A First-order Phase Transition to Metallic Hydrogen (and more)

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Ashkan Salamat Postdoc
Now at UNLV faculty

Ori Noked Postdoc
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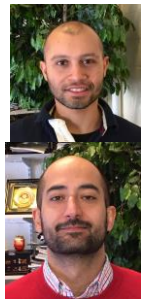
Rachel Husband Postdoc



This reseach

Recent Ph.Ds.

Vasya Dzyabura
Jieping Fang

Recent undergraduate Students

Armin Pourshafeie to Stanford
Luke Burkhart to Yale

Research Areas:

- **High pressure with DACs: Metallic Hydrogen**
- **Measurement of the neutron electric dipole moment**

Former Students now in NNSA Labs:

Jon Eggert	LLNL
Will Evans	LLNL
Hector Lorenzana	LLNL

SSAA

Todd Ditmire	U of T

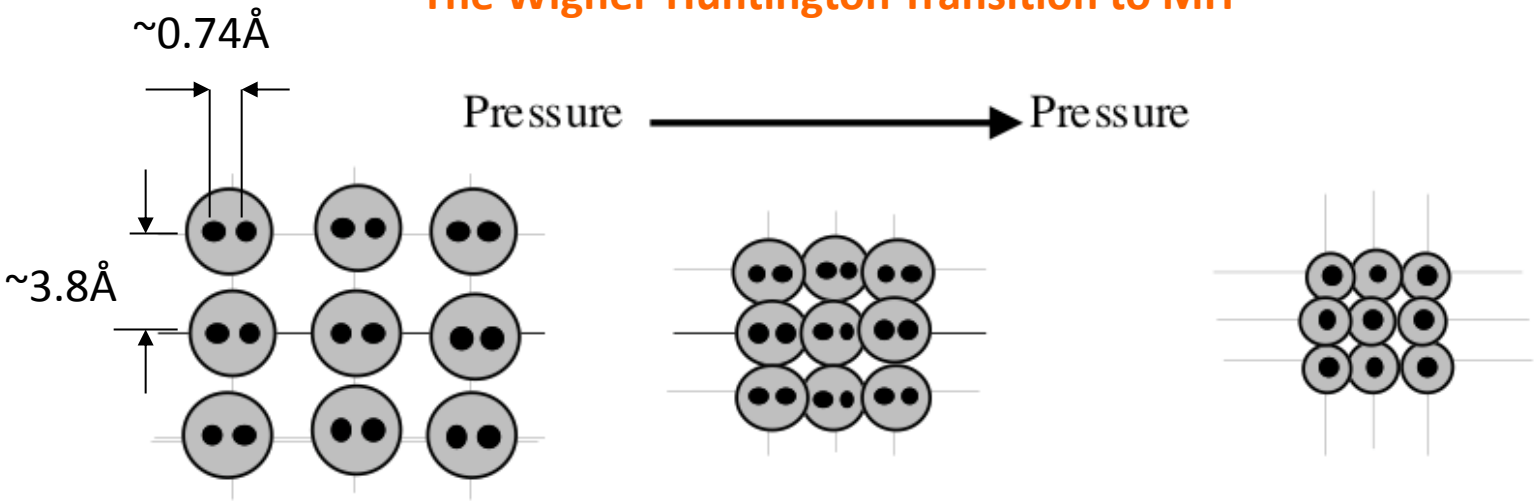
New Junior Faculty in SSAA Research areas

Shanti Deemyad	U of Utah
Ashkan Salamat	UNLV

Our research is focused on making and studying hydrogen in a Diamond Anvil Cell-- DAC

Solid or Liquid Metallic Hydrogen

The Wigner-Huntington Transition to MH



Molecular solid insulater

molecular metal

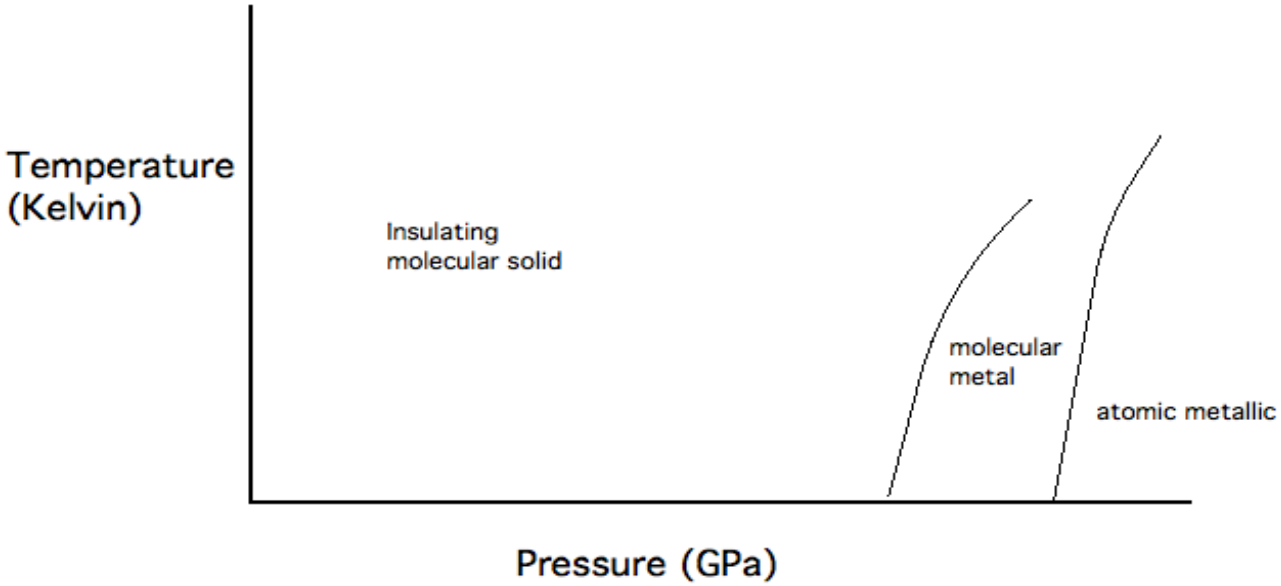
High Tc superconductivity

atomic metal

Liquid?
High Tc Supercond.
Metastable Metal?

Density:	1	11-12?	~13?
Pressure	0	300-400 GPa?	400-600 GPa?

The Simple High Pressure Phase Diagram

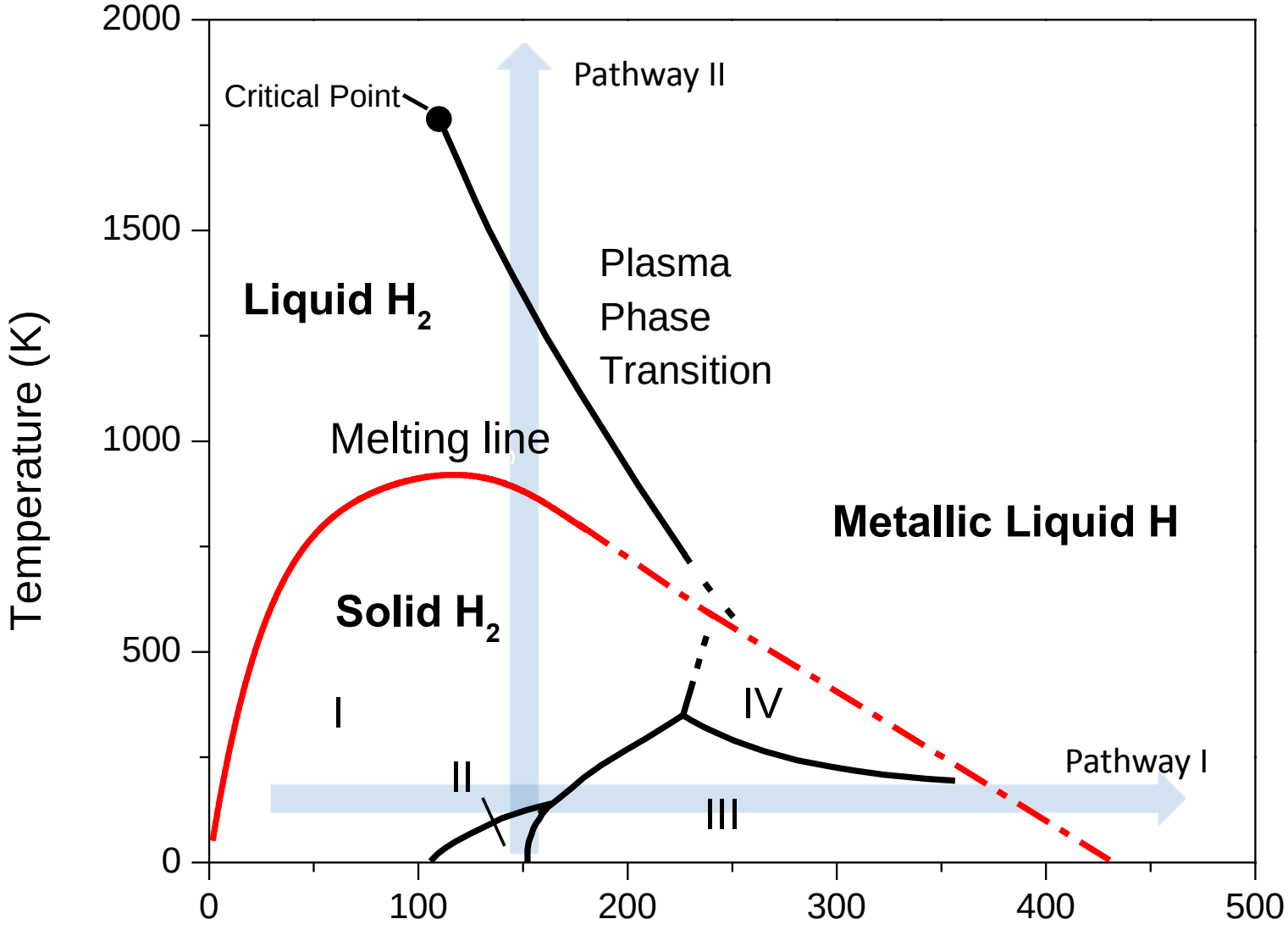


We shall see that it becomes much more complex!

