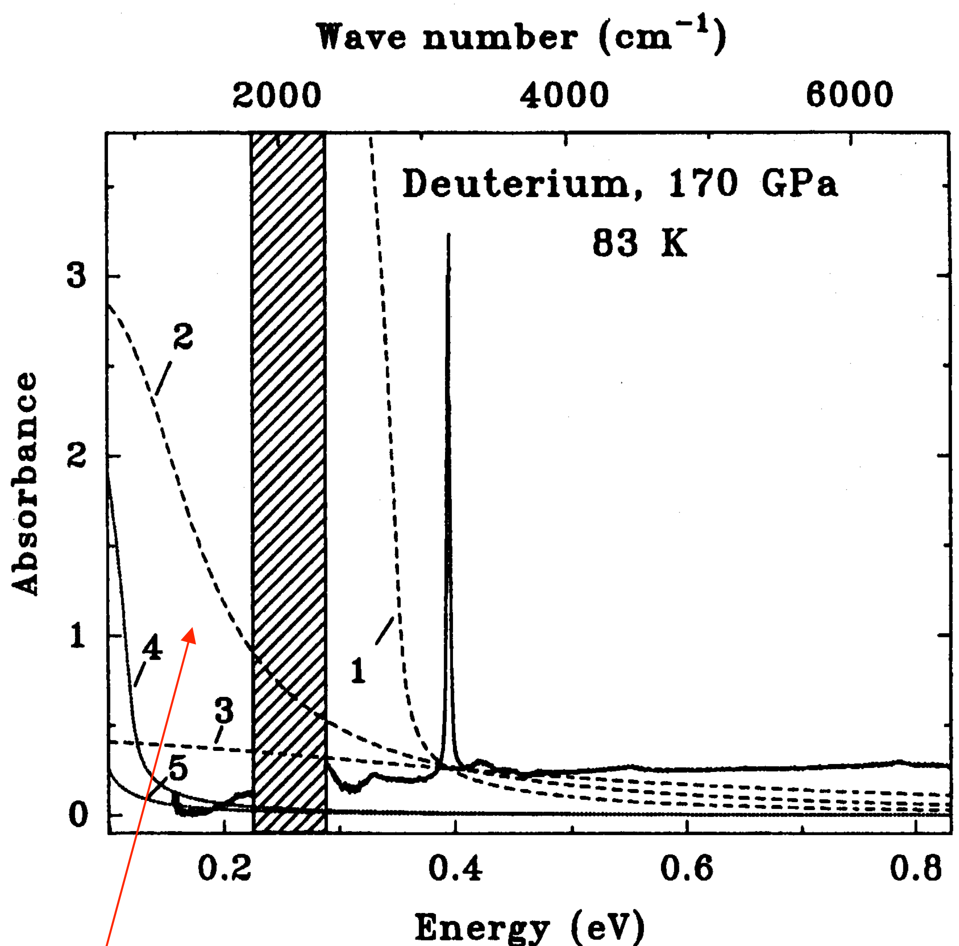
Infrared Vibron



[Hanfland *et al. Phys. Rev. Lett.* (1993); Hemley et al., *ibid.* (1994) Chen *et al. ibid* (1995)]

Carnegie Institution

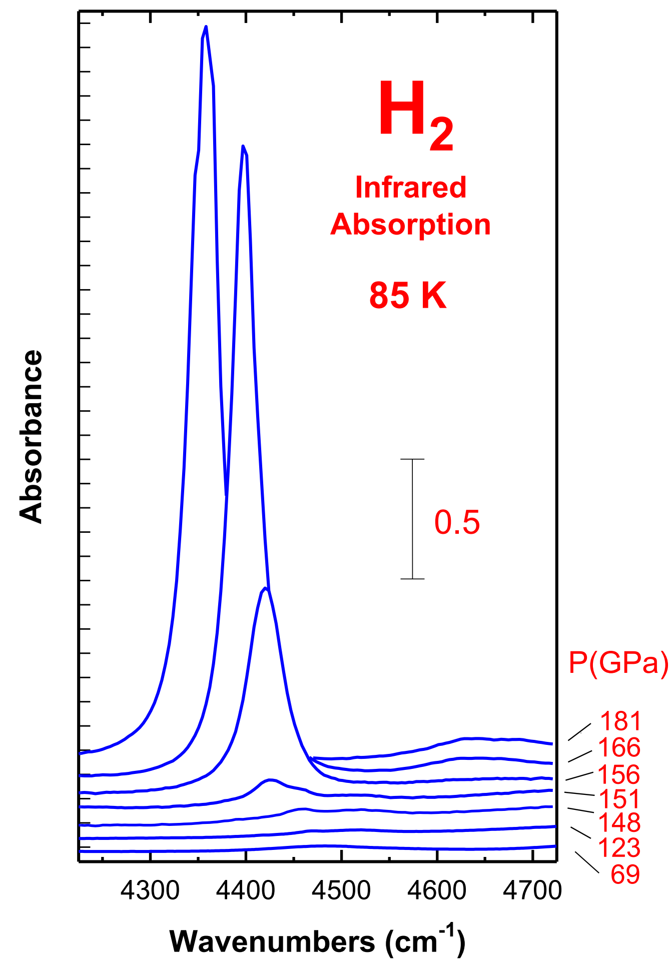
Infrared spectroscopy of phase III: charge transfer instead of metallization



[Hemley *et al. Phys. Rev. Lett.* (1996); see also, Chen *et al. ibid* (1996)]

Predicted Drude models

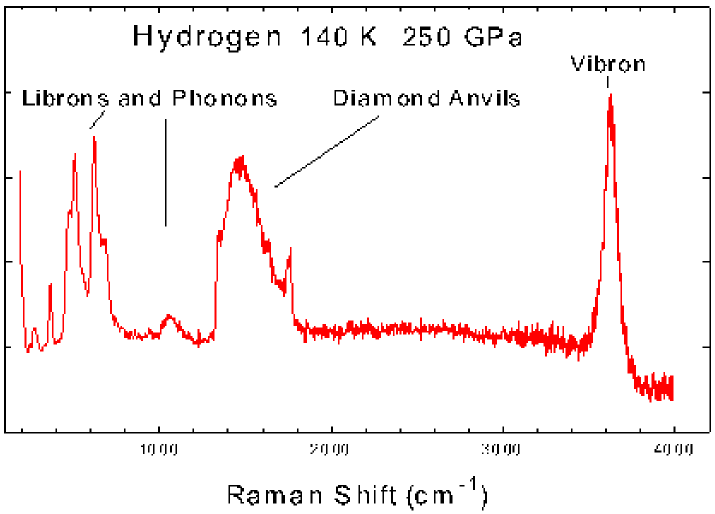
Infrared Vibron

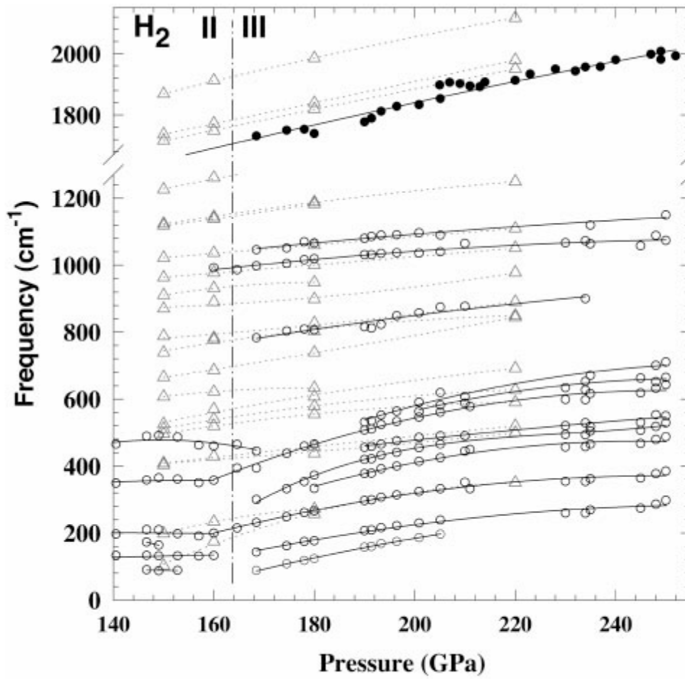


[Hanfland *et al. Phys. Rev. Lett.* (1993); Hemley et al., ibid. (1994) Chen *et al. ibid* (1995)]

Vibrational spectroscopy of phase III: low to high frequency excitations

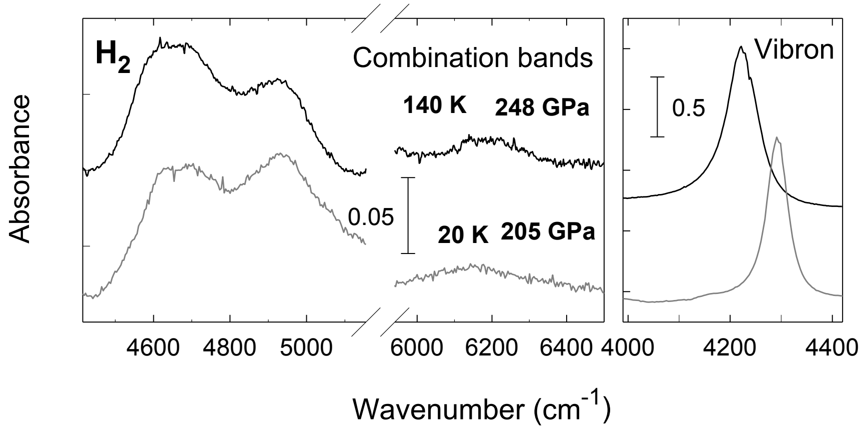
RAMAN





[Souza *et al.* *Phys. Rev. Lett.* (1997); Kohanoff *et al.*, ibid (1999); Edwards & Ashcroft *Nature* (1999)]

INFRARED



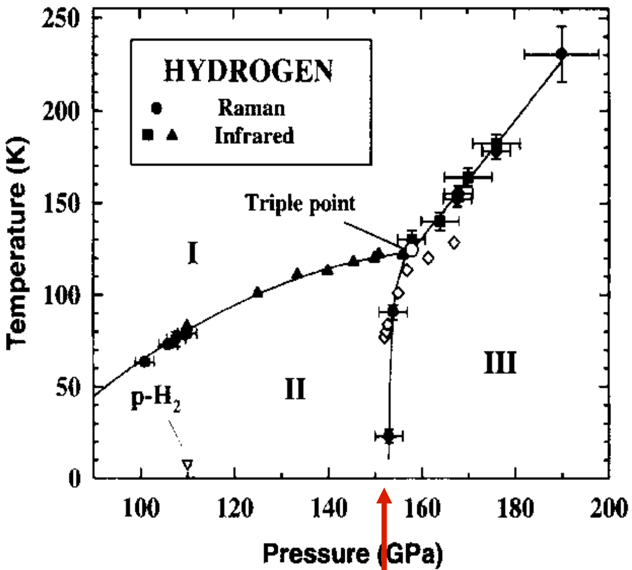
[Goncharov *et al., PNAS* (2001)]

- J no longer a good quantum number
- Vibron softening/orient. ordering
- Molecules persist to >320 GPa

[Loubeyre *et al., Nature* (2002)]

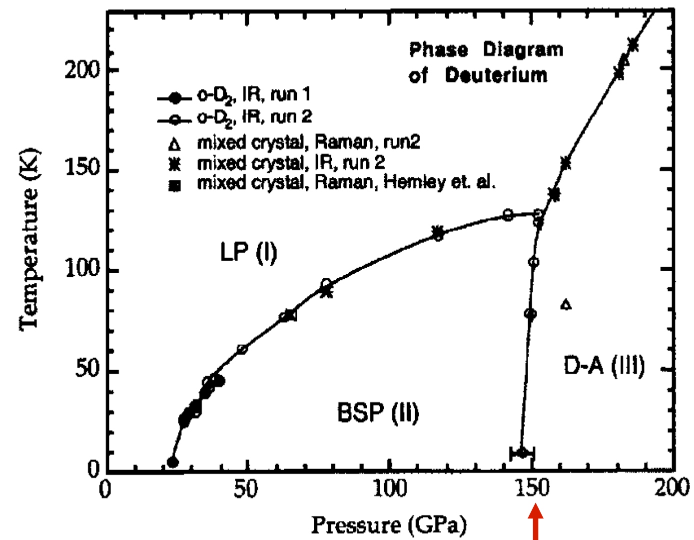
Carnegie Institution

P-T phase diagram of the solid hydrogens from vibrational spectroscopy to 200 GPa



[Mazin *et al.*, *Phys. Rev. Lett.* (1997)]

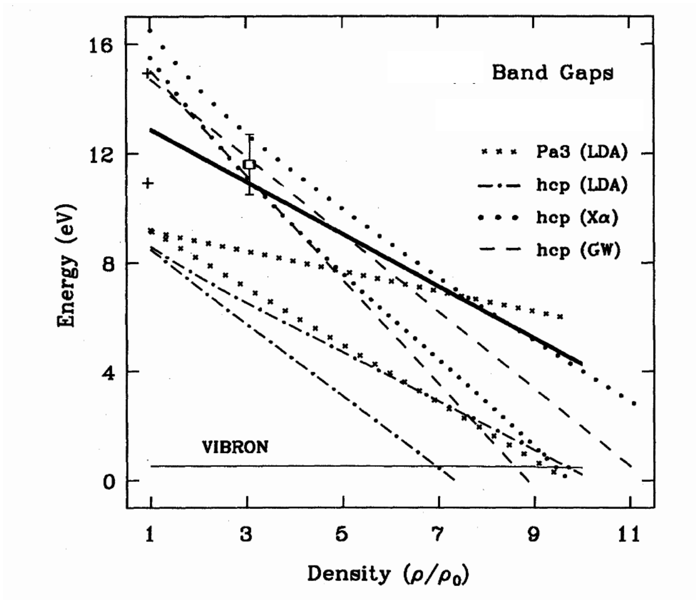
$\rho/\rho_0 = 9$



[Chen *et al.*, *Phys. Rev. Lett.* (1995)]

$\rho/\rho_0 = 9$

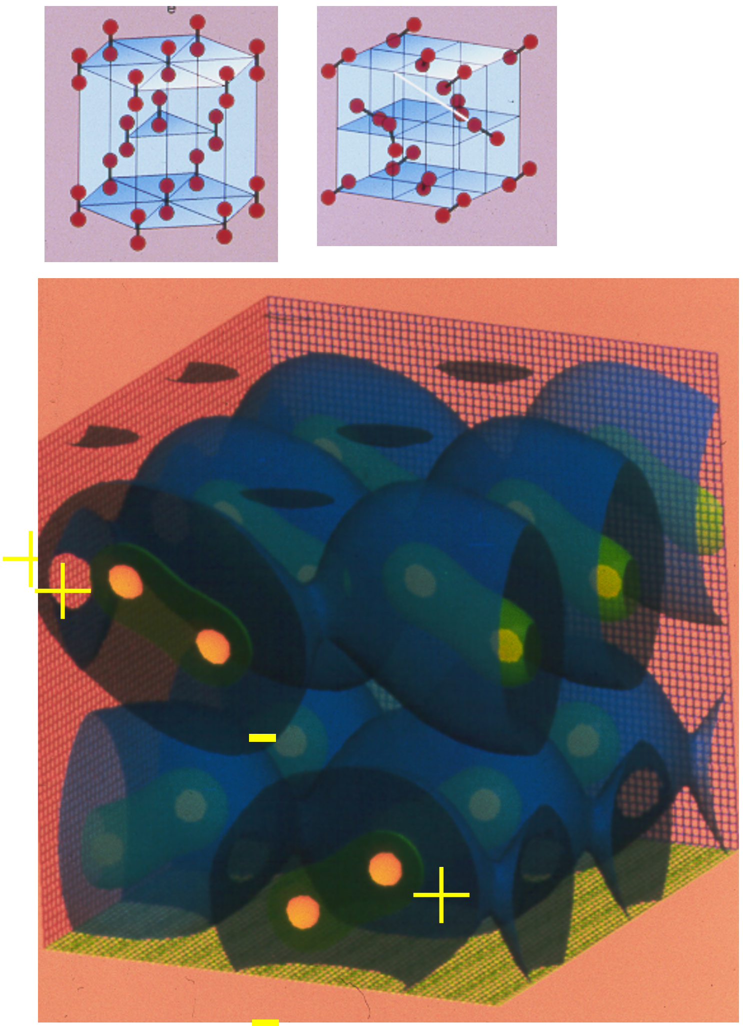
THEORETICALLY PREDICTED BAND GAPS



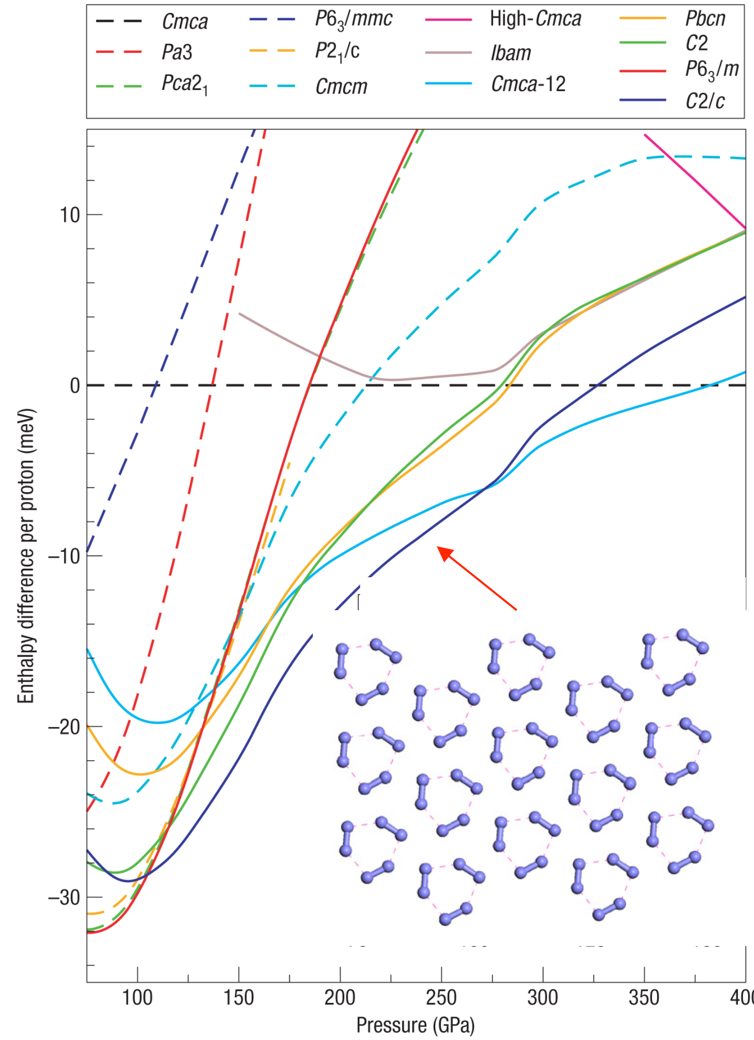
Relationship to metallization?

Theory challenge for phase III: preventing band overlap with the correct crystal structure

Molecular orientation and band overlap



[Kaxiras *et al.*, *Phys. Rev. Lett.* (1992)]



[Pickard & Needs, *Nature Physics* (2007)]

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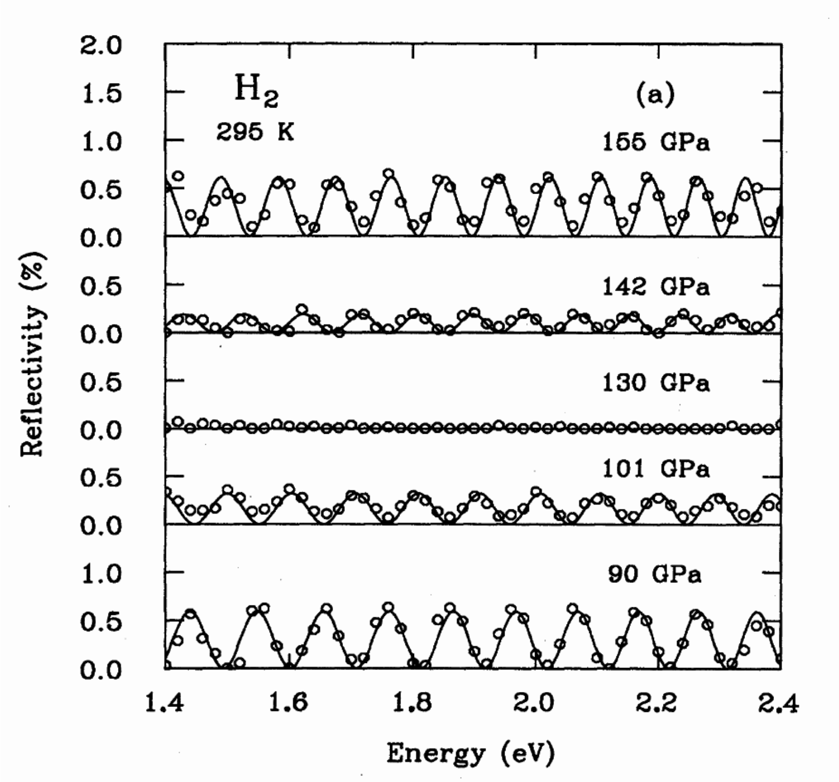
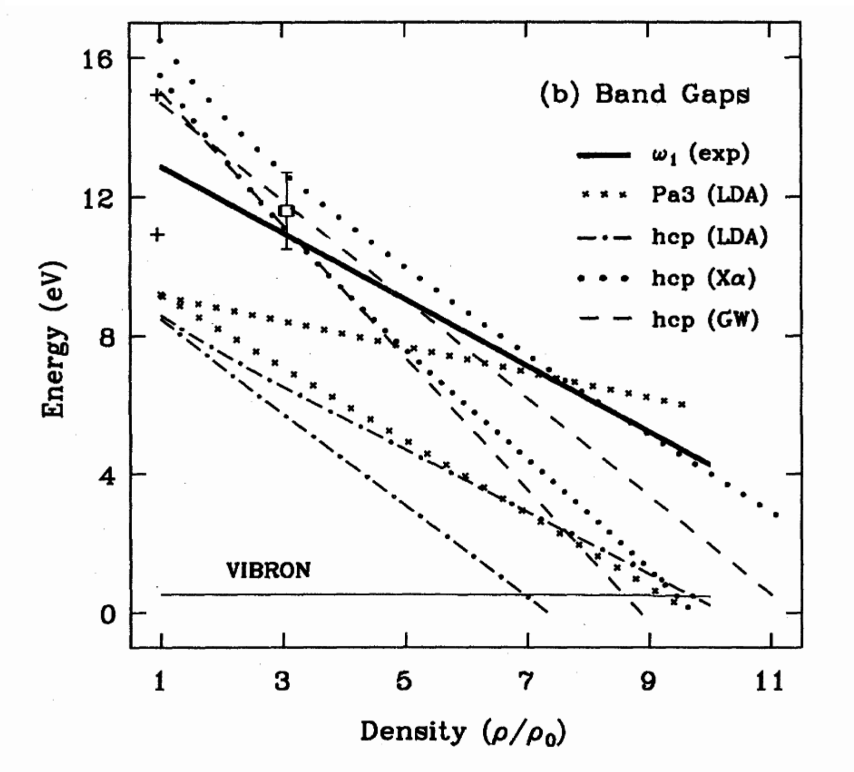
Dielectric Properties

Refractive Index and Oscillator Models





$$I(\nu) = A\sin\{4\pi B[1 + (\nu - \nu_0)C]\nu + D\}.$$

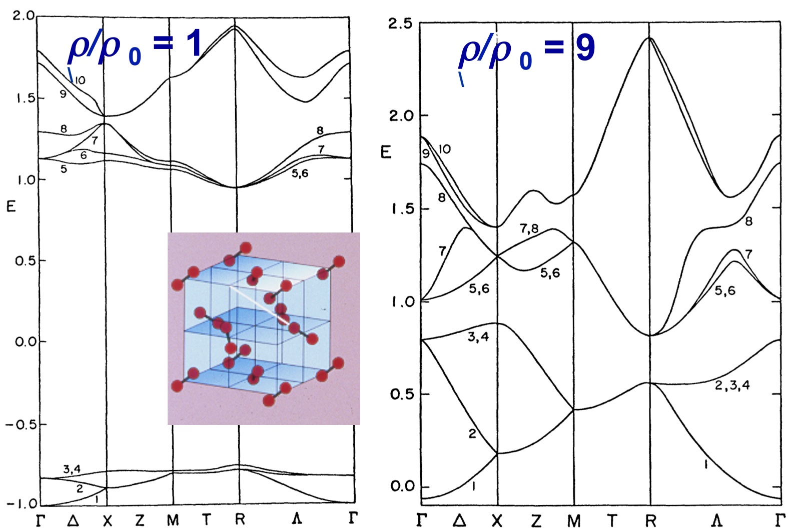
$$C = (1/n_0)(dn/d\nu)_{\nu_0}$$

$$n^2 - 1 = E_d E_0 / (E_0^2 - \hbar^2\omega^2)$$

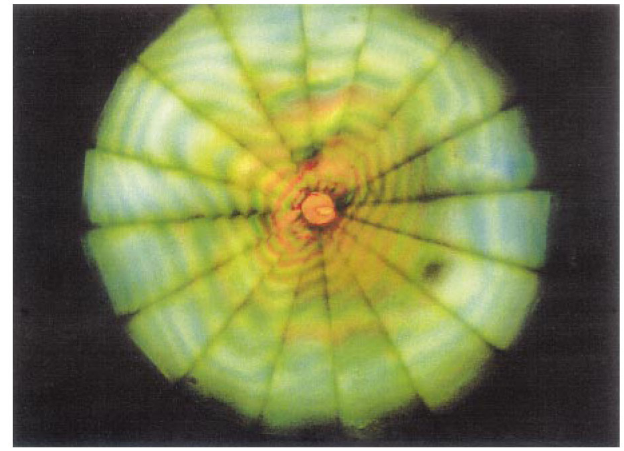
[Hemley *et al.*, *Nature* (1991)]

- **Oscillator model fits to $\varepsilon(\omega)$**
- **Constrains the direct band gap not indirect gaps**
- **Direct measurements?**

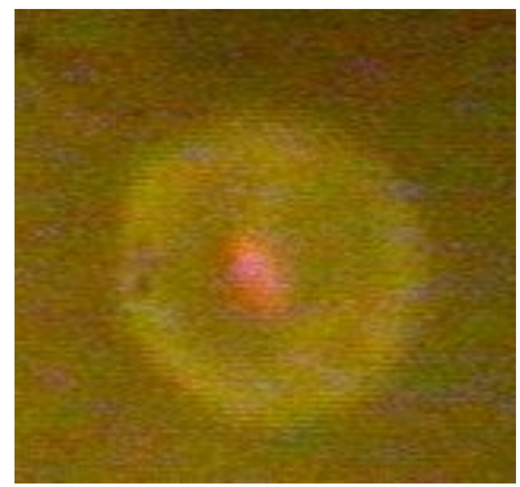
Visible absorption at ~300 GPa



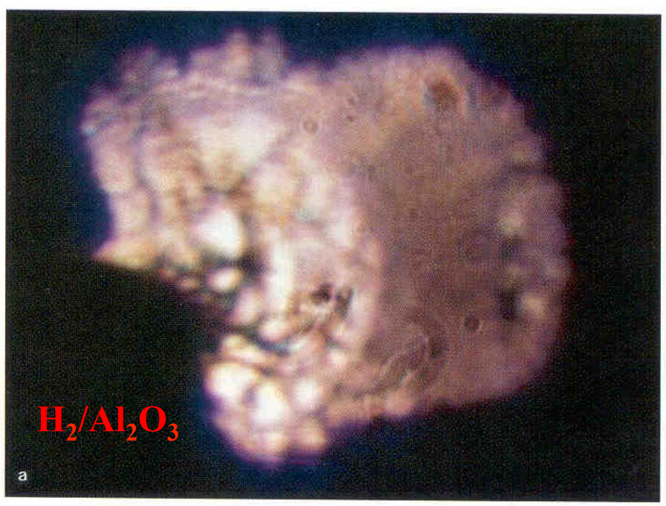
[Friedli & Ashcroft, *Phys. Rev. B* (1977);
[Ramaker et al., *Phys. Rev. Lett.* (1975)]



342 GPa
[Narayana et al., *Nature* (1998)]



320 GPa
[Loubeyre *et al*. *Nature* (2002)]



>250 GPa
[Mao & Hemley, *Science* (1989), *Rev. Mod. Phys.* (1994)]

- Optical absorption
- Variable sample thickness/ diamond absorption
- Direct gap, not indirect gap

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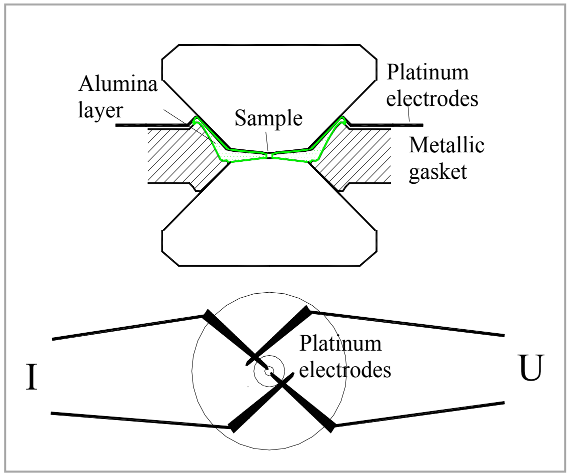
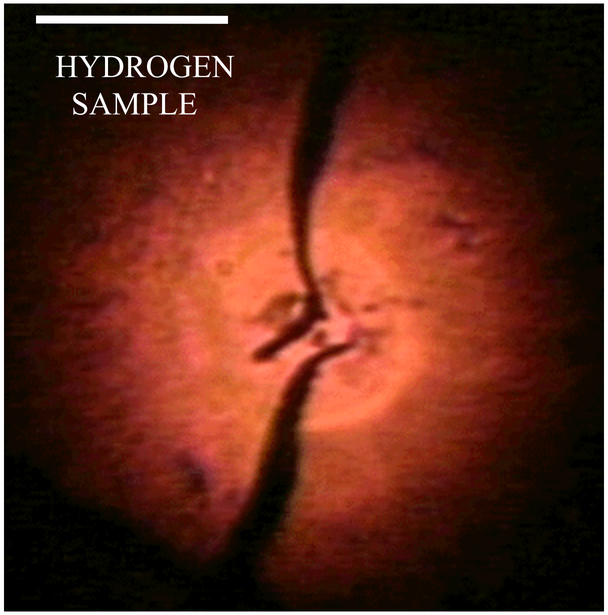
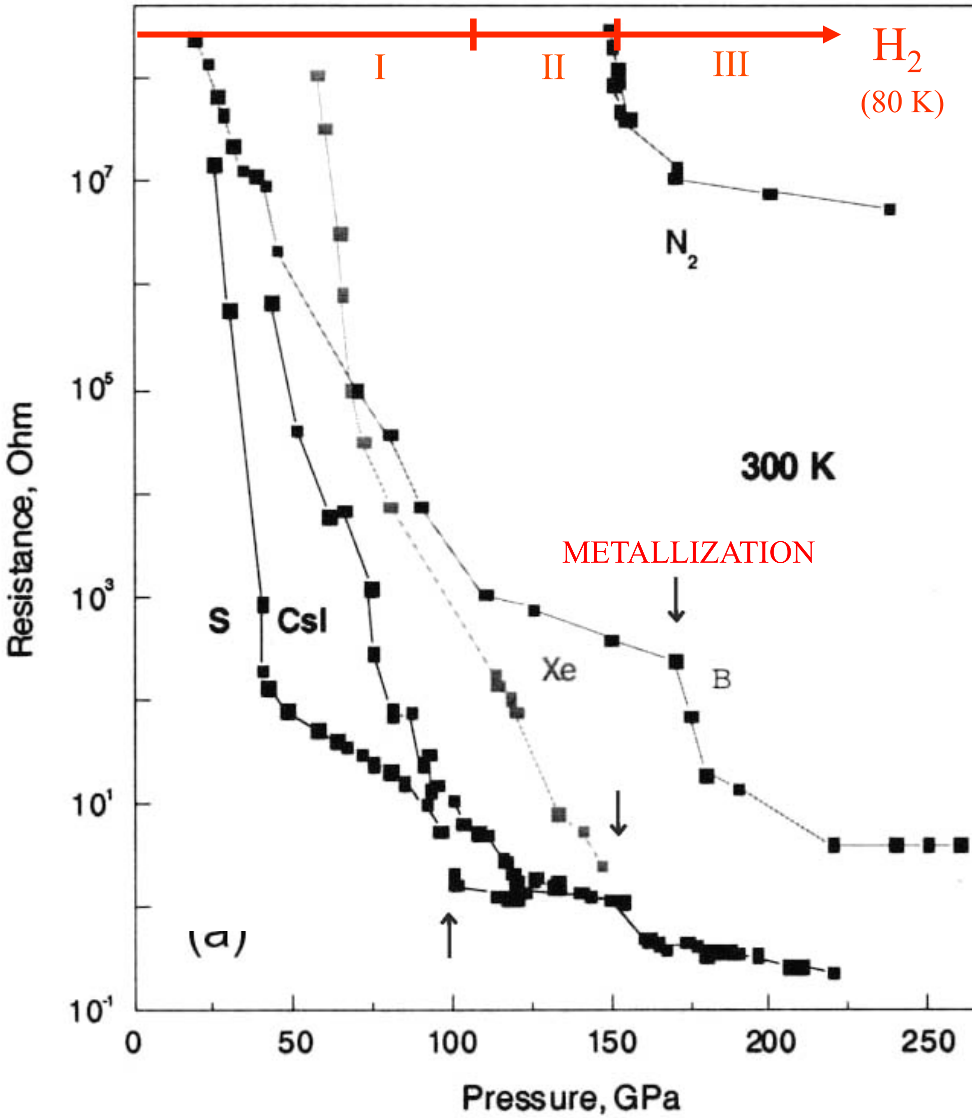
Early Electrical Transport Measurements