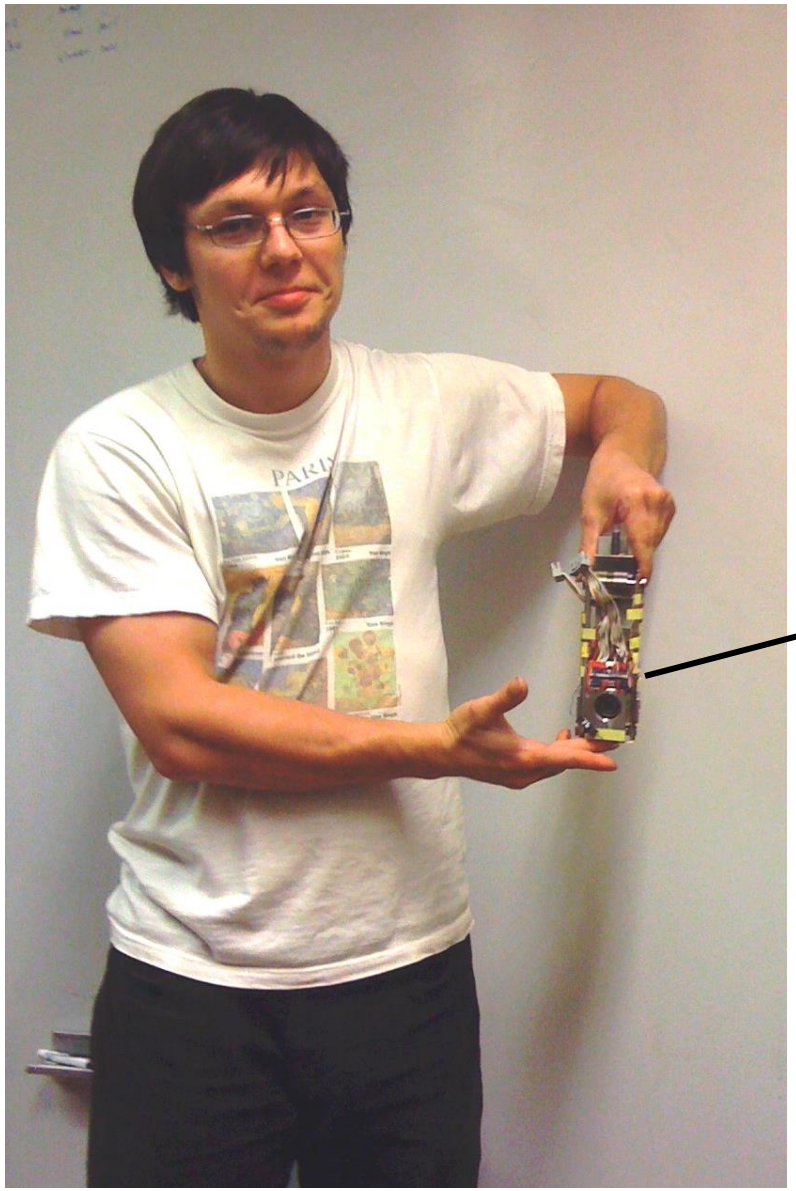
In recent decades a remarkable change has developed in the view of the phase diagram of hydrogen.

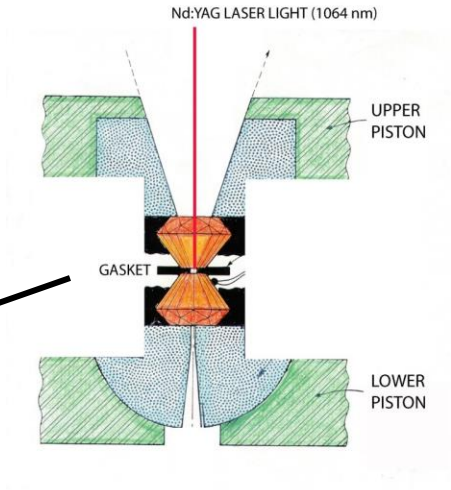
It was realized that there are
<u>two pathways to metallic hydrogen</u>

shown in the next viewgraph.


Temperature (K)
2000
1500
1000
500
0
0
100
200
300
400
500
Critical Point
Pathway II
Liquid H_2
Plasma
Phase
Transition
Melting line
Metallic Liquid H
Solid H_2
I
II
III
IV
Pathway I

We use Diamond Anvil Cells to create multi-megabar static pressures





A liquid-liquid phase transition to Atomic Metallic Hydrogen

For Pathway II, We need:

- **Megabar pressures: 100-200 GPa (100GPa=1 megabar) in a diamond anvil cell**
- **High temperatures: 1000-2000 K**

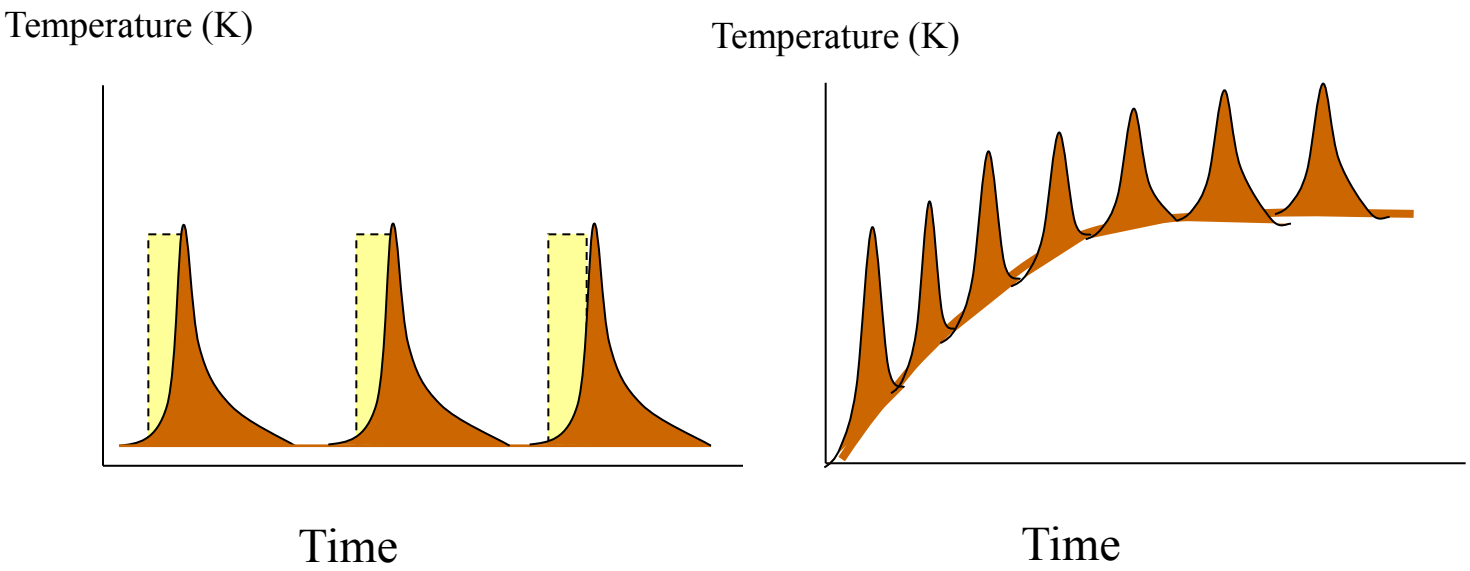
Challenges:

- **Hydrogen diffuses into the gasket of the DAC—loss of sample**
- **Hydrogen diffuses into diamonds at high temperature; diamonds embrittle and fail**