40 μm

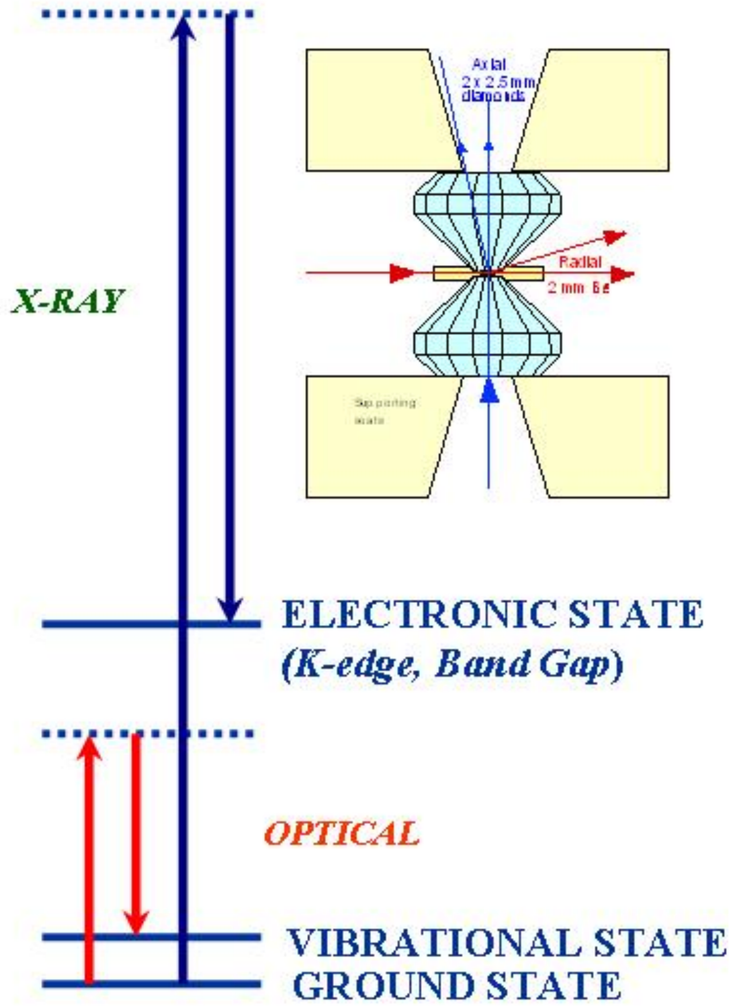


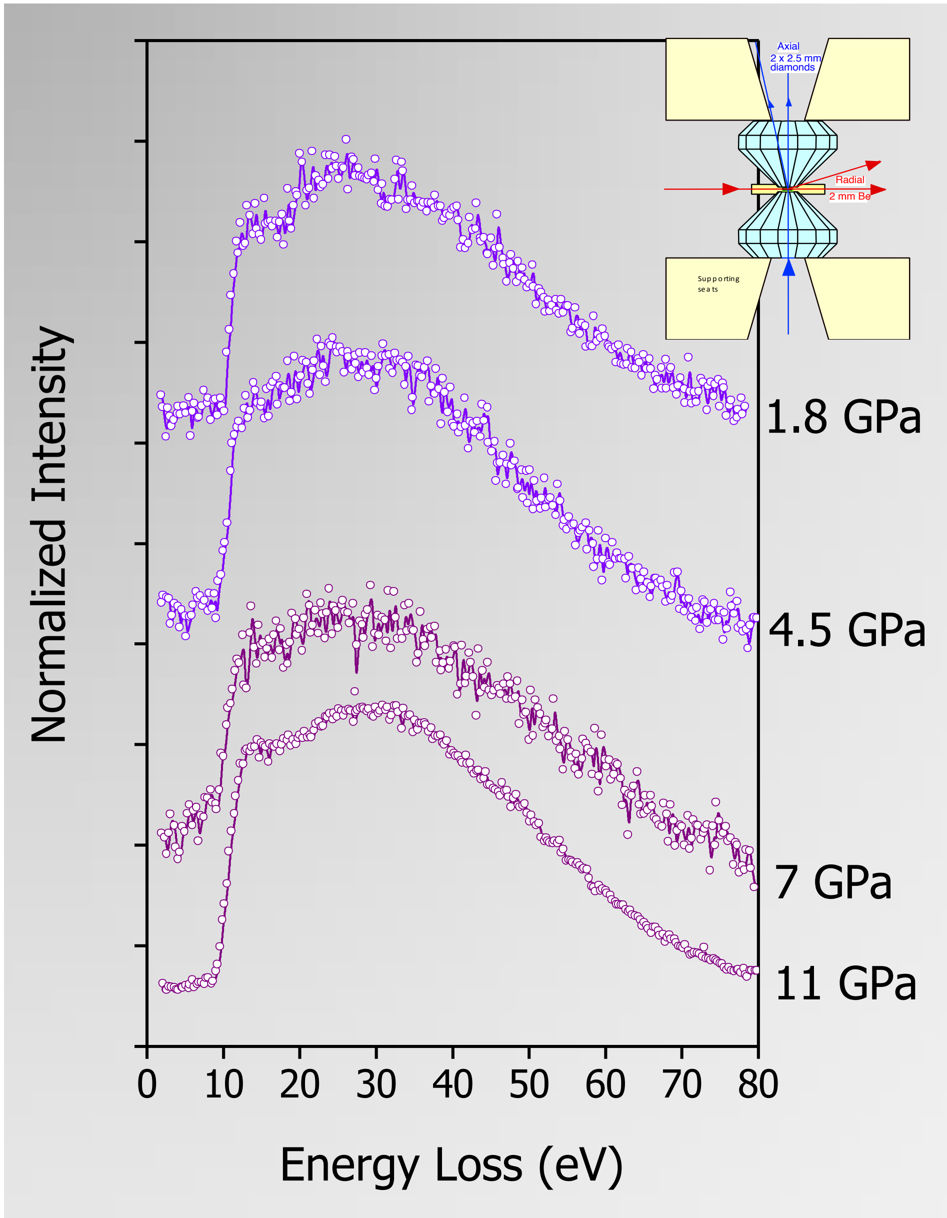


[Eremets *et al.*, *Frontiers in High-Pressure Science* (2002)]

Carnegie Institution

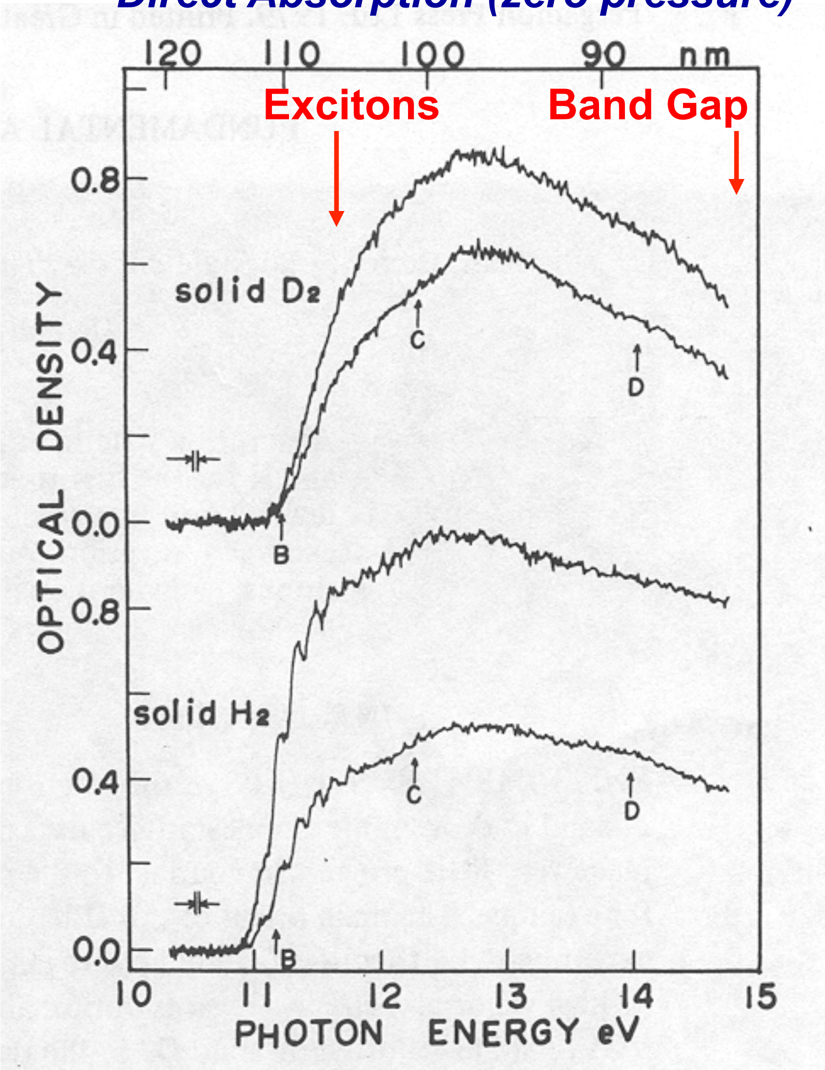
X-Ray Raman of dense hydrogen: direct measurements of the band gap



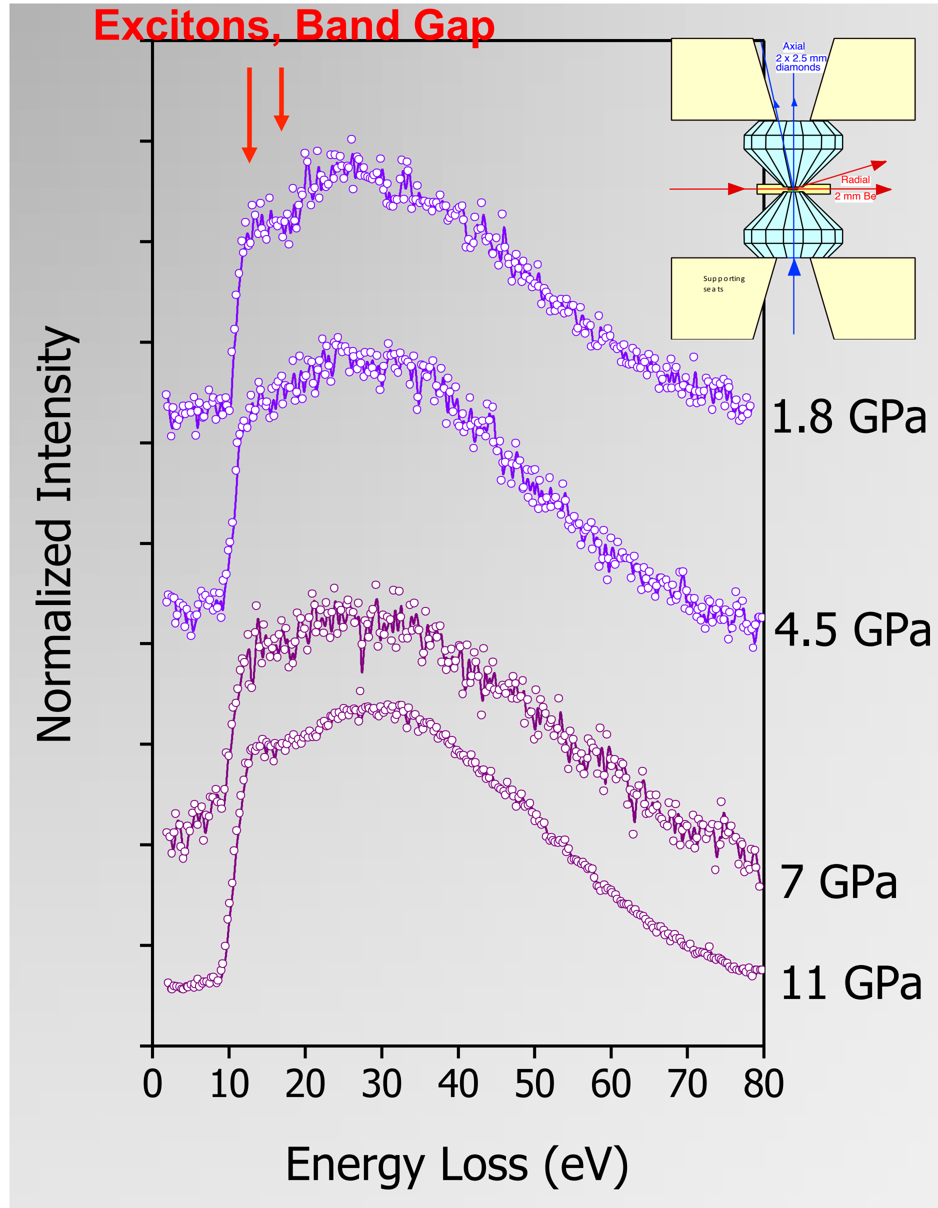


X-Ray Raman of dense hydrogen: direct measurements of the band gap

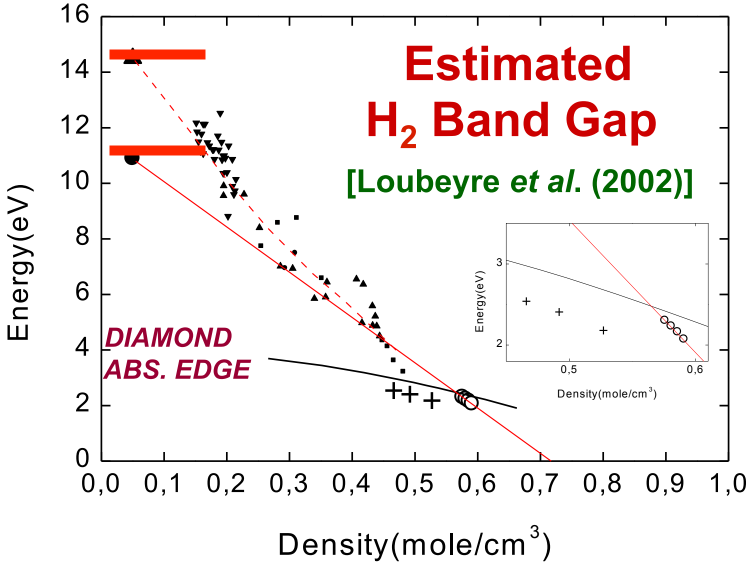
Direct Absorption (zero pressure)

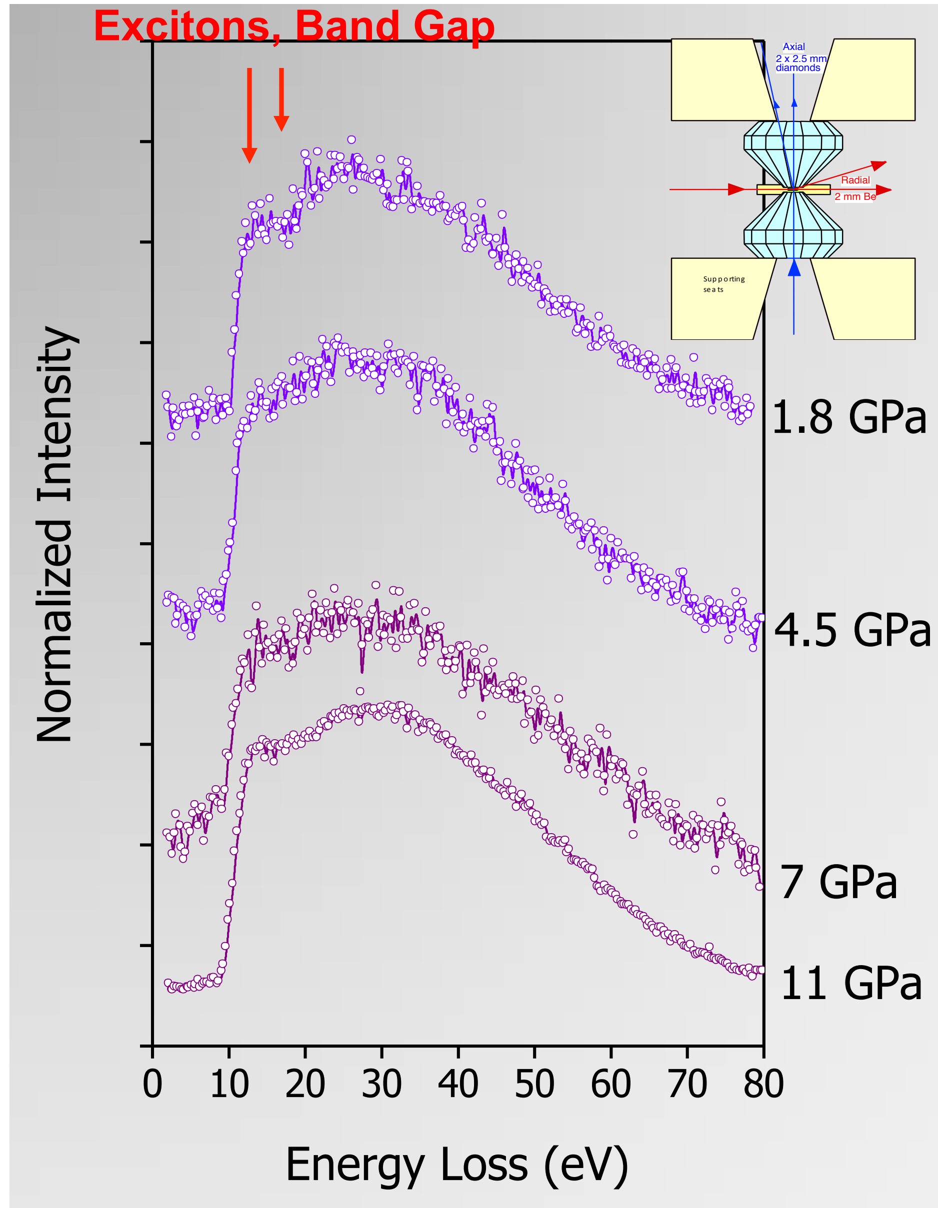


[Inoue *et al*. (1979)]



X-Ray Raman of dense hydrogen: direct measurements of the band gap





I. Introduction

II. Isolated Molecule/Zero Pressure

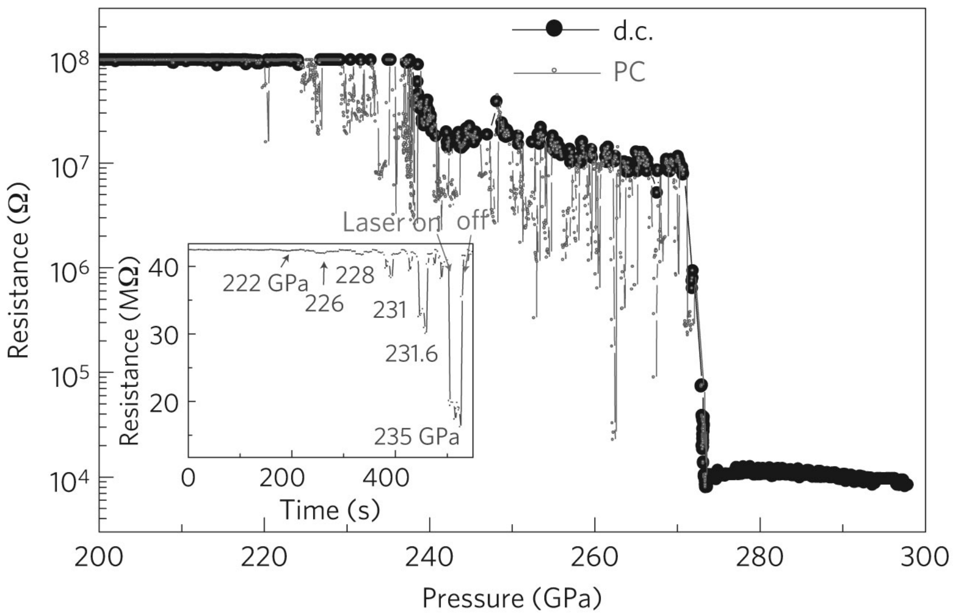
III. Hydrogen under Pressure

IV. New Phases

V. High Pressures and Temperatures

Evidence for new transitions

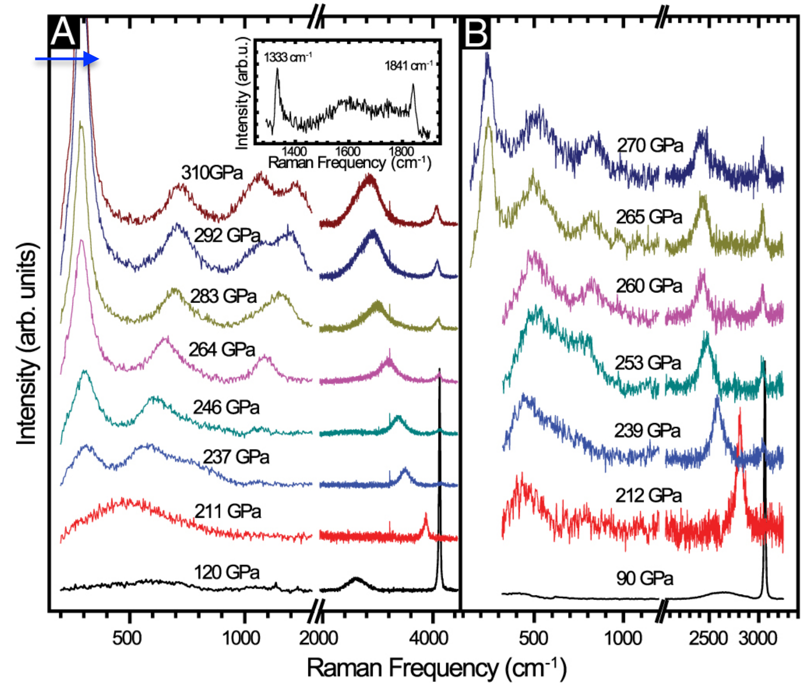
Electrical Conductivity Onset



[Eremets & Troyan, *Nature Materials* (2011)]

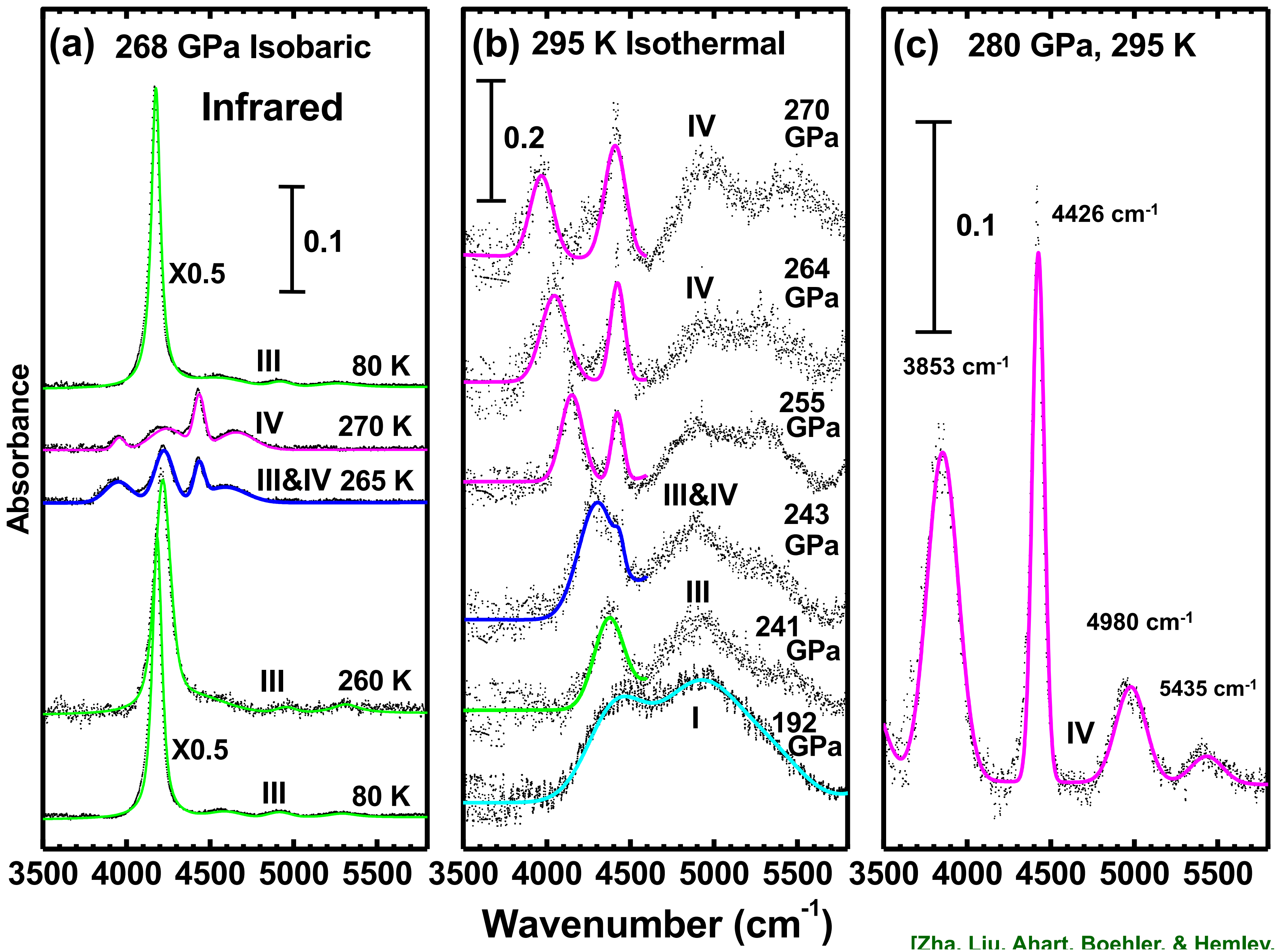
Raman Evidence for New Phase

H_2 D_2



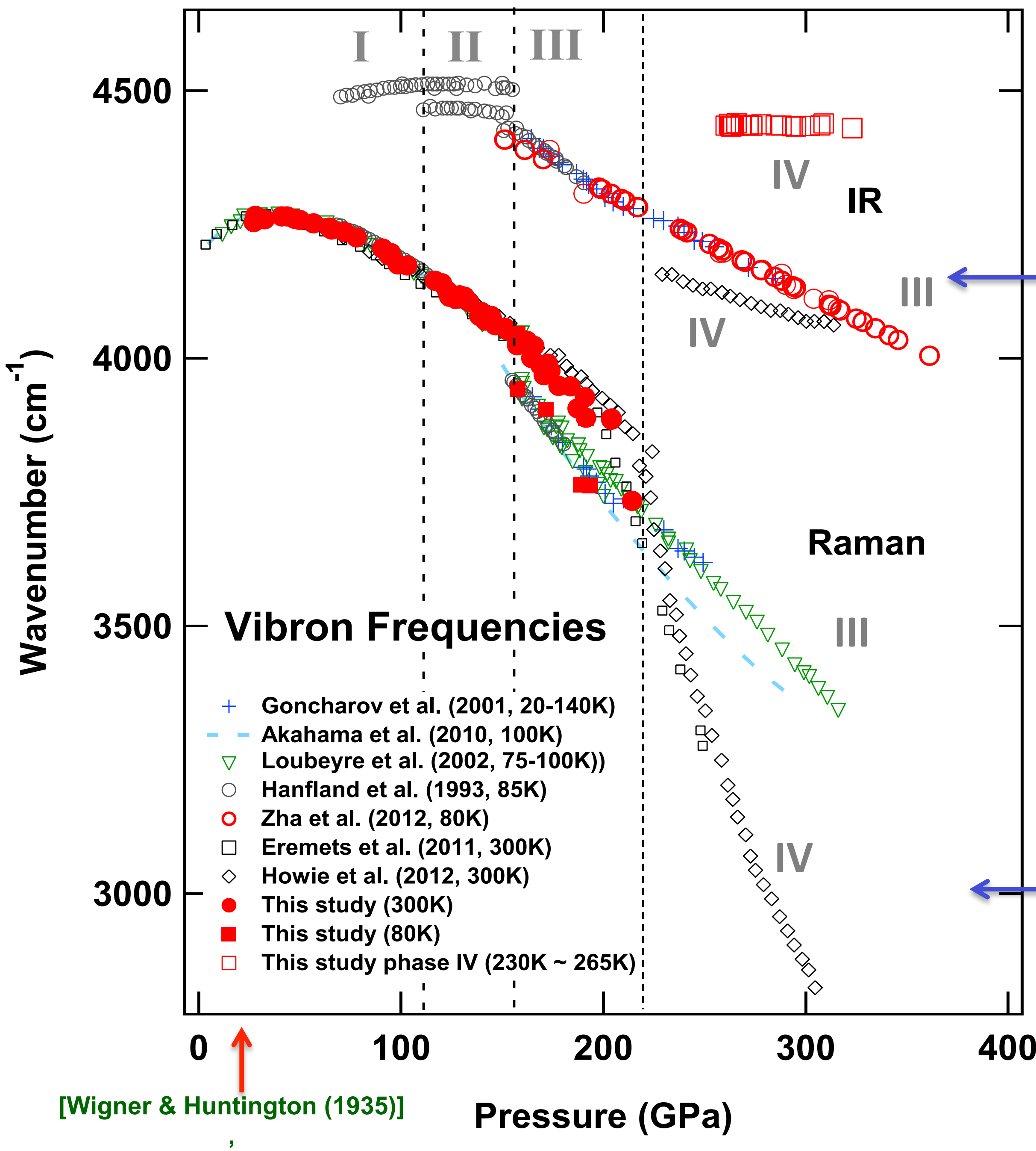
[Howie et al., *Phys. Rev. Lett.* (2012)]

Synchrotron infrared spectroscopy of phase IV



[Zha, Liu, Ahart, Boehler, & Hemley,

Pressure shifts and phase transitions

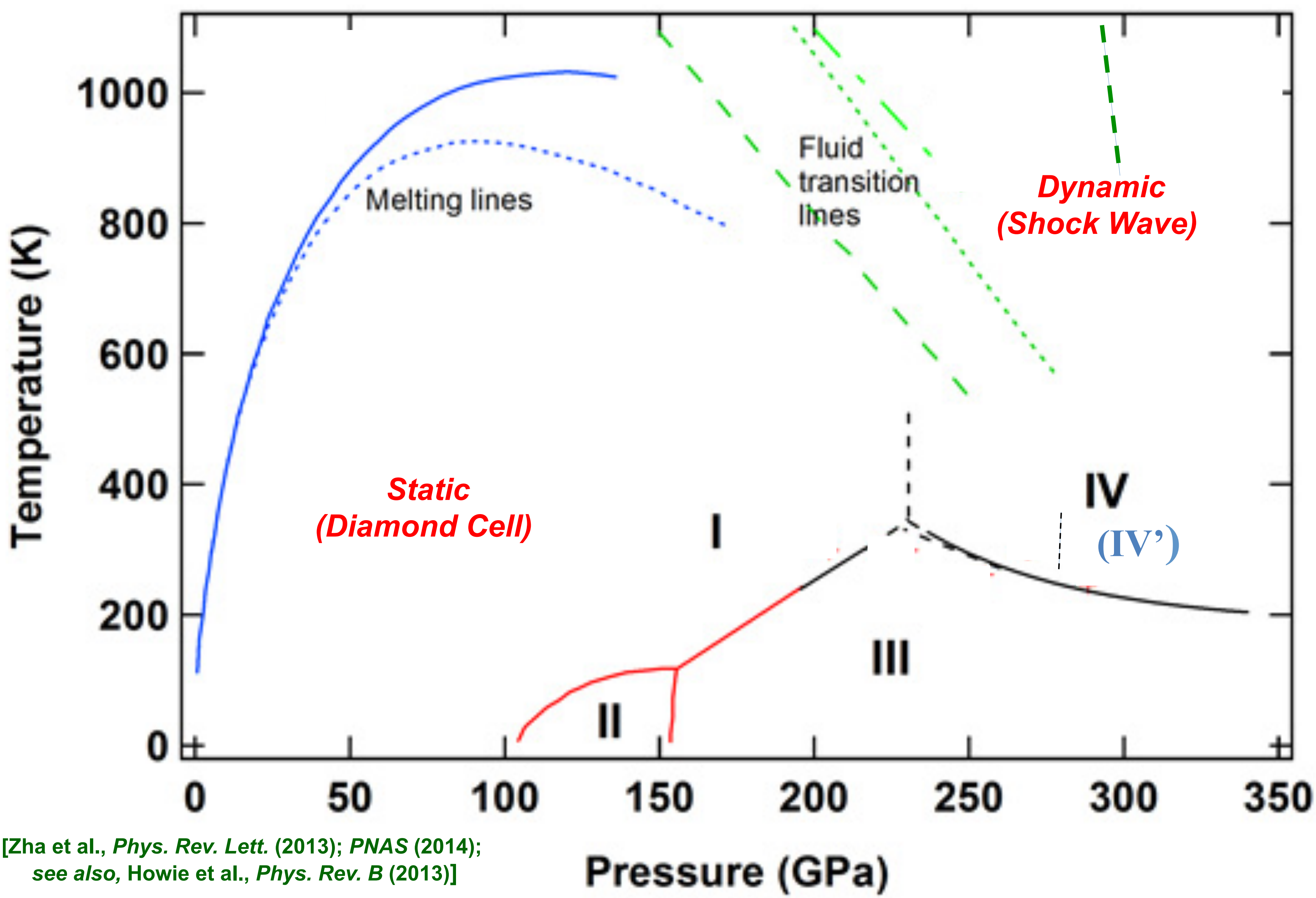


Weakly Interacting H_2

Strongly Interacting H_2

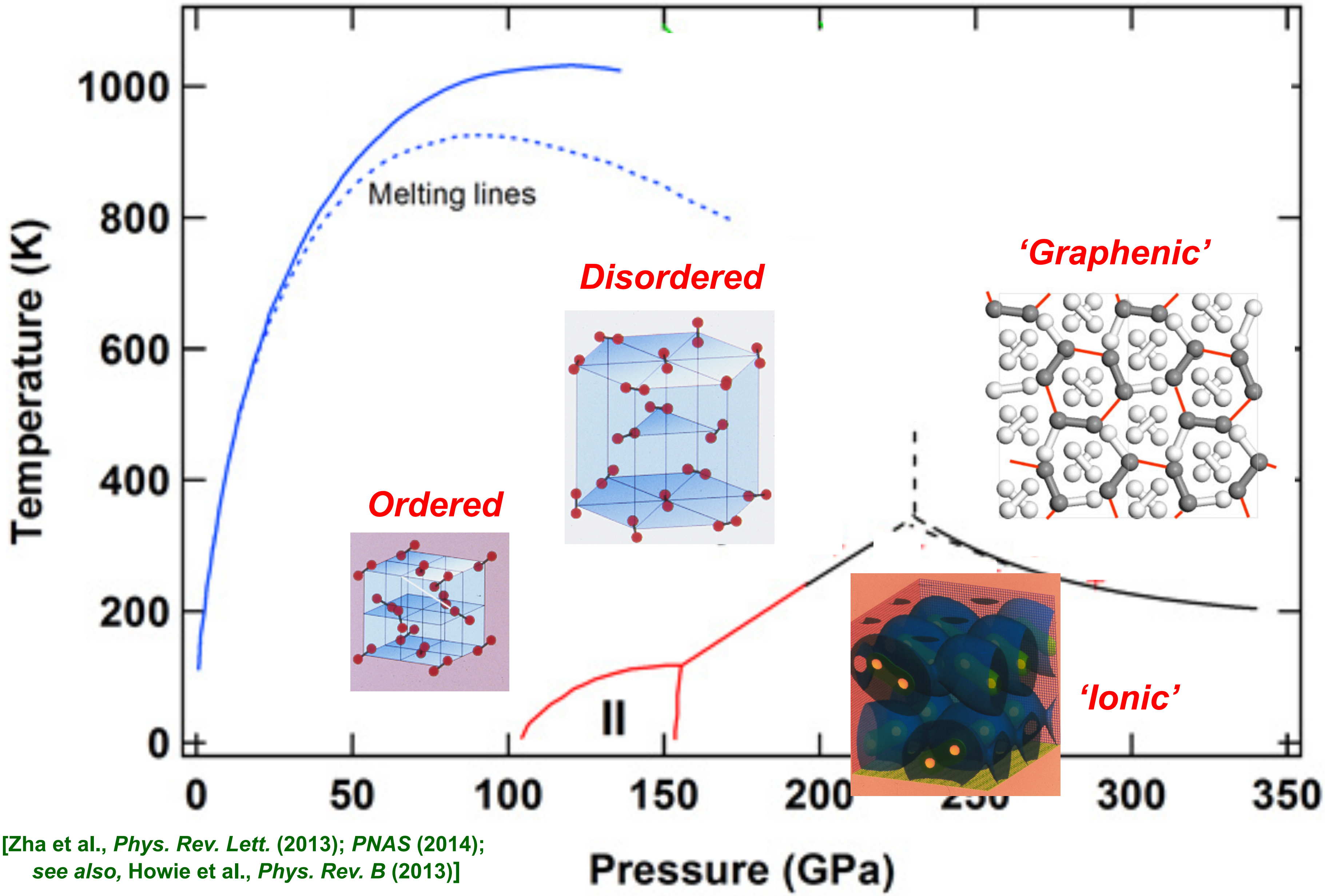
[Zha, Liu, Ahart, Boehler, & Hemley, *Phys. Rev. Lett.* (2013)]