Wigner-Huntington Transition

For Pathway I, we need pressures of 400 to 600 GPa

Laser Pulses and Heating

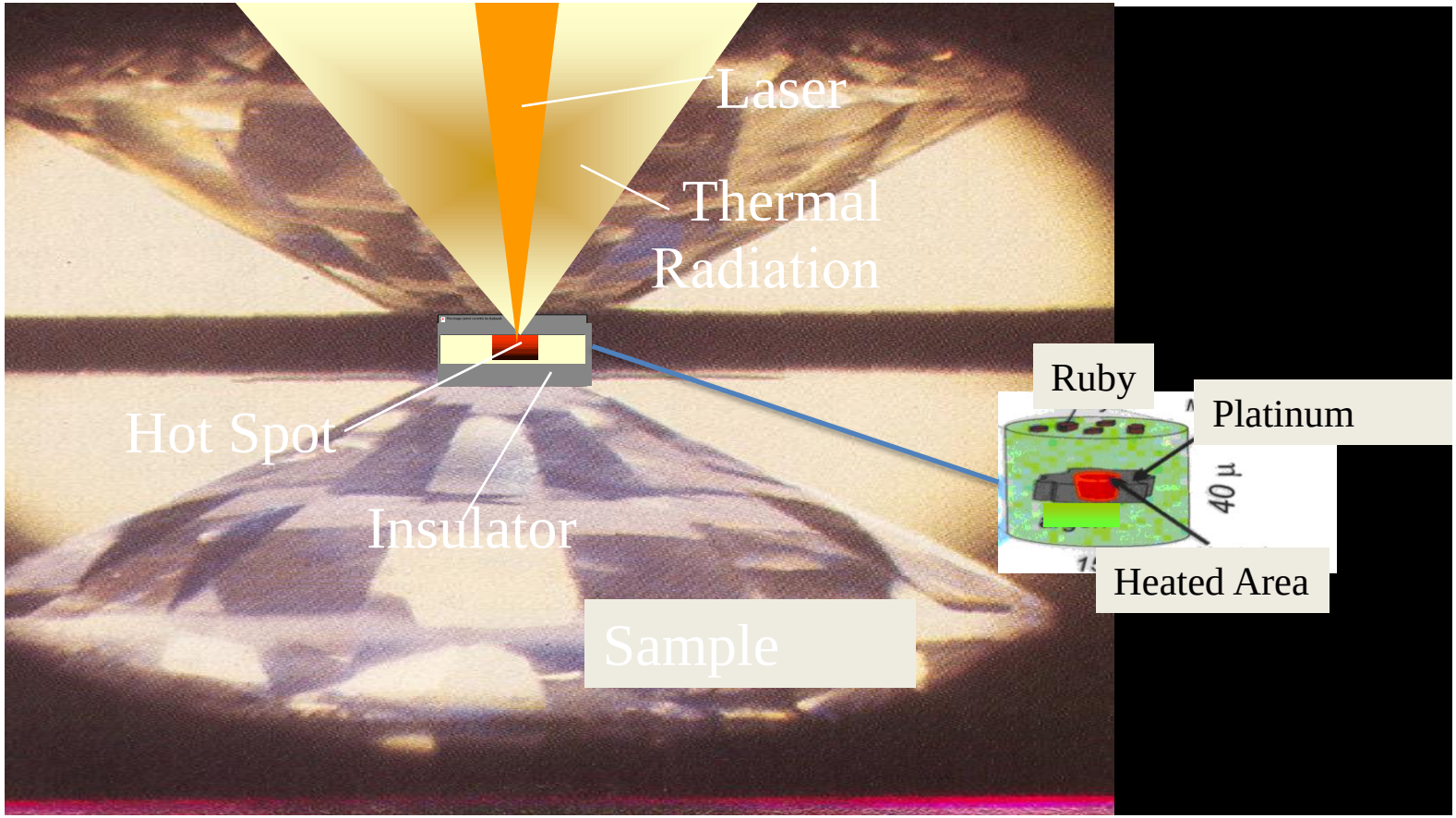


Pulsed laser heating. a) Recovery of initial temperature between pulses. The square waves represent the laser power. b) Increase of steady state temperature for short pulse period.

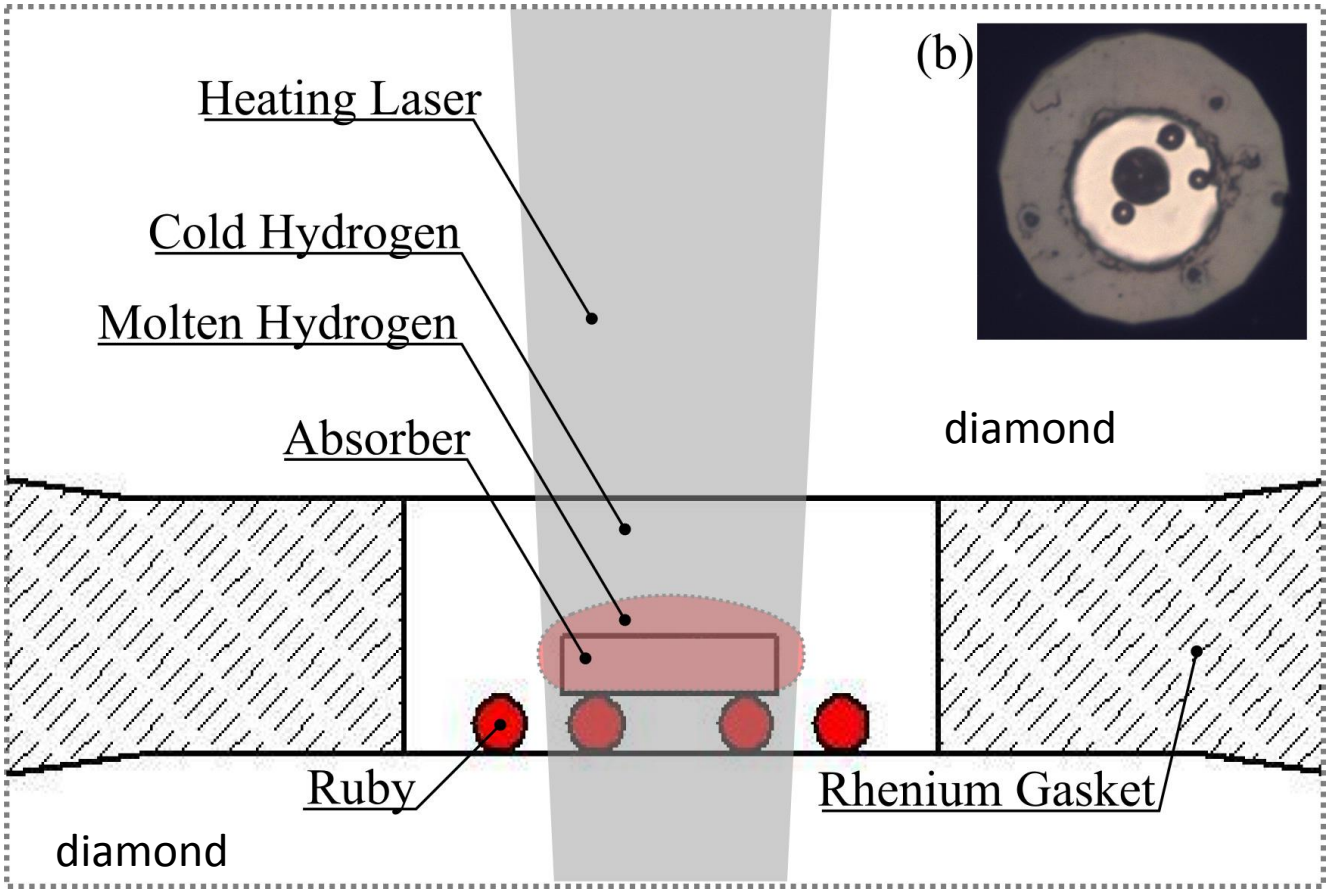
Pulsed laser heating inhibits hydrogen diffusion effects;

Sample thermalizes in picoseconds; pulses ~100-300 nanosecond

Laser heated sample in diamond anvil cell



Sample can be heated to several thousand Kelvin with a laser



50 microns

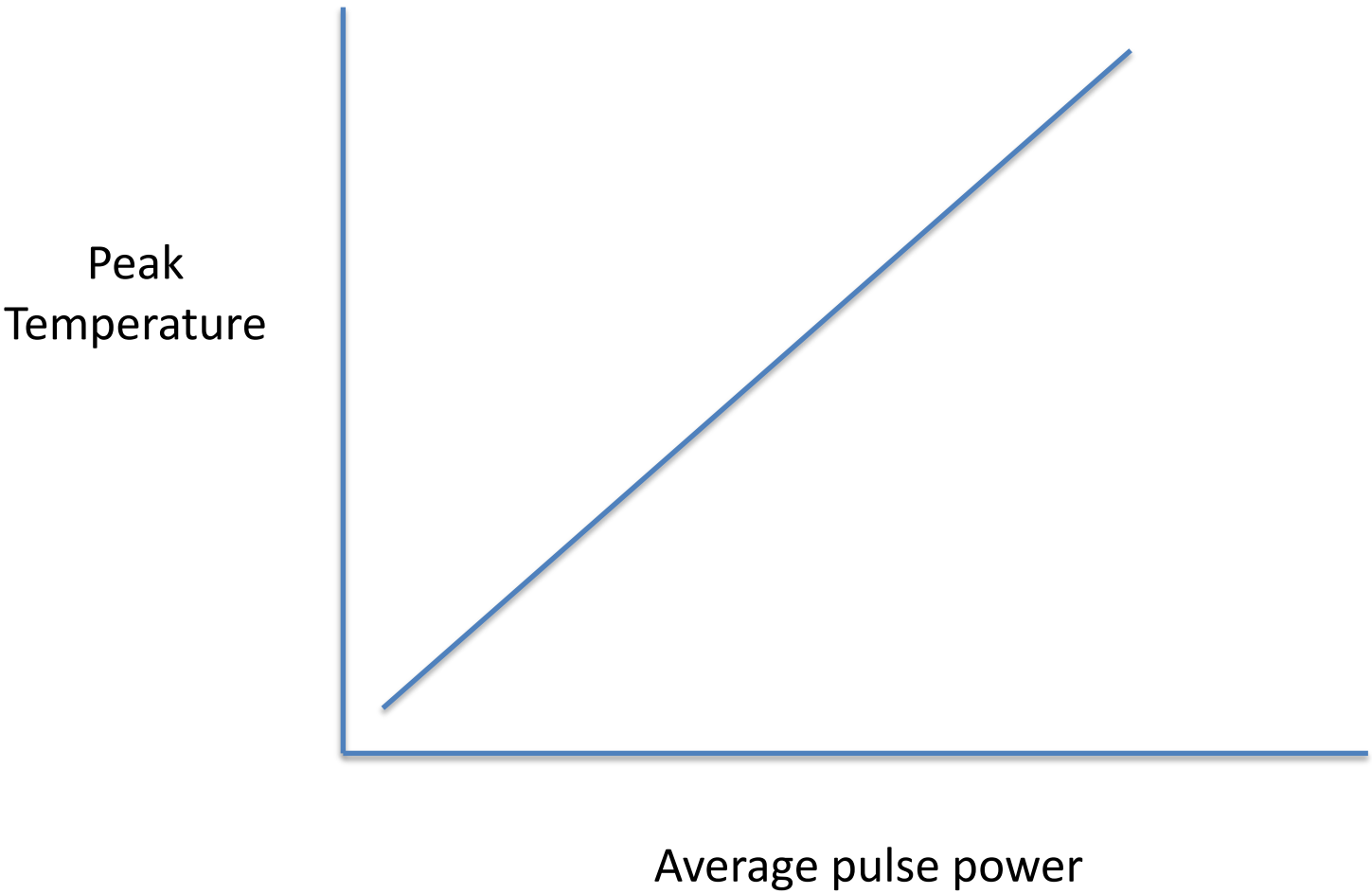
Pulsed laser heating of absorber buried in hi-P hydrogen