Hydrogen Phase Diagram



[Zha et al., *Phys. Rev. Lett.* (2013); *PNAS* (2014);
see also, Howie et al., *Phys. Rev. B* (2013)]

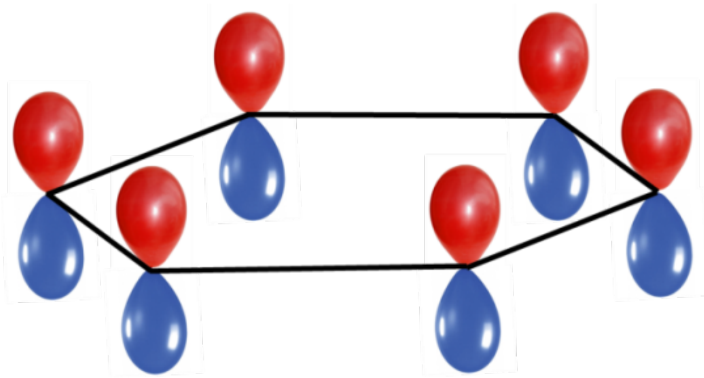
Hydrogen Phase Diagram



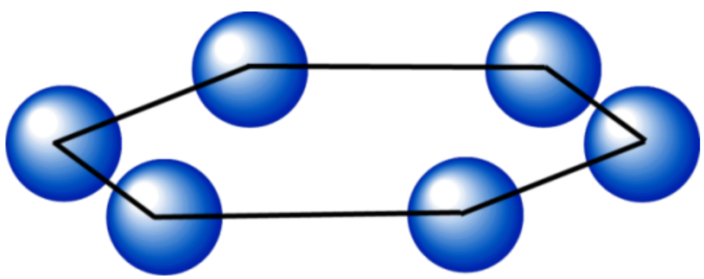
[Zha et al., *Phys. Rev. Lett.* (2013); *PNAS* (2014); *see also,* Howie et al., *Phys. Rev. B* (2013)]

Phase IV is a graphene-based layer structure

C-graphene

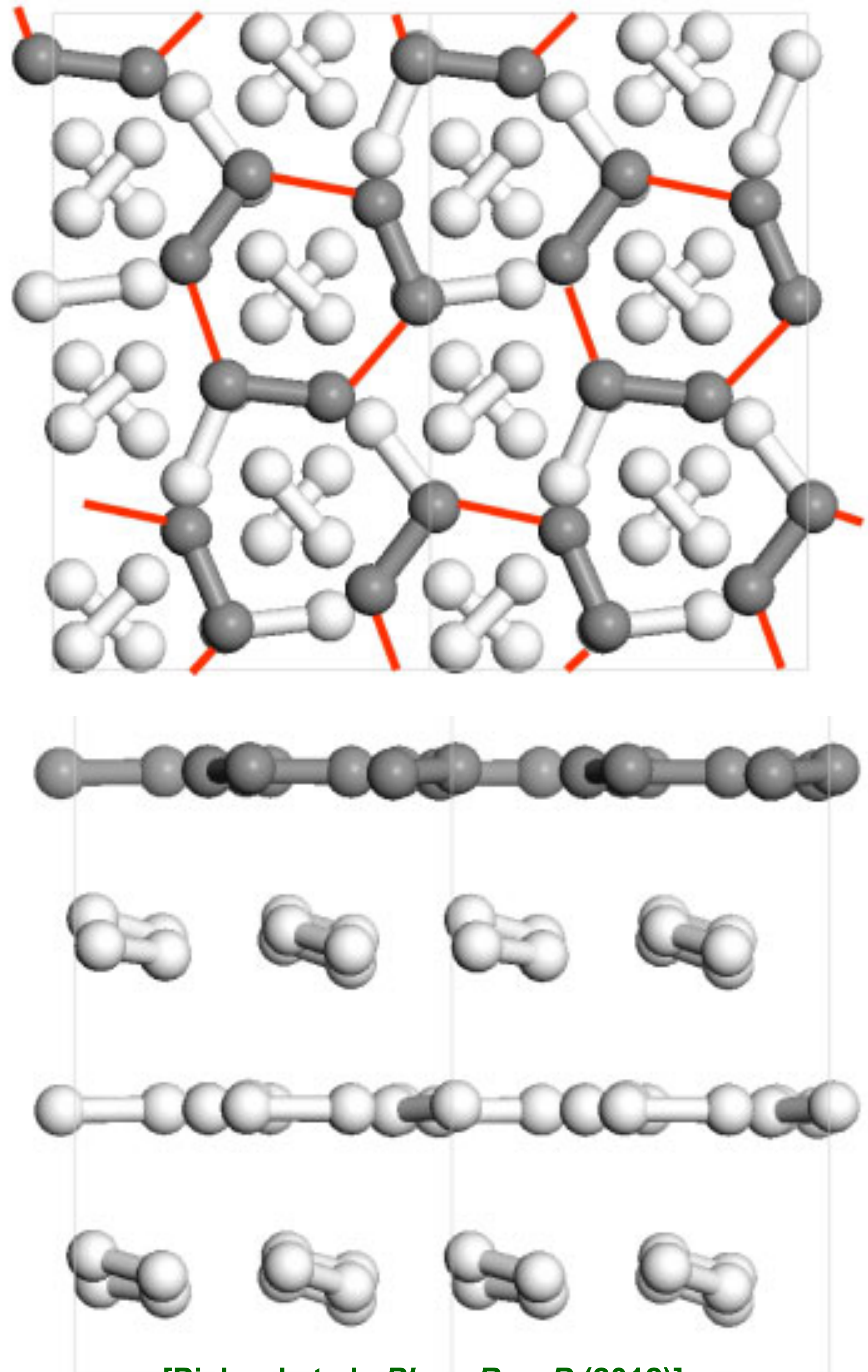


H-graphene



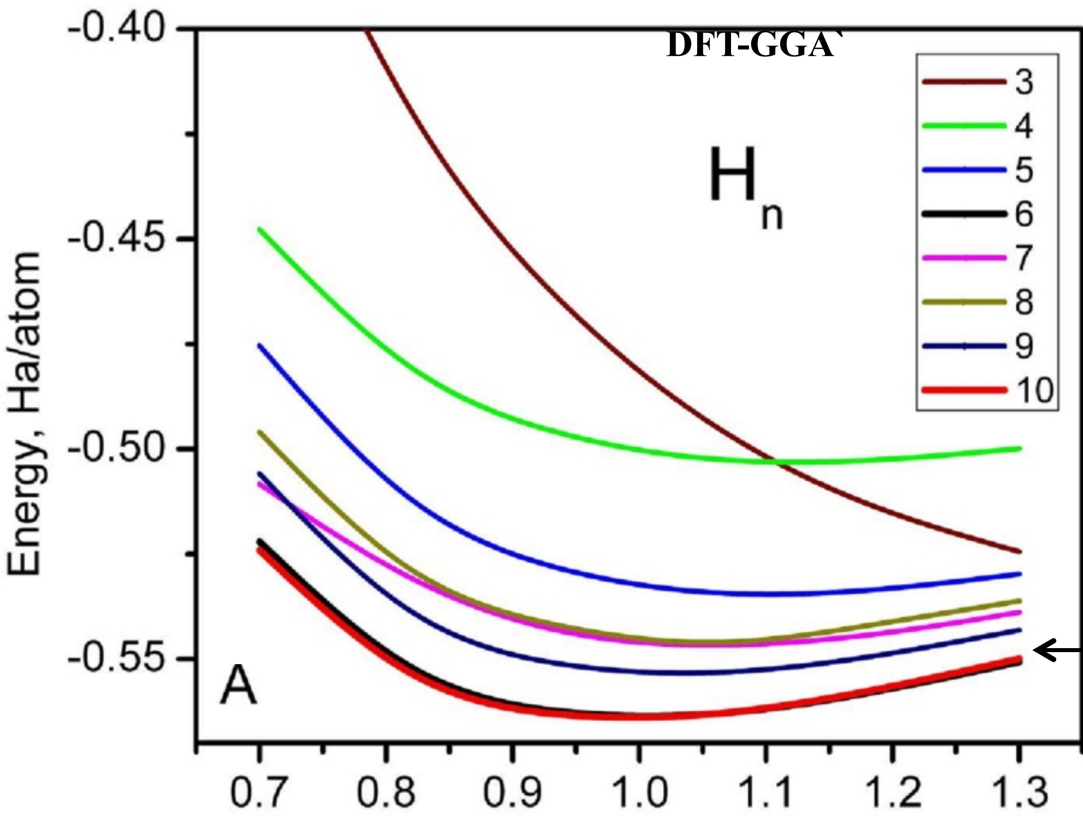
H_6 'aromatic cluster'

[Naumov & Hemley, *Acct. Chem. Res.* (2014);
see also, LeSar & Herschbach, *J. Phys. Chem.* (1981);
Dixon et al., *Faraday Disc.* (1977)]



[Pickard et al., *Phys. Rev. B* (2012)]

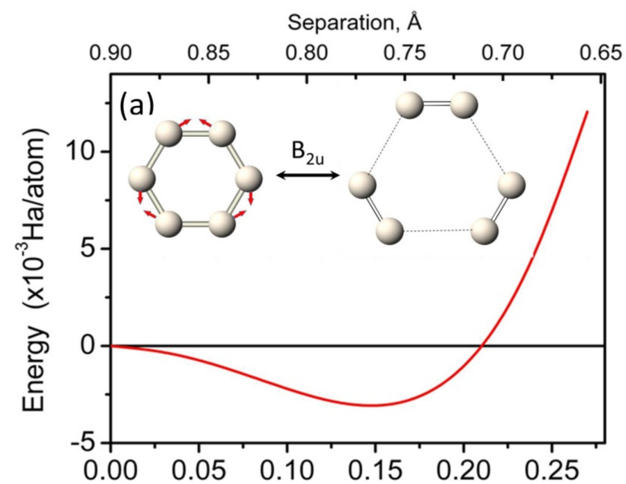
Molecular calculations for H_n rings



Hückel's rule:

Aromatic compounds must have 4n +2 π electrons, n = 0, 1, 2, 3… to fill a π shell

← ***n*** **= 6, 10**



- D_{6h} to D_{3h}
- **Both are aromatic**

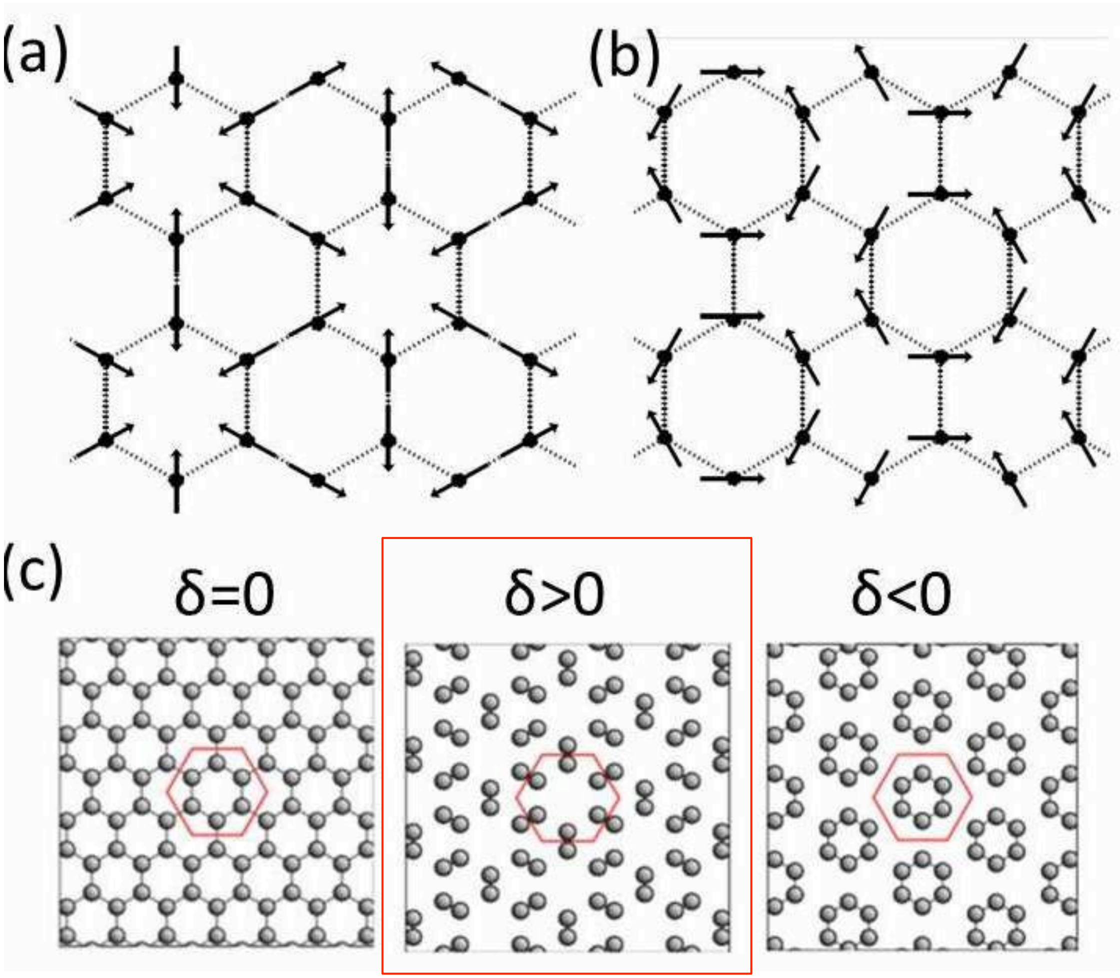
[Naumov & Hemley, *Accts. Chem. Res.* (2014)]

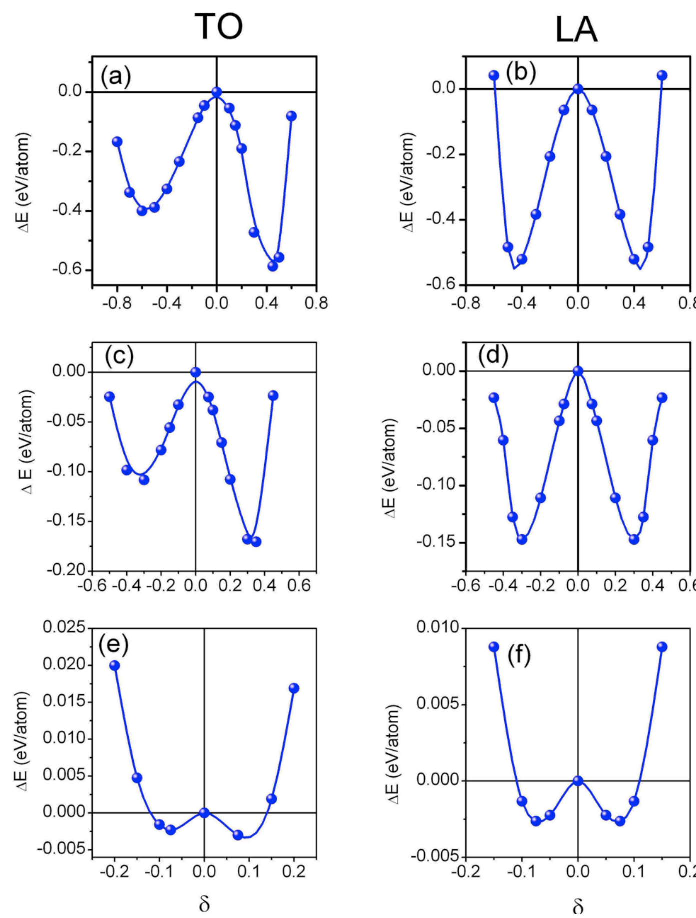
Distortions and relationship to graphene / graphite structures

[Naumov, Cohen & Hemley, *Phys. Rev. B* (2013)]