Long pulses (~300ns) heat the sample with little energy

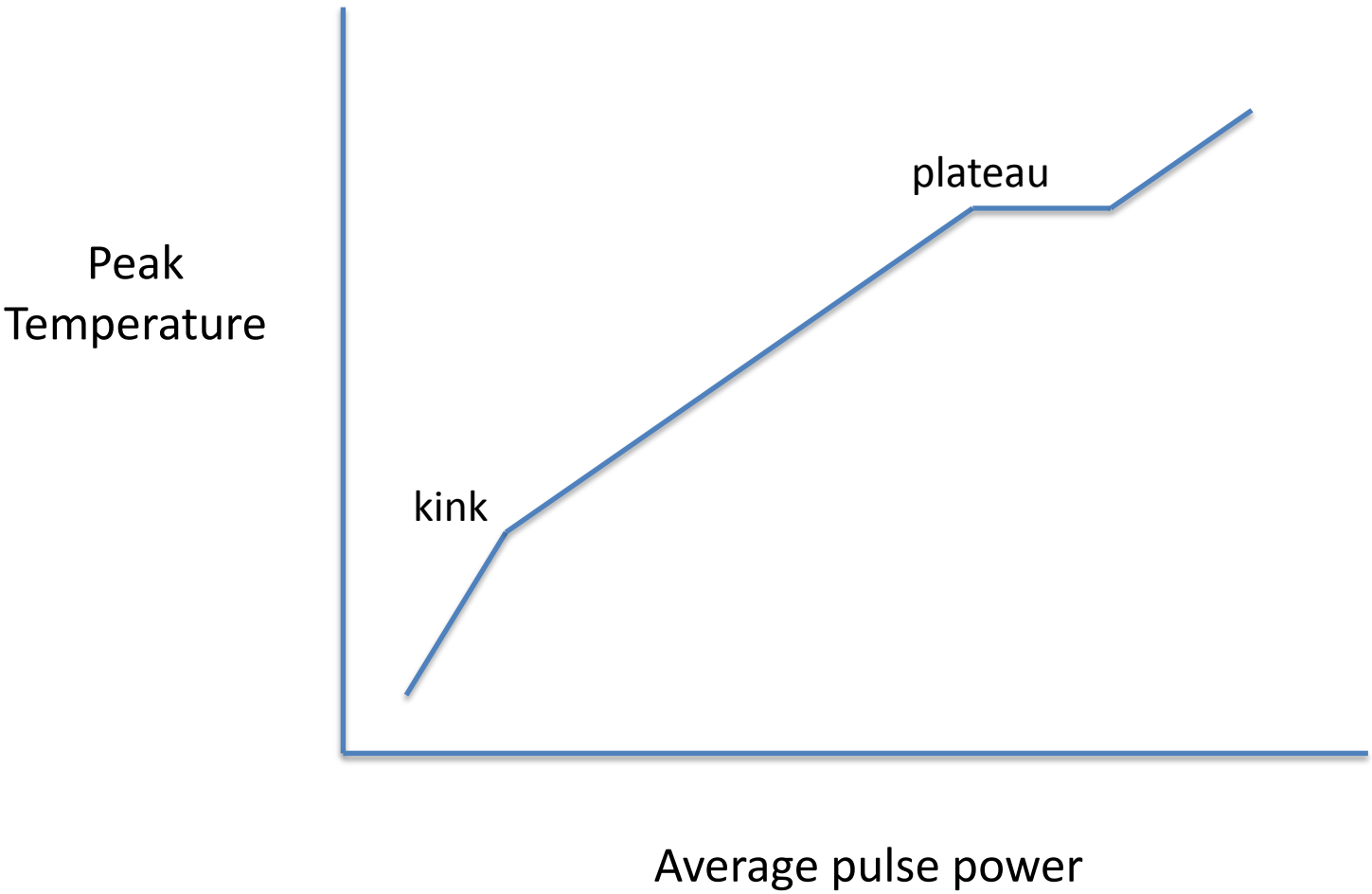
Sufficient time for local thermodynamic equilibrium, but no time to diffuse

We measure heating curves:

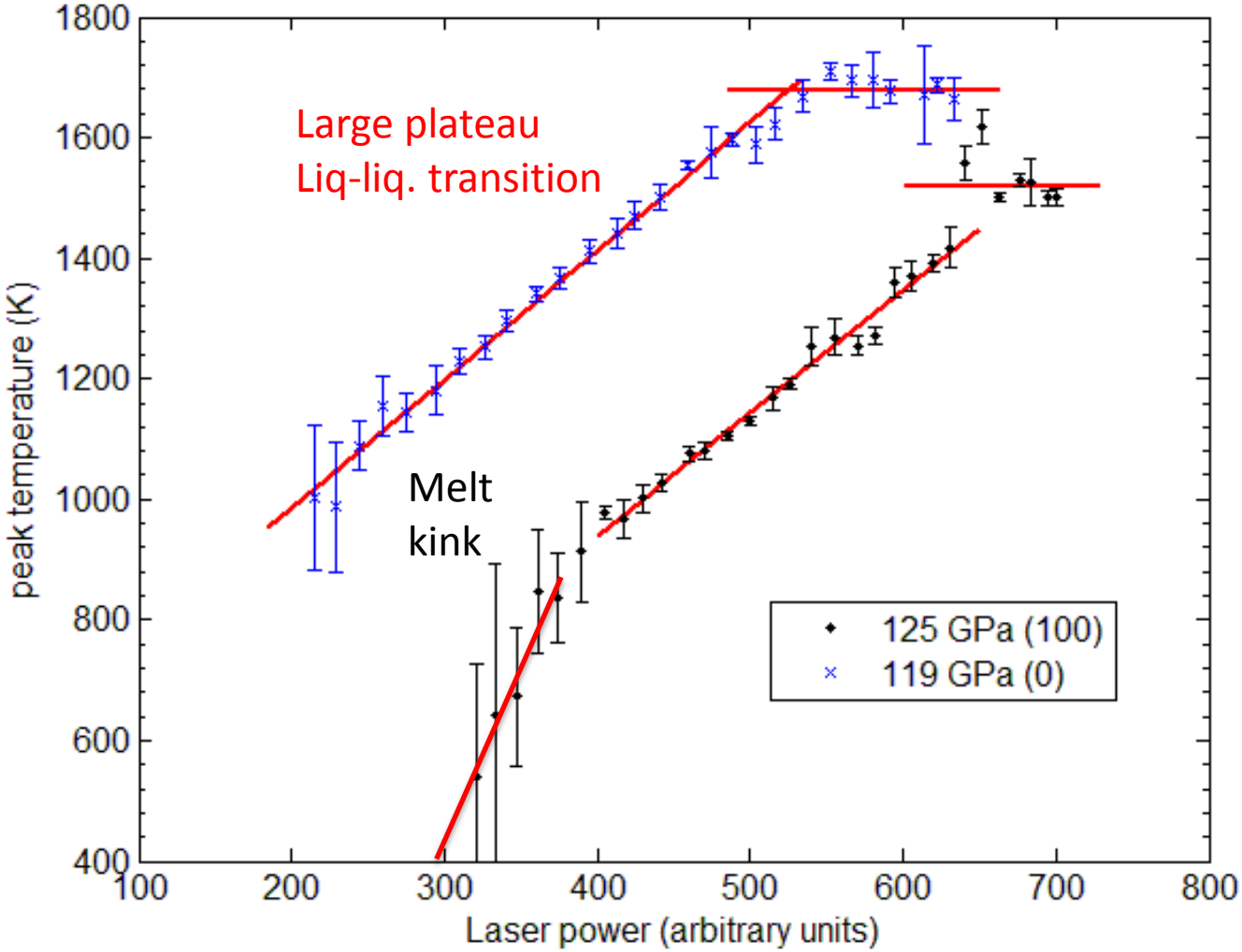
If there are no phase transitions we expect a monotonically Increasing curve