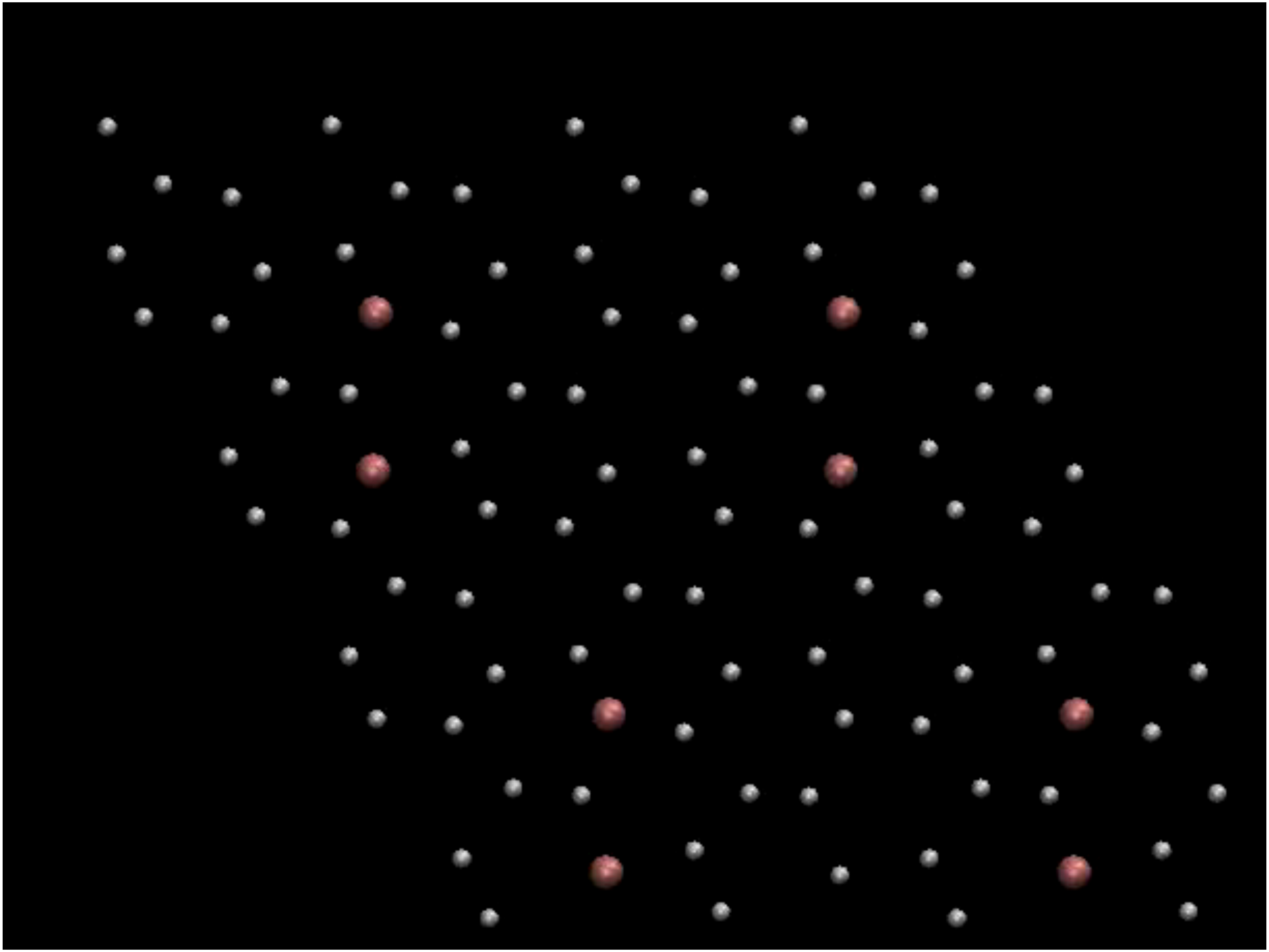
DFT-GGA Calculations

TO LA

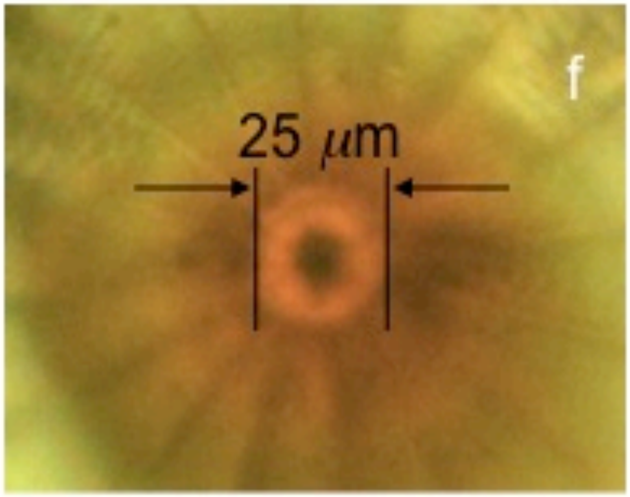




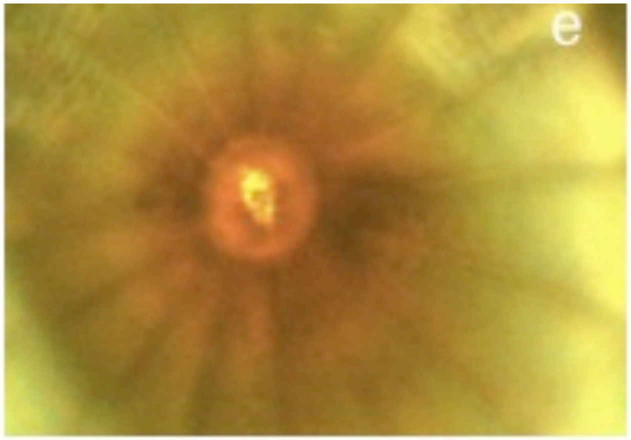
PEIERLS DISTORTION -> Residual Pairing



Is hydrogen metallic at these *P-T* conditions?



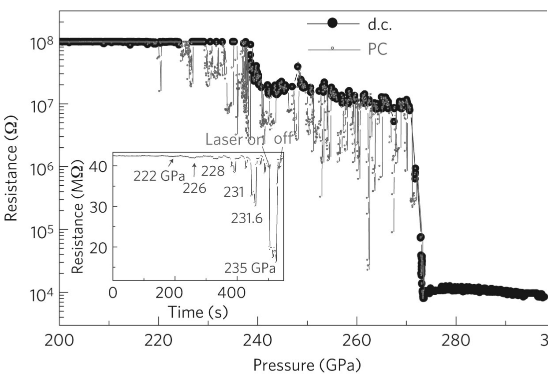
Reflection



Reflection/
Transmission

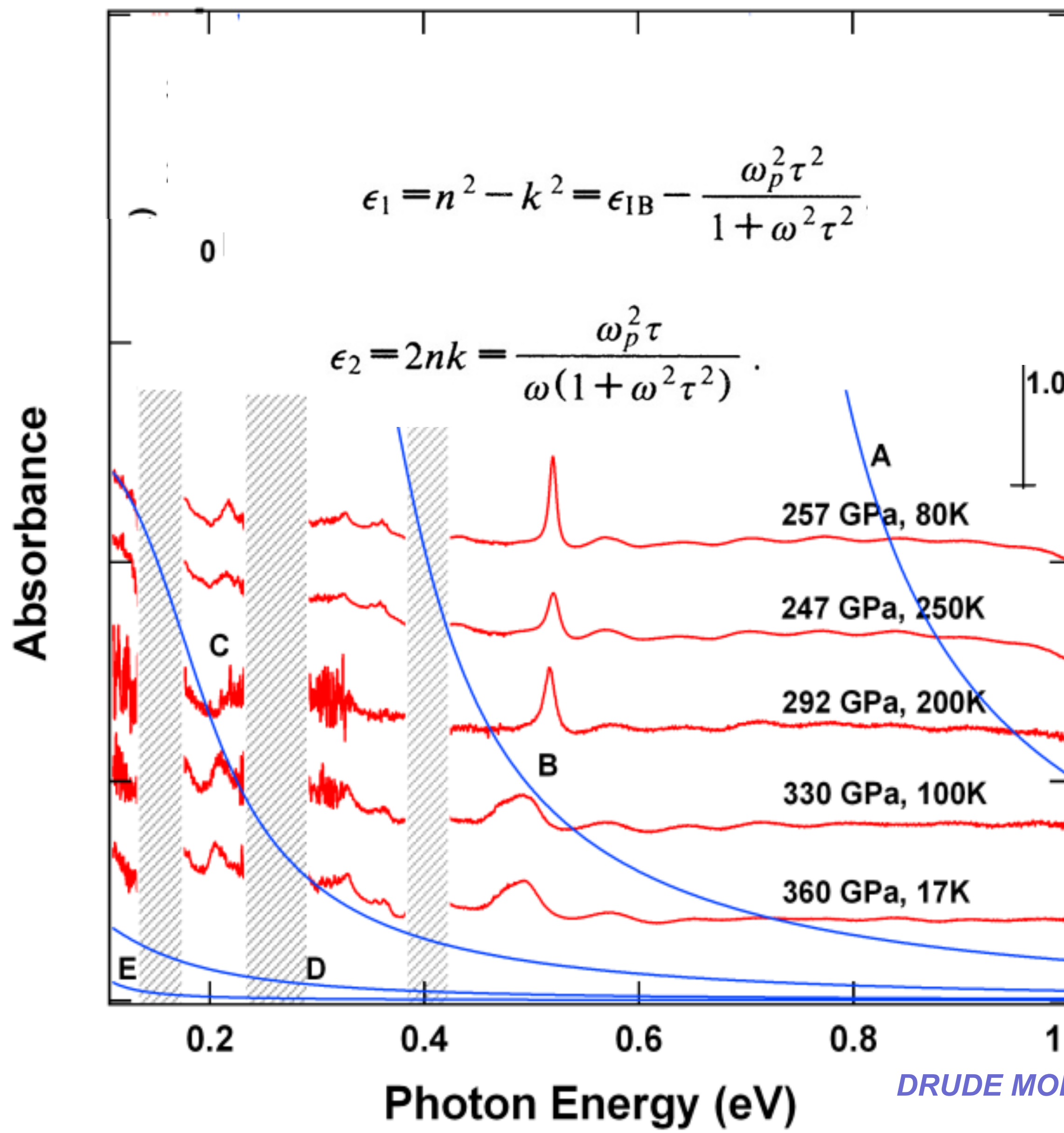
360 GPa, 292 K

Is hydrogen metallic at these *P-T* conditions?



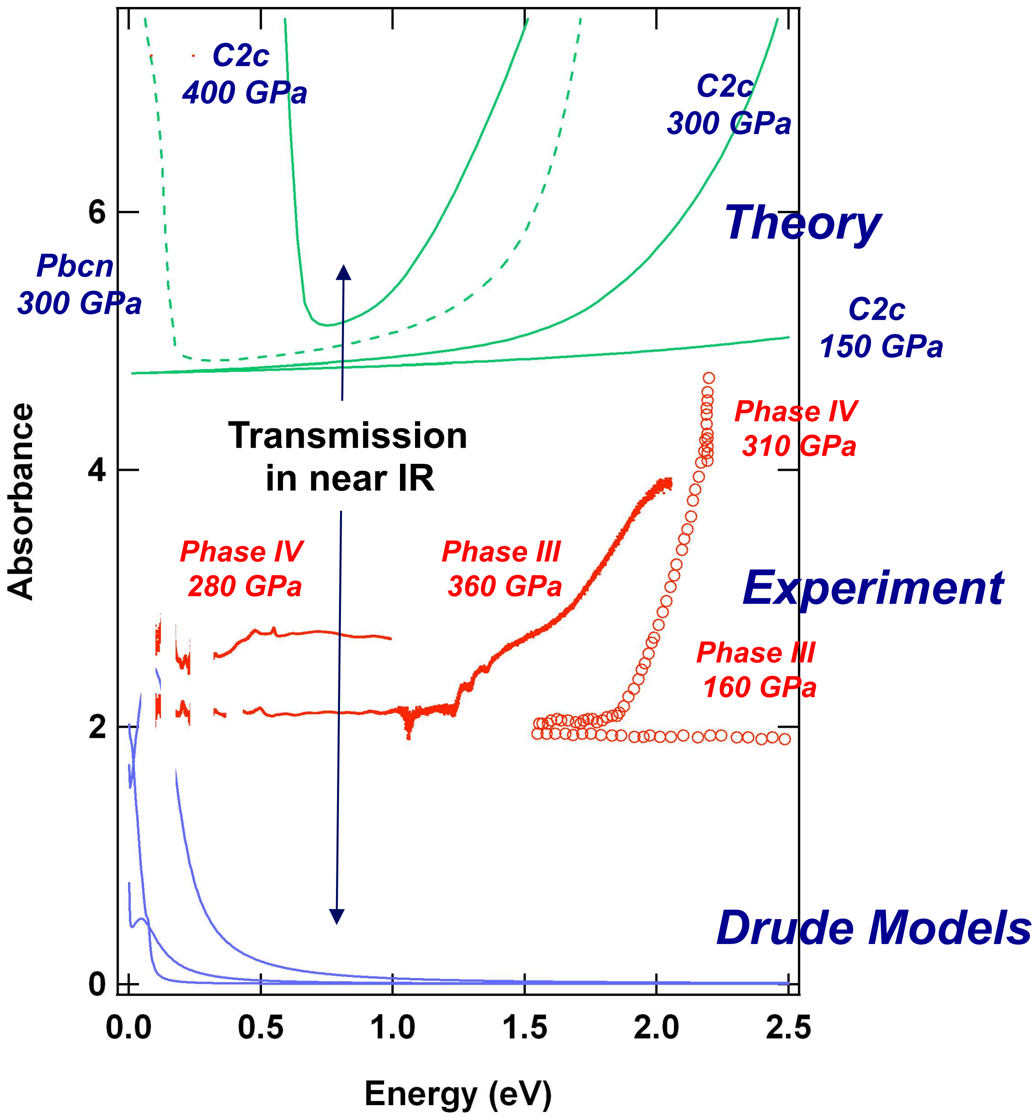
[Eremets & Troyan, *Nature Materials* (2

- Transparent to at least 0.1 eV
- Plasma frequency ω < 0.2 eV
- Semiconducting or semimetallic?
- Transition to semimetal at 270 GPa (phase VI'?)



DRUDE MODELS

A new mechanism for hydrogen metallization

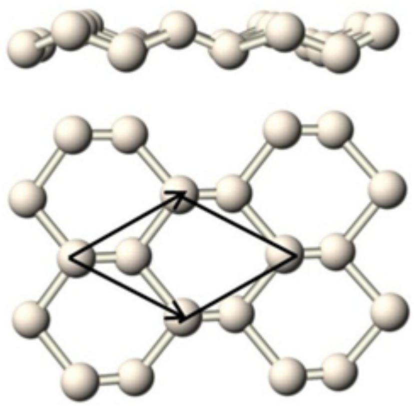


- ***Borderline of semiconductor-semimetal at 300 GPa***
- ***Parallels to graphite (not alkali metals)***
- ***Higher pressures in solid?***

[Cohen, Naumov, & Hemley *PNAS* (2013); Naumov & Hemley, *Acct. Chem. Res.* (2014)]

Other structures of dense hydrogen:
Topological semimetals and surface metallization

***Cmca*-4**

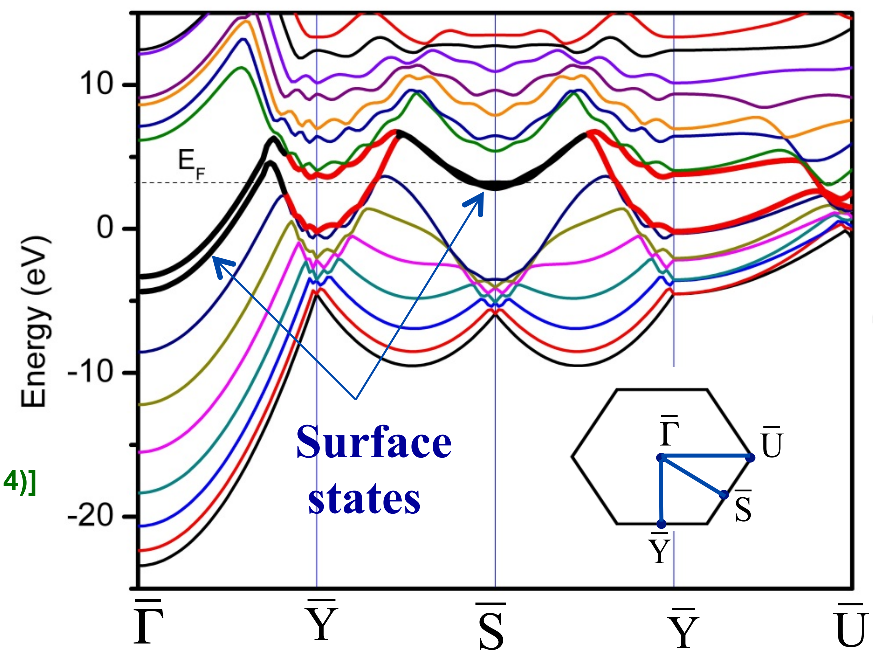


[Edwards & Ashcroft, *PNAS* (2004);
Naumov & Hemley *Acc. Chem. Res.* (2014)]

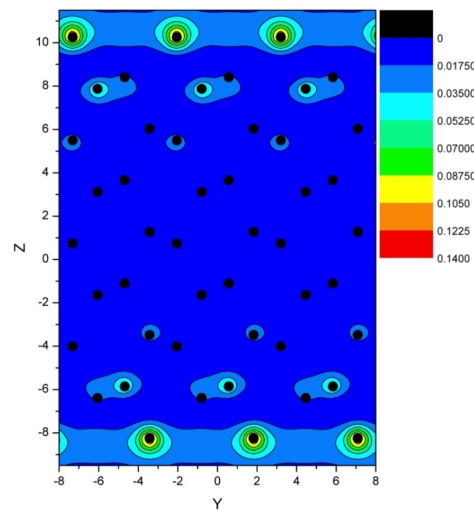
Predicted stable < 300 GPa

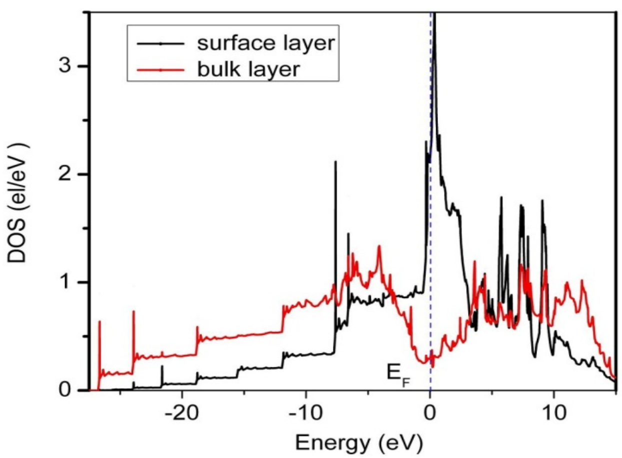
[Liu et al., *J. Chem. Phys.* (2012)]

Band structure of a 8L slab



Charge density of a SS at Γ





Metallic SSs also exist In other candidate structures including *$Pca2_1$* ,*C2/m* and *$Cmc2_1$* .