A phase transition leads to a kink or plateau in the curve



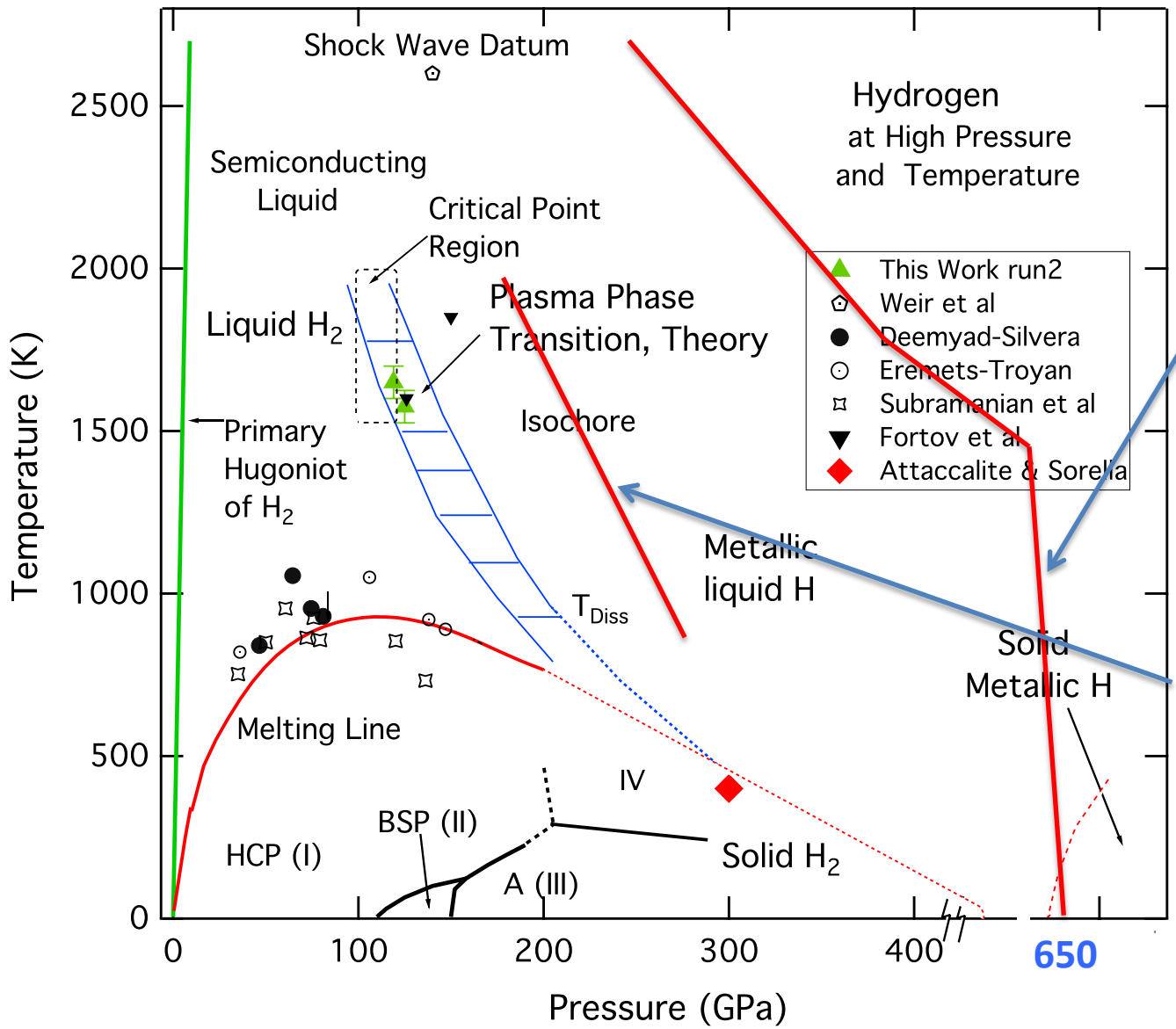
Temperature vs pulsed laser power: This was for a Platinum absorber, 2013



Dzyabura, Zaghoo, IFS, PNAS

Theoretical Developments

Saumon&Chabrier
1990's 15,000K



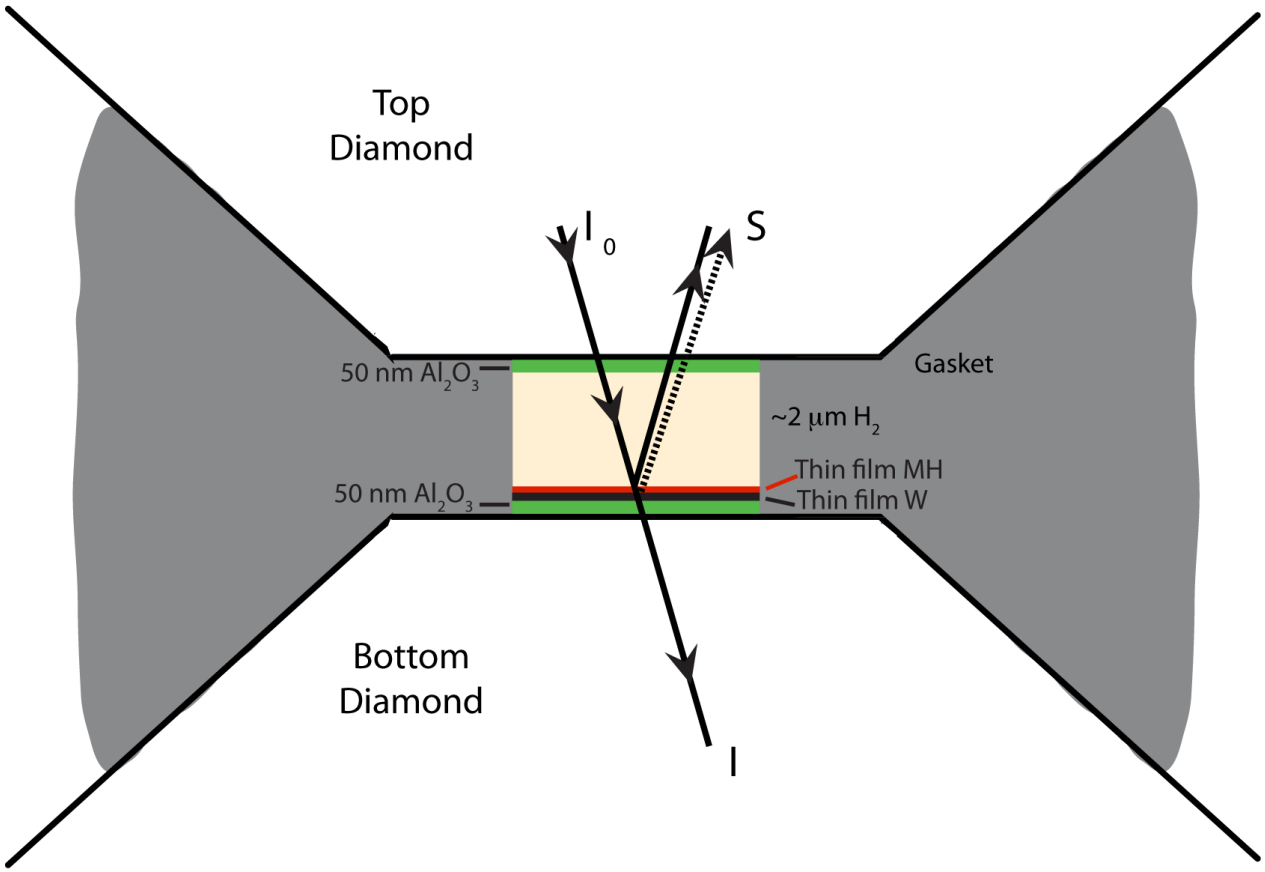
Mazzola, Ynoki, Sorella
Nature Comm. March 2014
Diffusion Monte Carlo

2013: Miguel A. Morales, Jeffrey M. McMahon Carlo Pierleoni, and David M. Ceperley:
<u>Nuclear Quantum Effects</u> shift the PPT line by ~ 100 GPa-Firs t principles calculations

Yang et al 2015 agree with blue crosshatch

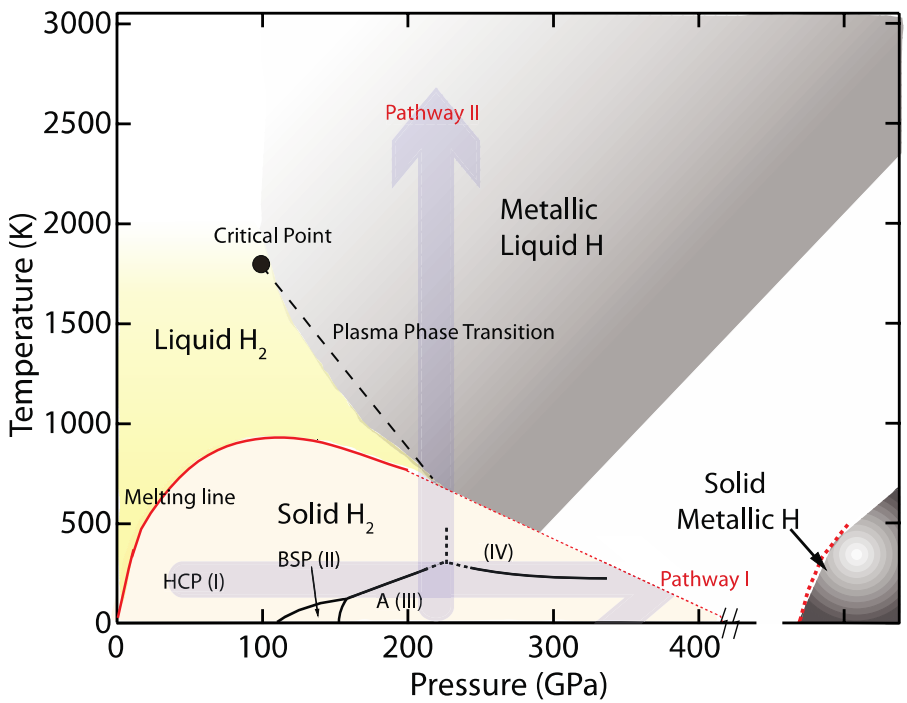
To show that hydrogen is metallic we want to measure transmission and reflection on the plateaus.