[Naumov *et al. in preparation*]

Surface (interfacial) Metallization?

Predicted metallic superfluid

PRL **95,** 105301 (2005) PHYSICAL REVIEW LETTERS week ending 2 SEPTEMBER 2005

Observability of a Projected New State of Matter: A Metallic Superfluid

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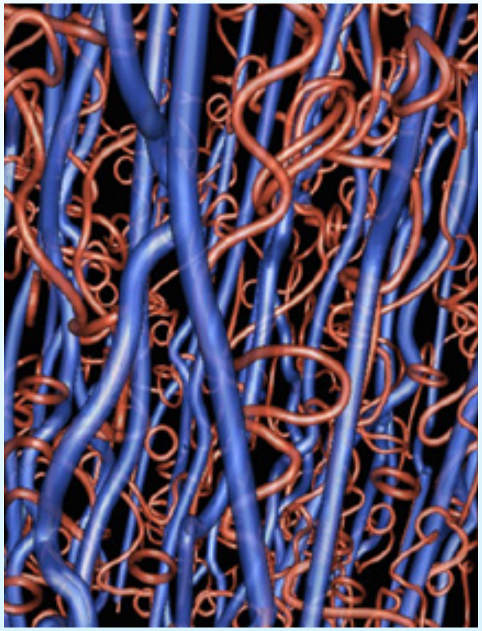
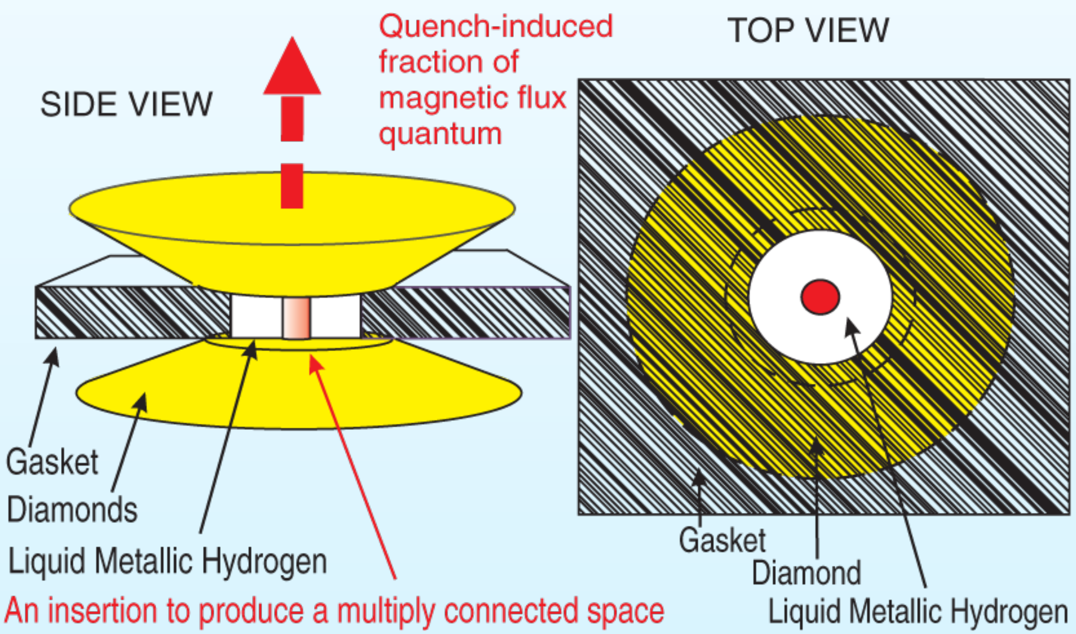
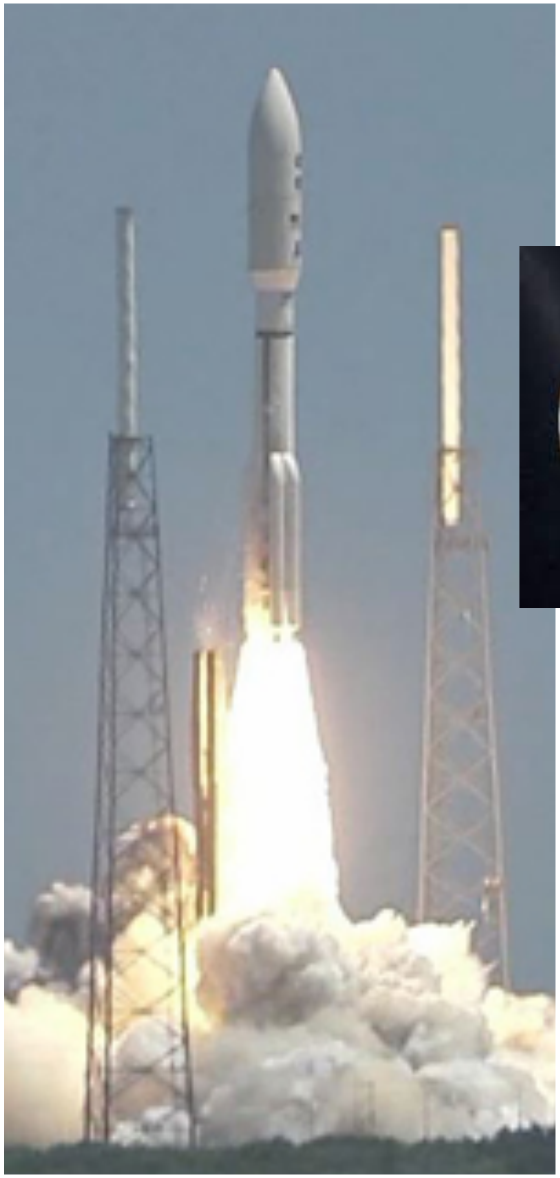
(Received 13 June 2005; published 1 September 2005)





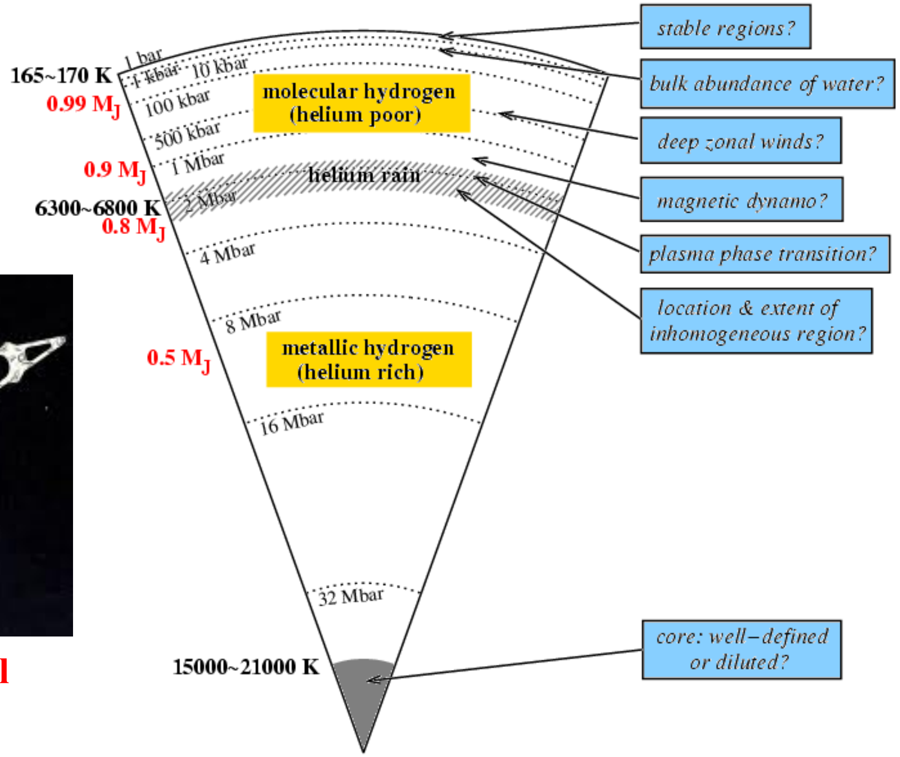
I. *Introduction*
II. *Isolated Molecule/Zero Pressure*
III. *Hydrogen under Pressure*
IV. *New Phases*
V. ***High Pressures and Temperatures***

Juno Mission will let Jupiter tell us



August 5, 2011 Launch

July 14, 2016 Arrival

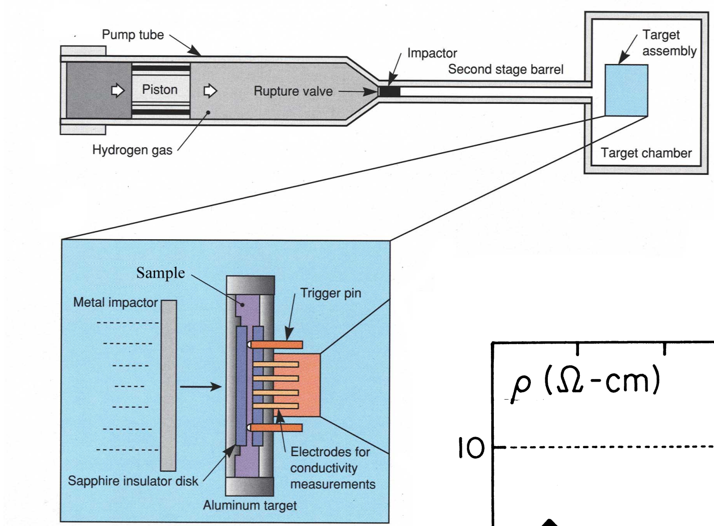


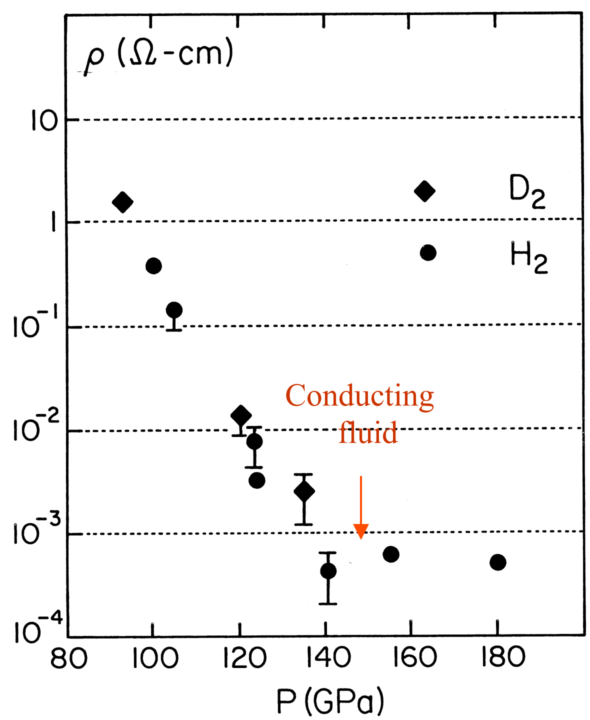
MISSION GOALS

- ***Internal structure?***
- ***Composition (e.g., H_2O)?***
- ***Gravity / magnetic fields?***
- ***Origin and evolution?***
- ***Size and existence of a core?***

DENSE HYDROGEN: Dynamic (Shock) Compression

ELECTRICAL CONDUCTIVTY

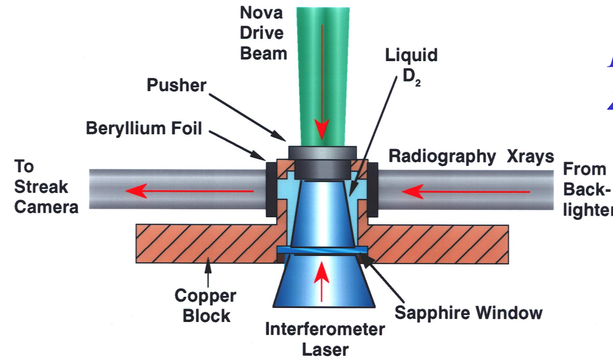


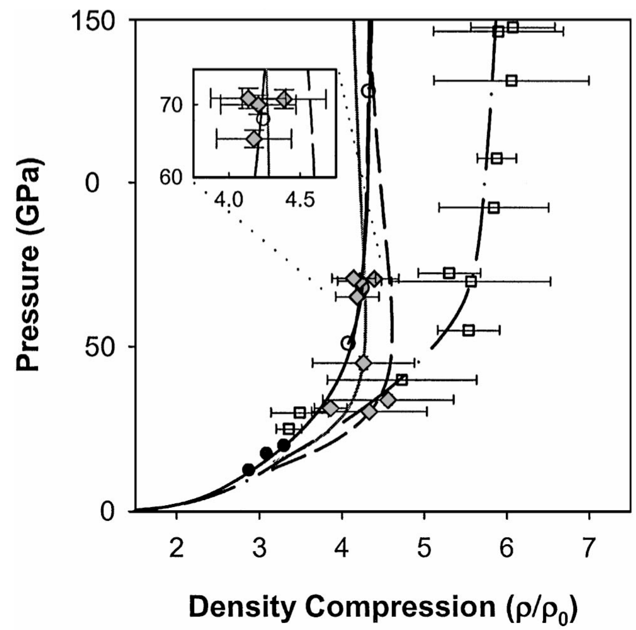


[Weir *et al., Phys. Rev. Lett.* (1996)]

HUGONIOT MEASUREMENTS

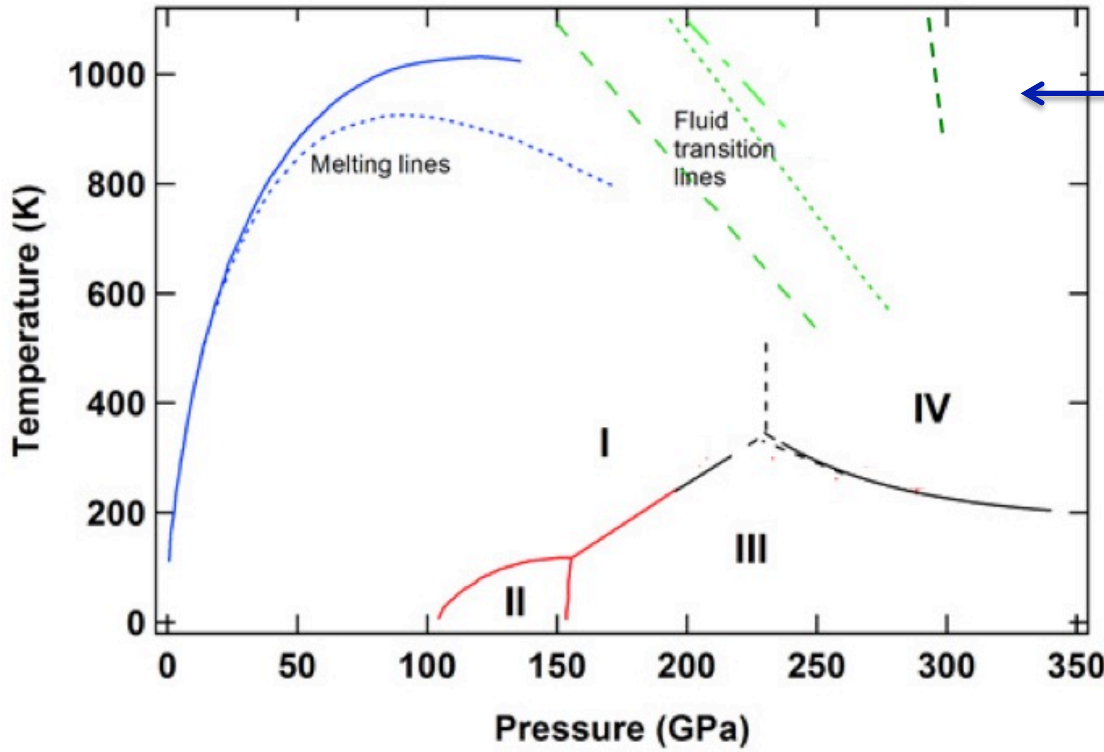
LASER
Z-PINCH

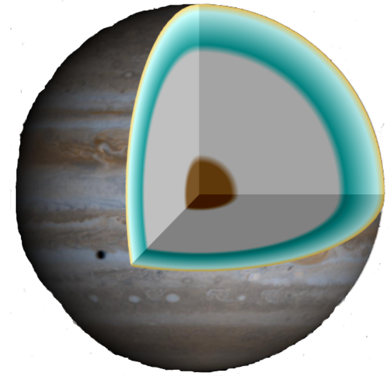




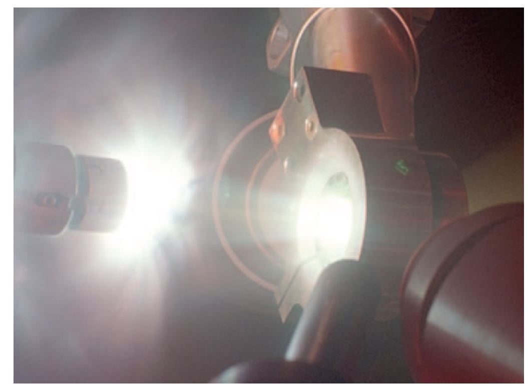
[Cauble *et al., Science* 281, 1178 (1998); Knudson *et al. Phys. Rev. Lett.* 87, 225501 (2001)]