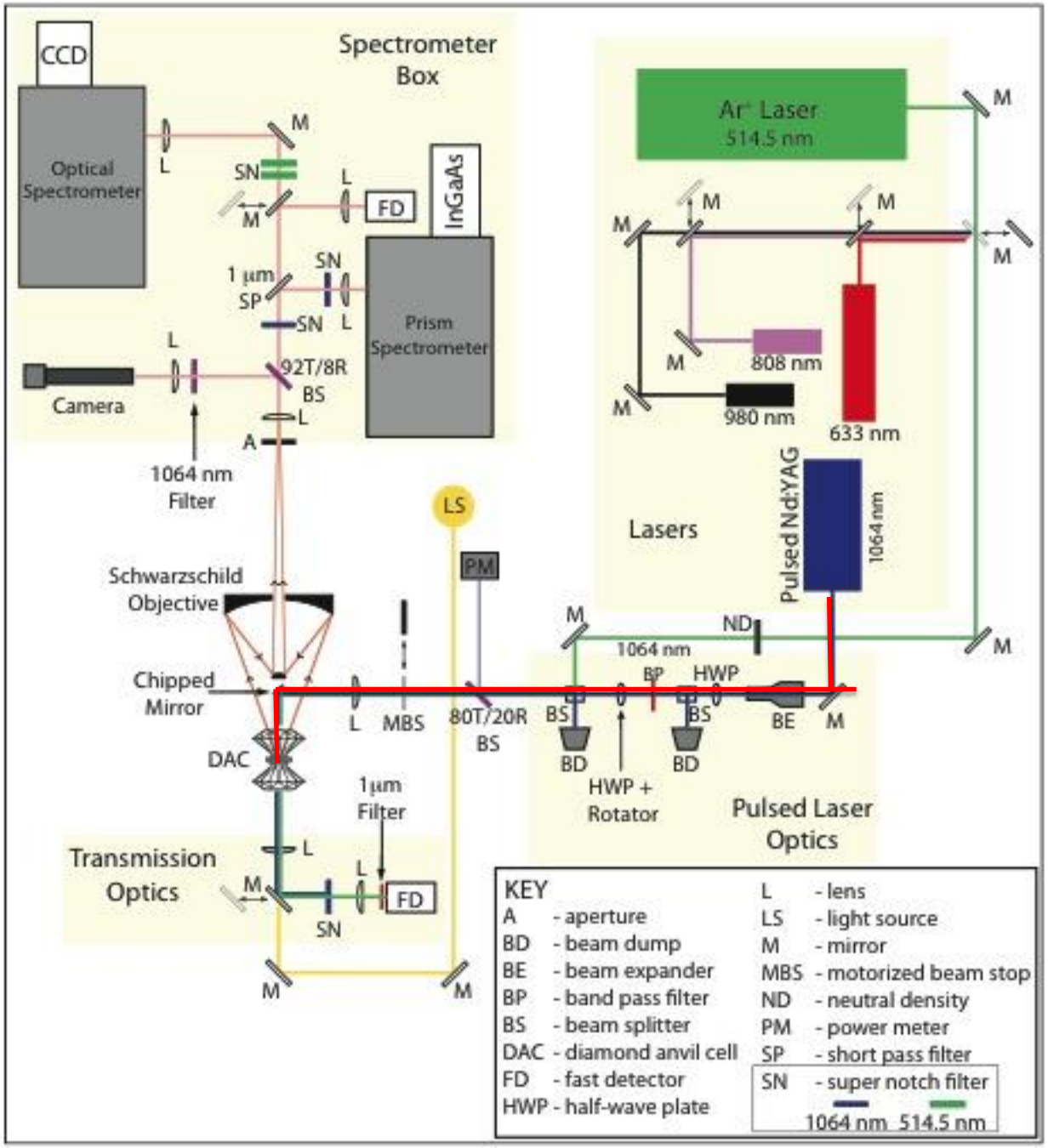
Heart of the DAC with semi-transparent laser absorber: enables measurement of transmission and reflection while pulsed laser heating

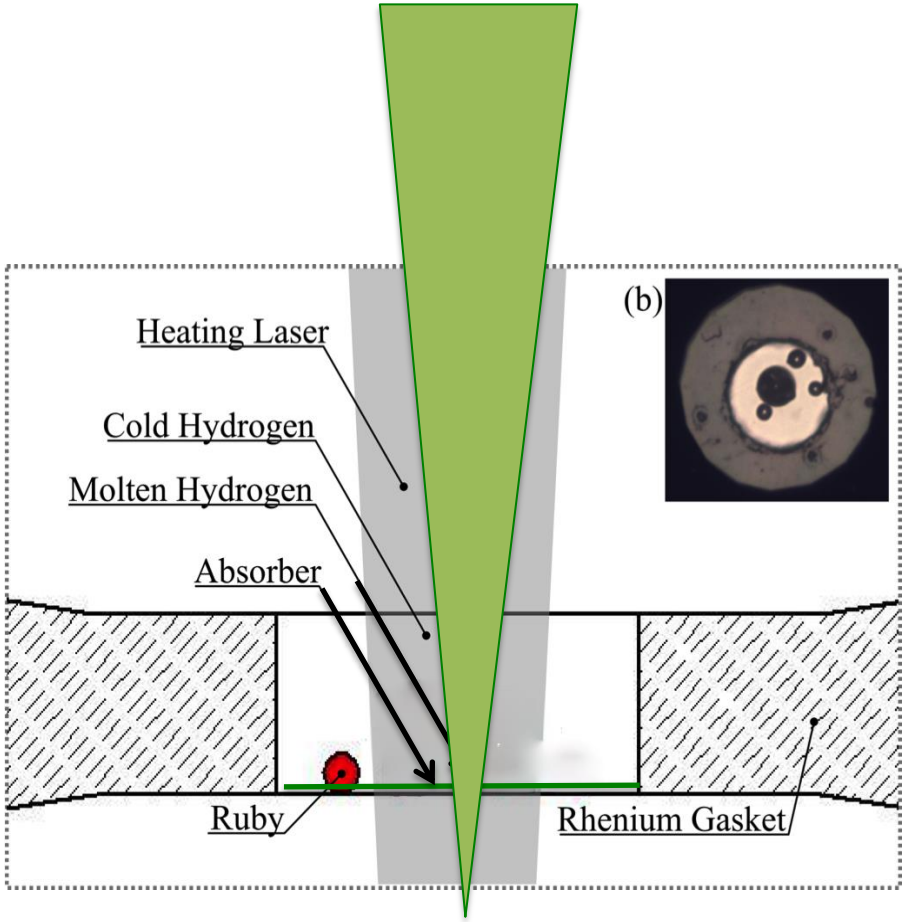


Now, Pathway II, we need high pressure and high temperature

Problems: Attempts by others found that with continuous heating to high temperature, hydrogen diffused into the gasket and the <u>sample was lost</u>; if a diffusion barrier was used on the gasket to inhibit diffusion, then hydrogen diffused into the diamond anvils and <u>the diamonds failed</u>




CCD
Spectrometer Box
Optical Spectrometer
L
M
SN
M
L
FD
InGaAs
1 µm
SP
SN
SN
L
Prism Spectrometer
L
92T/8R
BS
Camera
L
A
1064 nm Filter
Ar⁺ Laser
514.5 nm
M
M
M
M
M
M
808 nm
980 nm
633 nm
Lasers
Pulsed Nd:YAG
1064 nm
LS
PM
Schwarzschild Objective
Chipped Mirror
DAC
L
MBS
80T/20R
BS
M
ND
M
1064 nm
BP
HWP
BS
BS
BE
M
BD
BD
HWP + Rotator
Pulsed Laser Optics
1µm Filter
Transmission Optics
L
M
L
FD
SN
M
M
KEY
A - aperture
BD - beam dump
BE - beam expander
BP - band pass filter
BS - beam splitter
DAC - diamond anvil cell
FD - fast detector
HWP - half-wave plate
L - lens
LS - light source
M - mirror
MBS - motorized beam stop
ND - neutral density
PM - power meter
SP - short pass filter
SN - super notch filter
1064 nm 514.5 nm



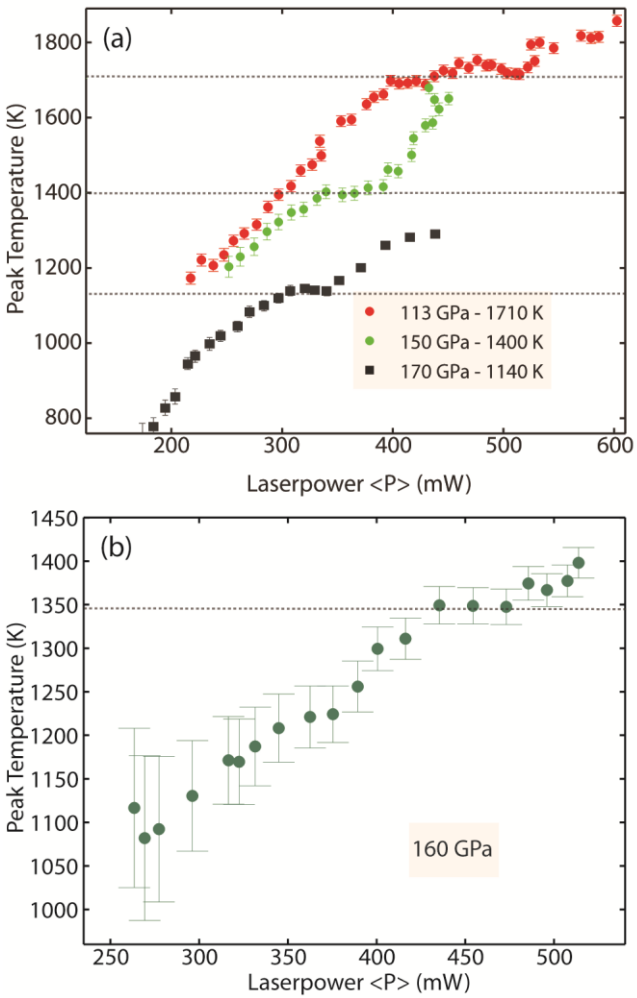
Semi transparent tungsten film on diamond as absorber.