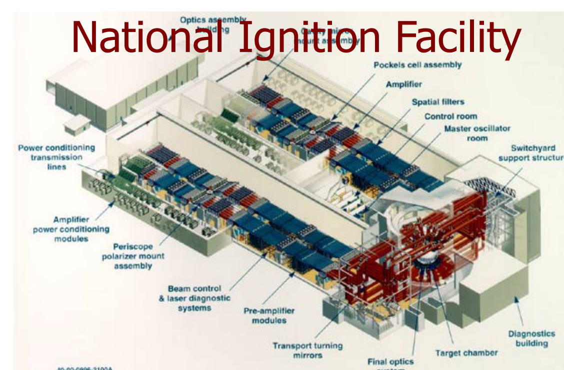
High *P-T* Transition in Fluid Hydrogen





LASER DRIVEN DYNAMIC COMPRESSION





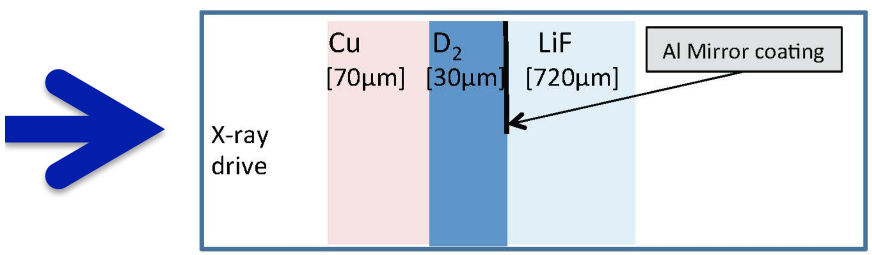
- ***Insulating to Conducting?***
- ***Dissociation: H_2 to H?***
- ***Pressure differences?***
- ***Relationship to the solid transition?***

Carnegie Institution

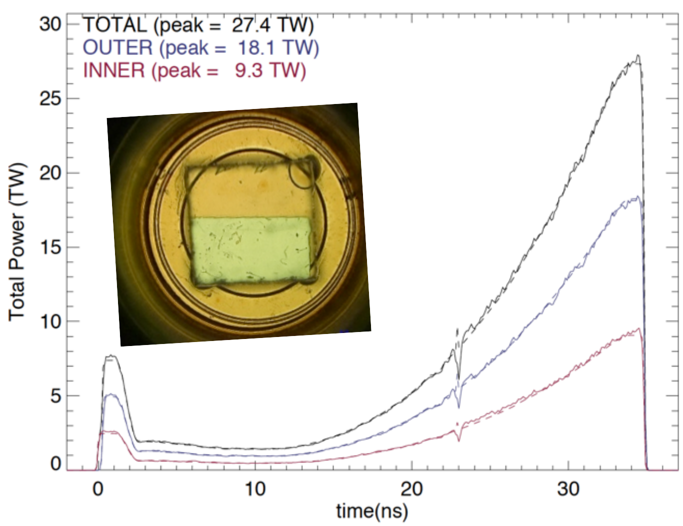
High *P-T* Transition in Fluid Hydrogen

176 beams of the NIF Laser were used,
with a custom 300 kJ - 35 ns long pulse shape

Sample Geometry

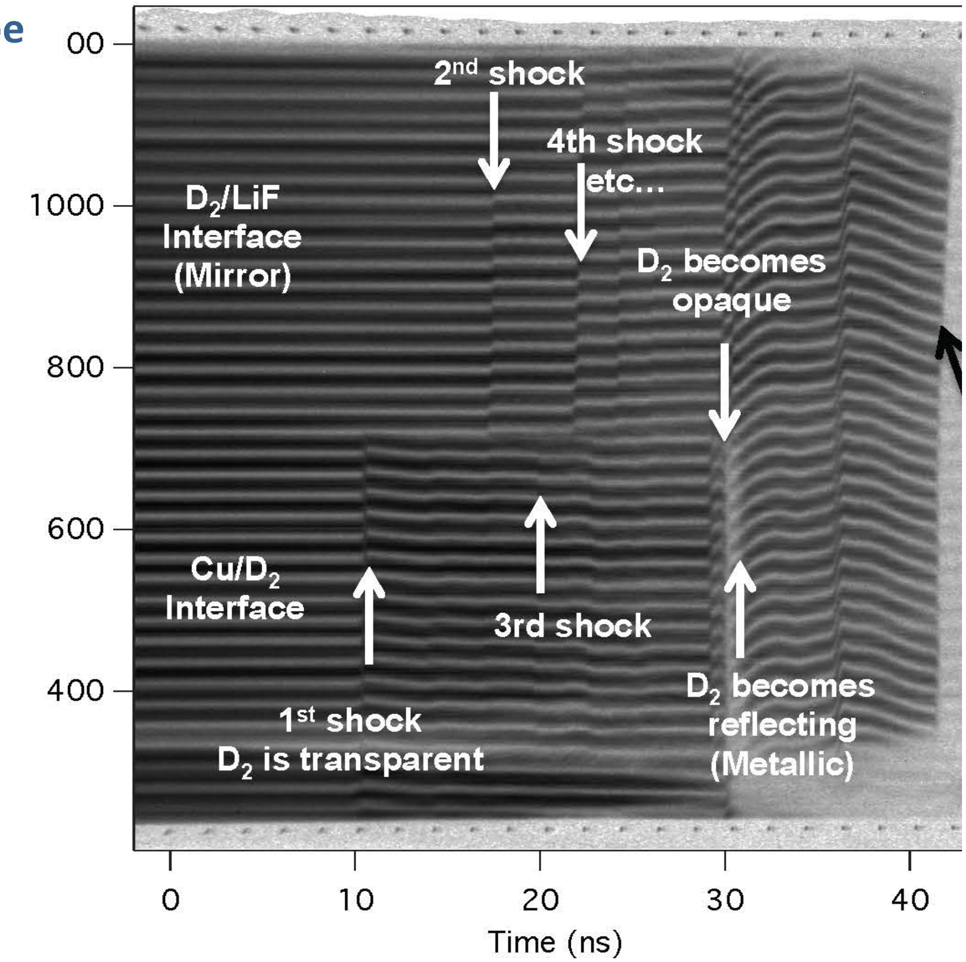


Laser Pulse



[Jeanloz & Hemley (PIs),
NIF Discovery Science Campaign]

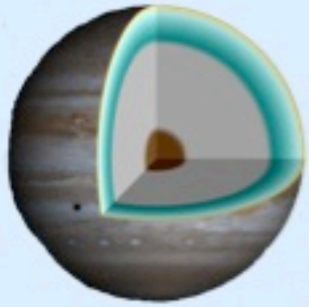
Optical/Velocity (VISAR) Data

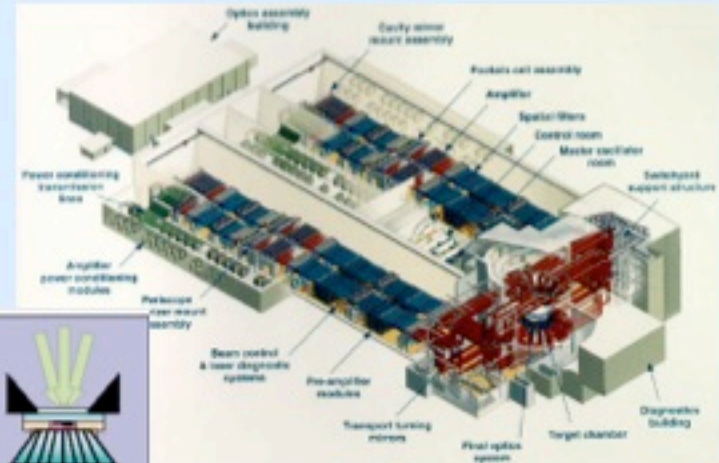


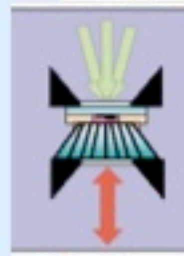
Evidence for changes in the hydrogen optical
properties during the reverberation compression

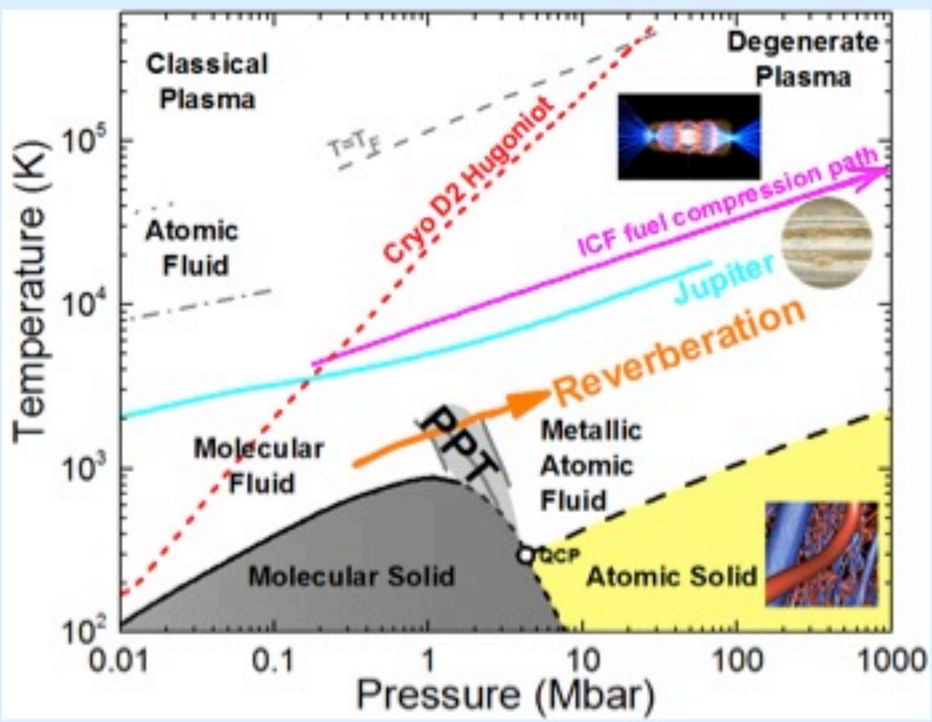
We direct experiments at the National Ignition Facility to understand the materials in ultra-extreme conditions

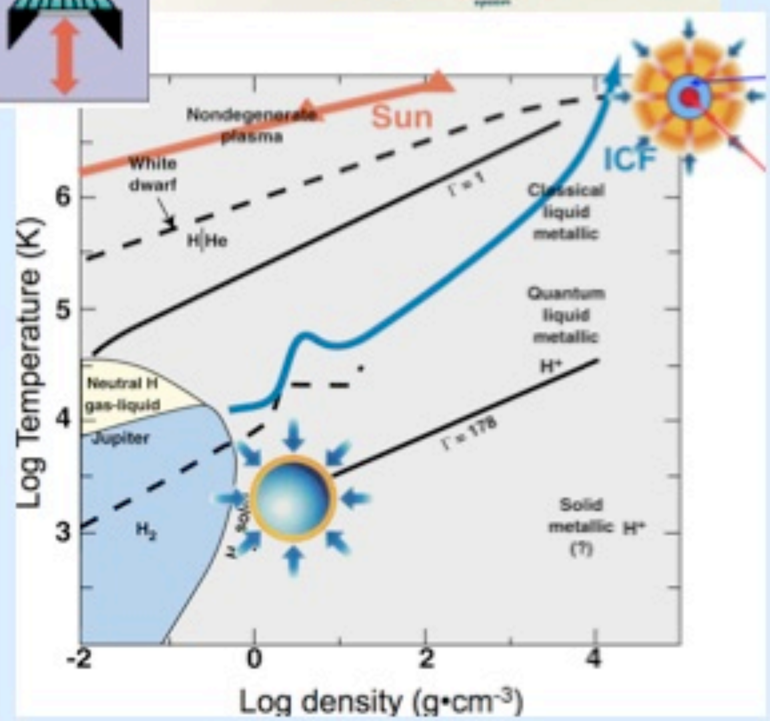
- Conducting transition in fluid of hydrogen at megabar pressures?
- Depth of magnetic field generation in Jupiter ?
- Melting curve of iron in exoplanets (>20 Mbar)











CONCLUSIONS AND OUTLOOK

1. **Dense hydrogen is a system of unexpected complexity.**
2. **There is no sign of the 'Wigner-Huntington metallization' to 340 GPa in phases III and IV (below 300 K).**
3. **The structure of phase IV is broadly consistent with the structures predicted theoretically.**
4. **There is a remarkable parallel between dense hydrogen and graphene that reveals a new mechanism for metallization.**
5. **New dynamic compression results reveal a transition in fluid starting at 140 GPa.**

ACKNOWLEDGEMENTS

Collaborators

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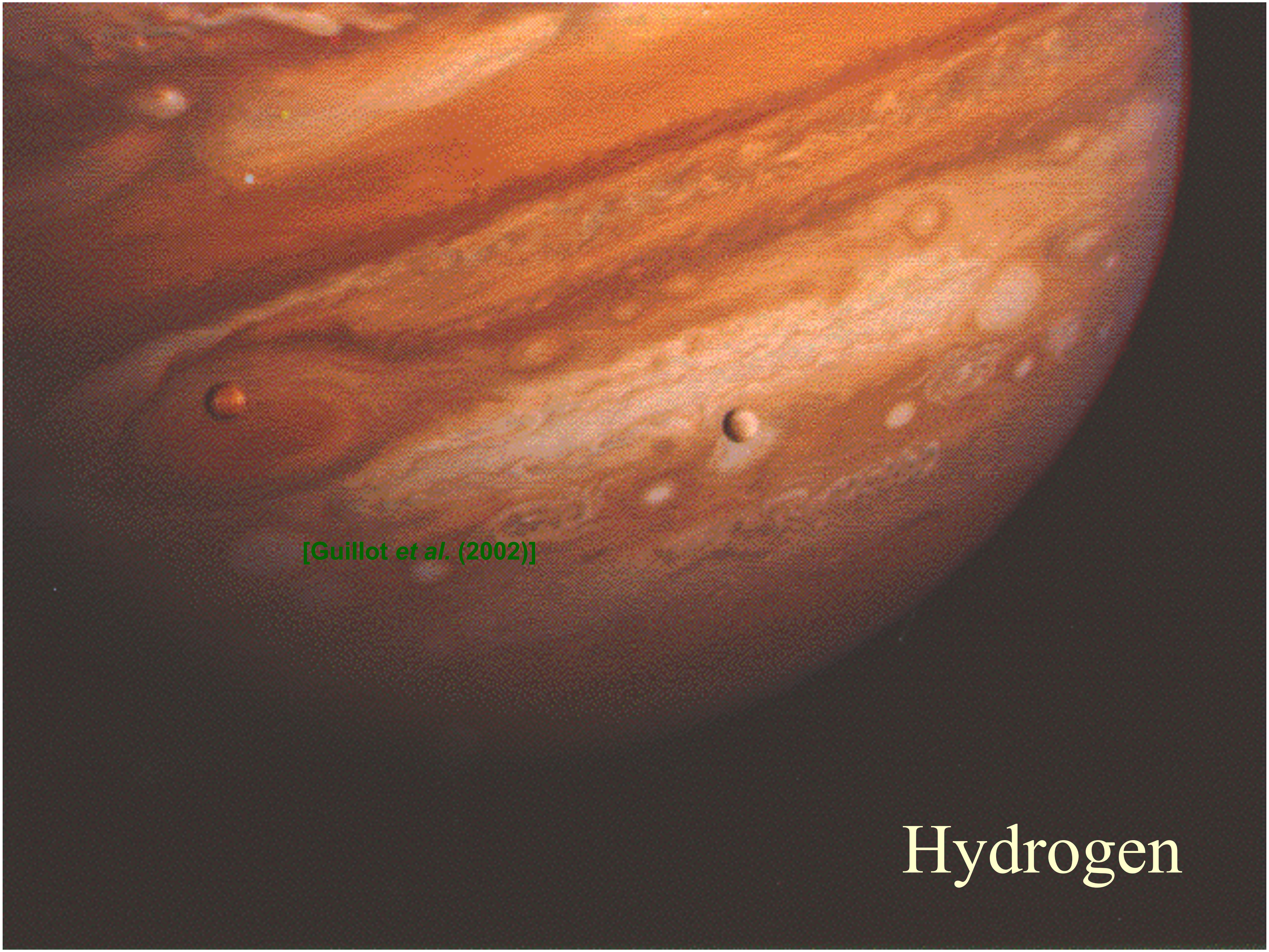
[Guillot *et al.* (2002)]
Hydrogen