Plateaus on Tungsten absorber

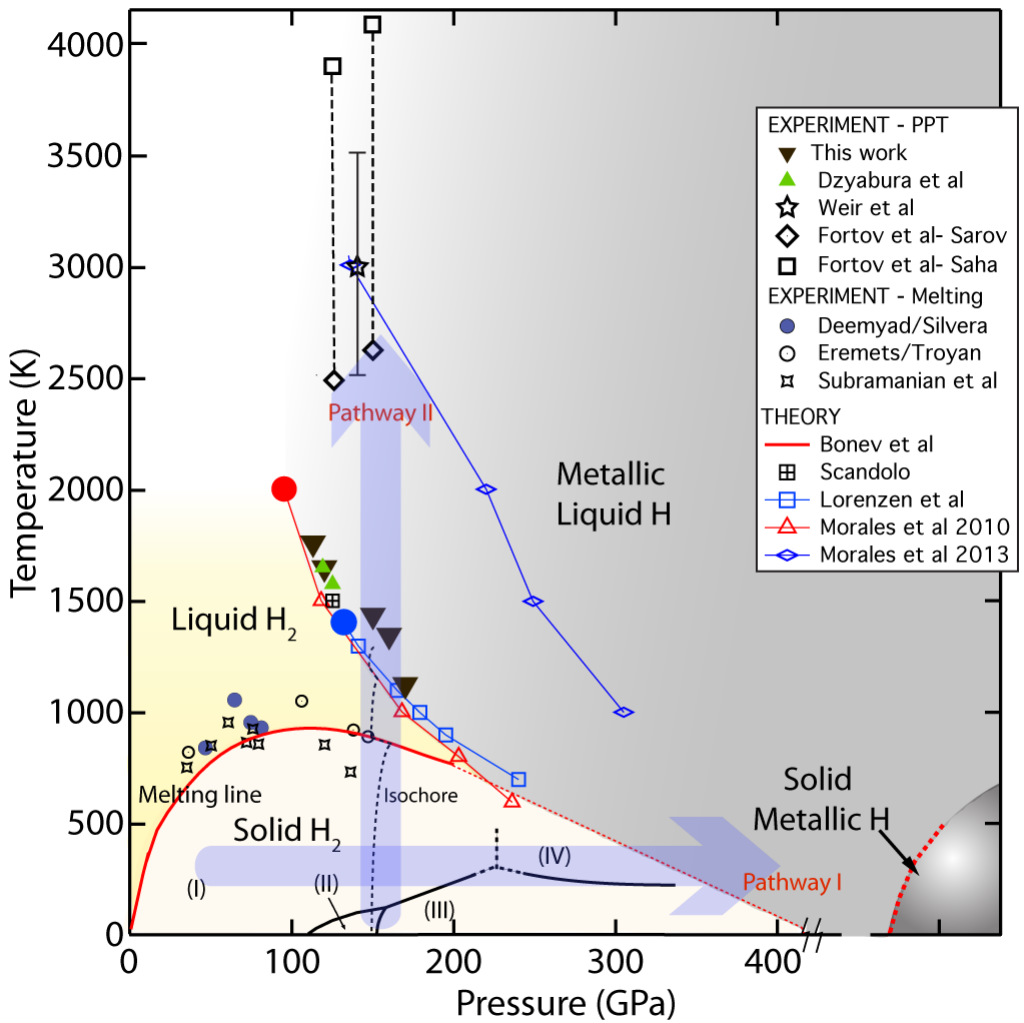
Origin of plateaus: two possibilities

- Reflectivity increases, so laser power must increase to maintain heating and temperature

- Latent heat of transformation: if this is the case, then it is evidence of a first-order phase transition

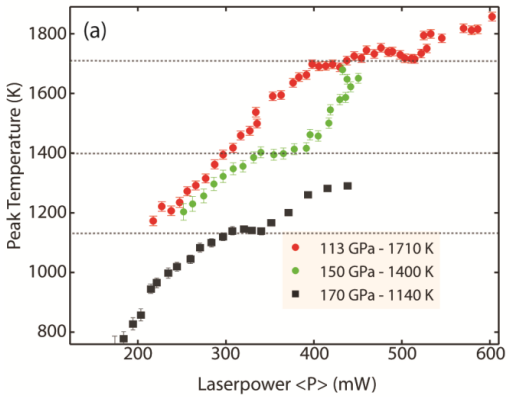


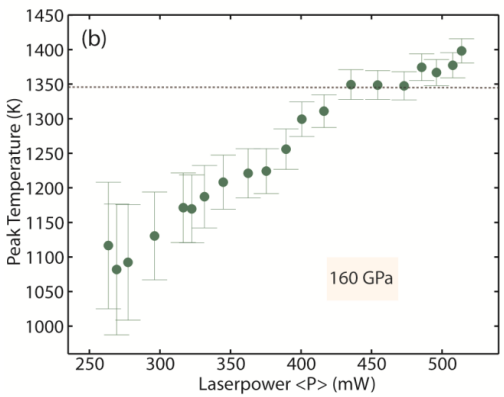
Pressure-Temperature points of Plateaus

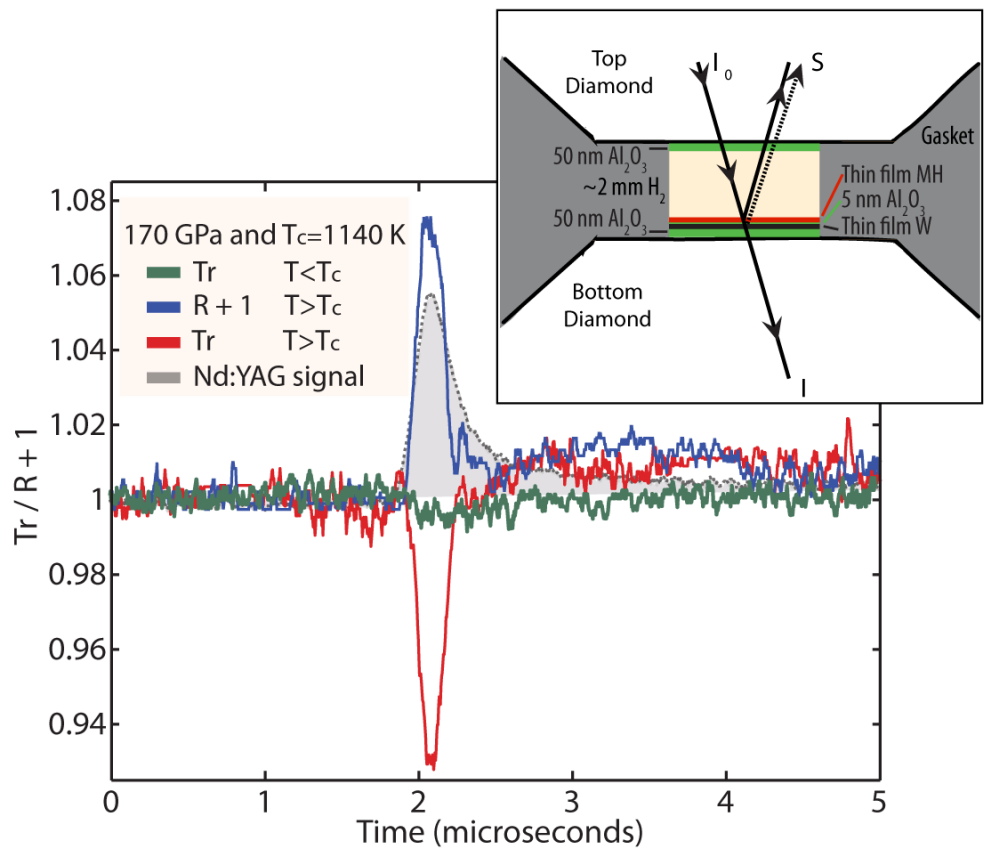


Zaghoo, Salamat, Silvera; accepted for publication

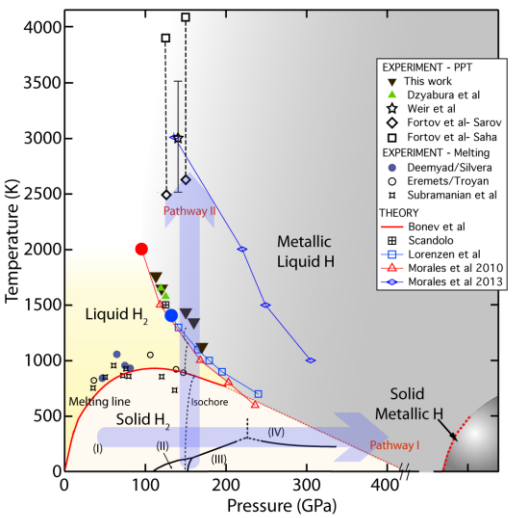
Transmission and Reflection on or at higher temperatures than the plateau

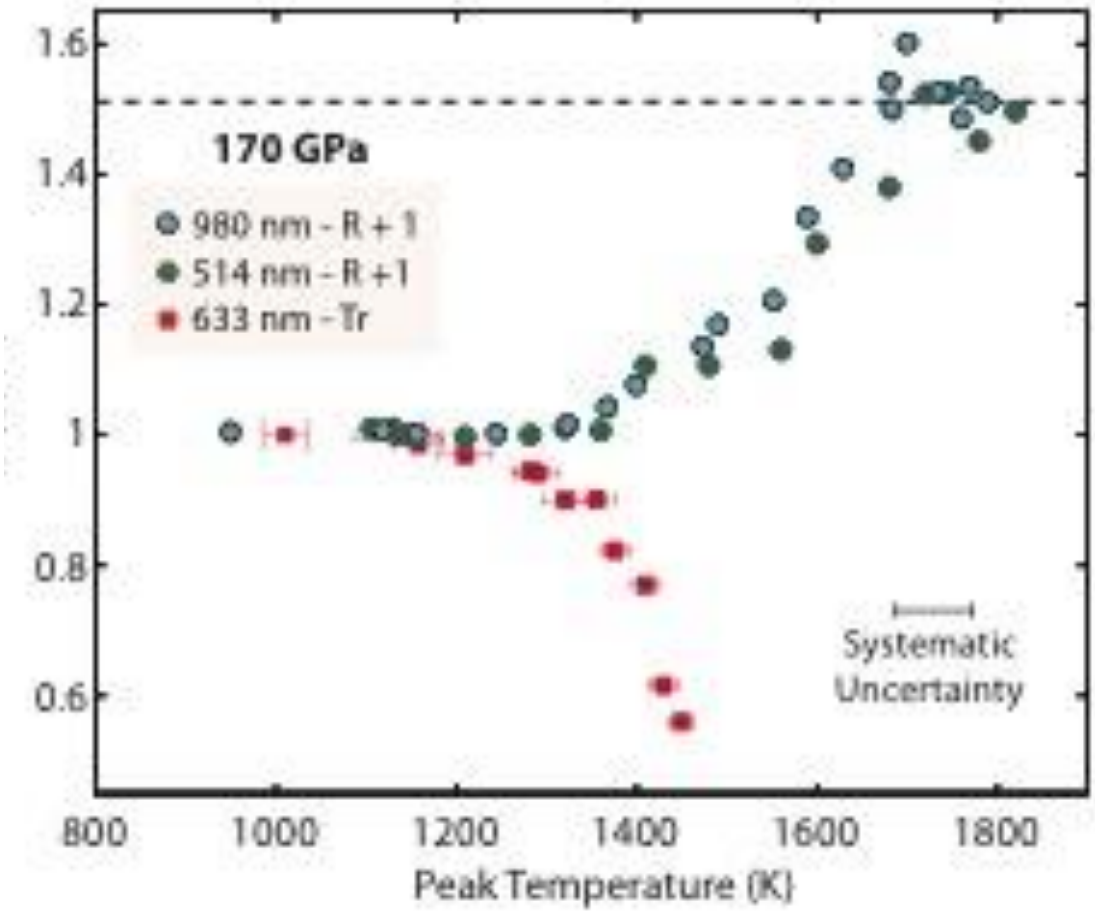






Heating to thick film and bulk reflectance limit





According to theory the PPT is a first-order phase transition.

The plateaus can arise either from latent heat of transformation (characterizes a first-order transition) or increased reflectivity at the plateau.

Reflectance is negligible so the plateau is due to latent heat and the transition is first order; the latent heat is most likely due to dissociation of molecules

At 170 GPa and 1240K we estimate the Fermi energy is ~13 EV, assuming complete dissociation of the molecules

So the MH is a metal with a well-defined Fermi surface; it is not a hot plasma with Maxwell-Boltzmann statistics.

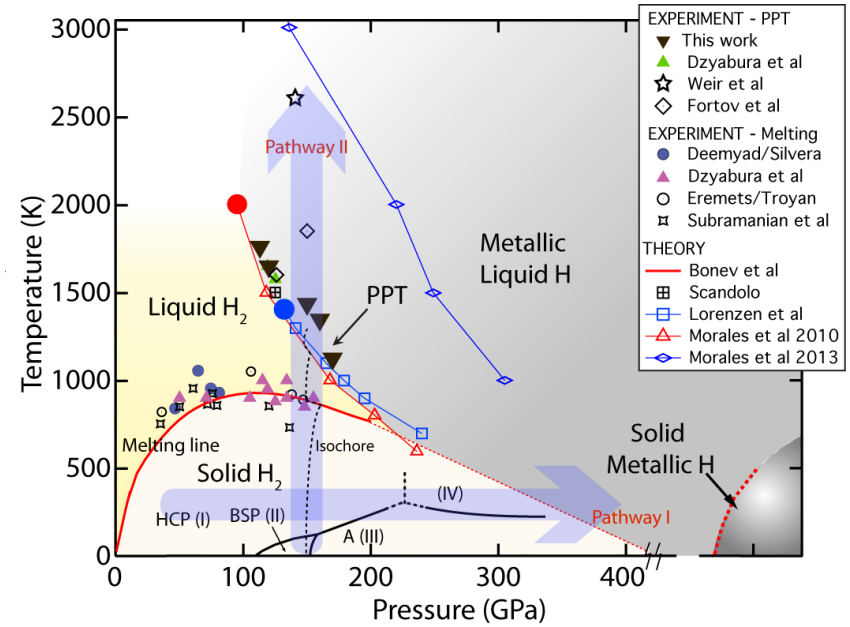
Estimate of DC electrical conductivity from Drude/dielectric function is ~13,000 (ohm-cm)^-1—much better than Mott minimum conductivity for a metal, assuming full dissociation of the hydrogen

Significance of weak reflectance on the plateau:

Plateaus due to latent heat

We have observed a first-order phase transition in agreement with theory:

The Plasma Phase Transition to metallic hydrogen

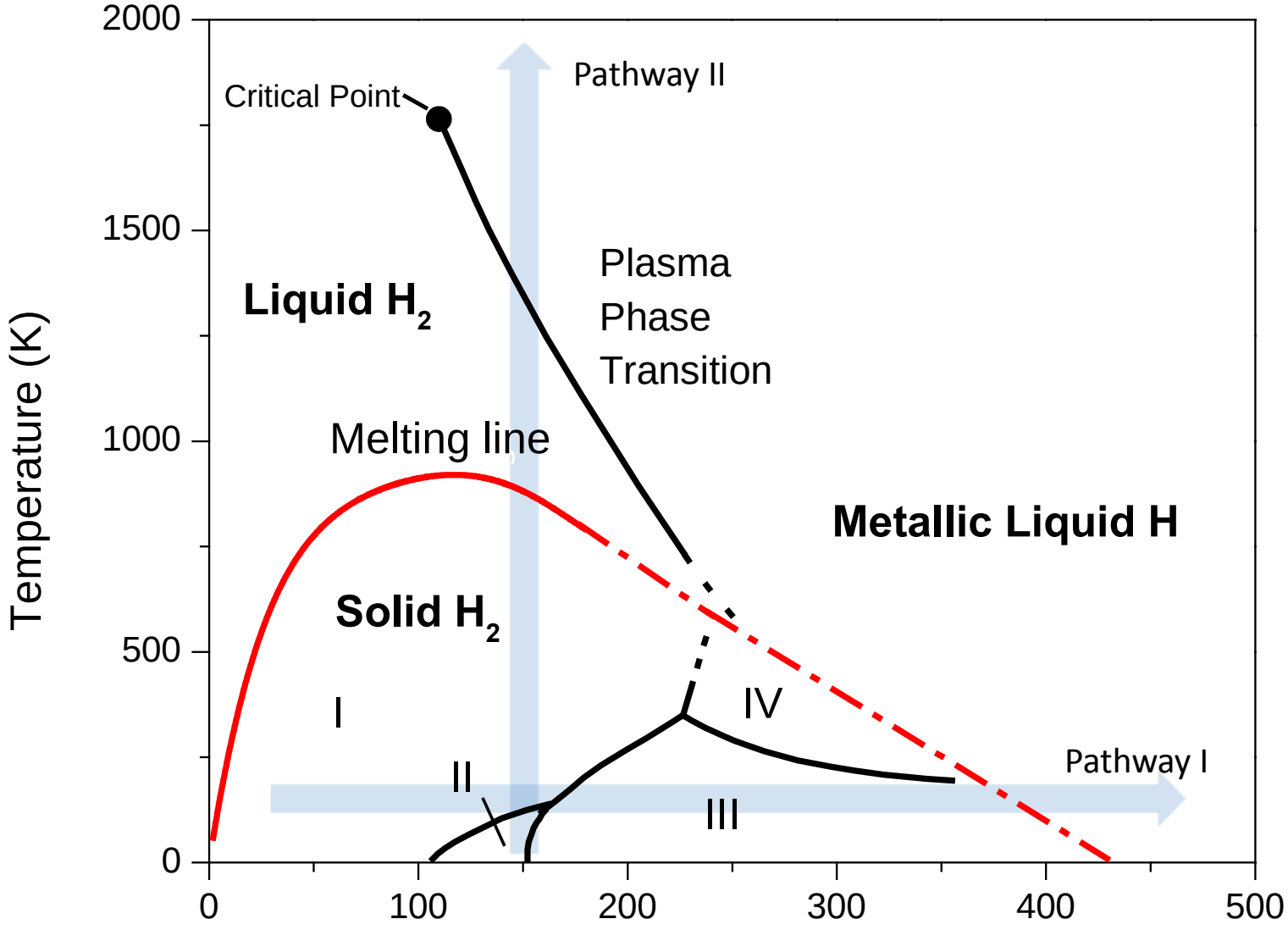


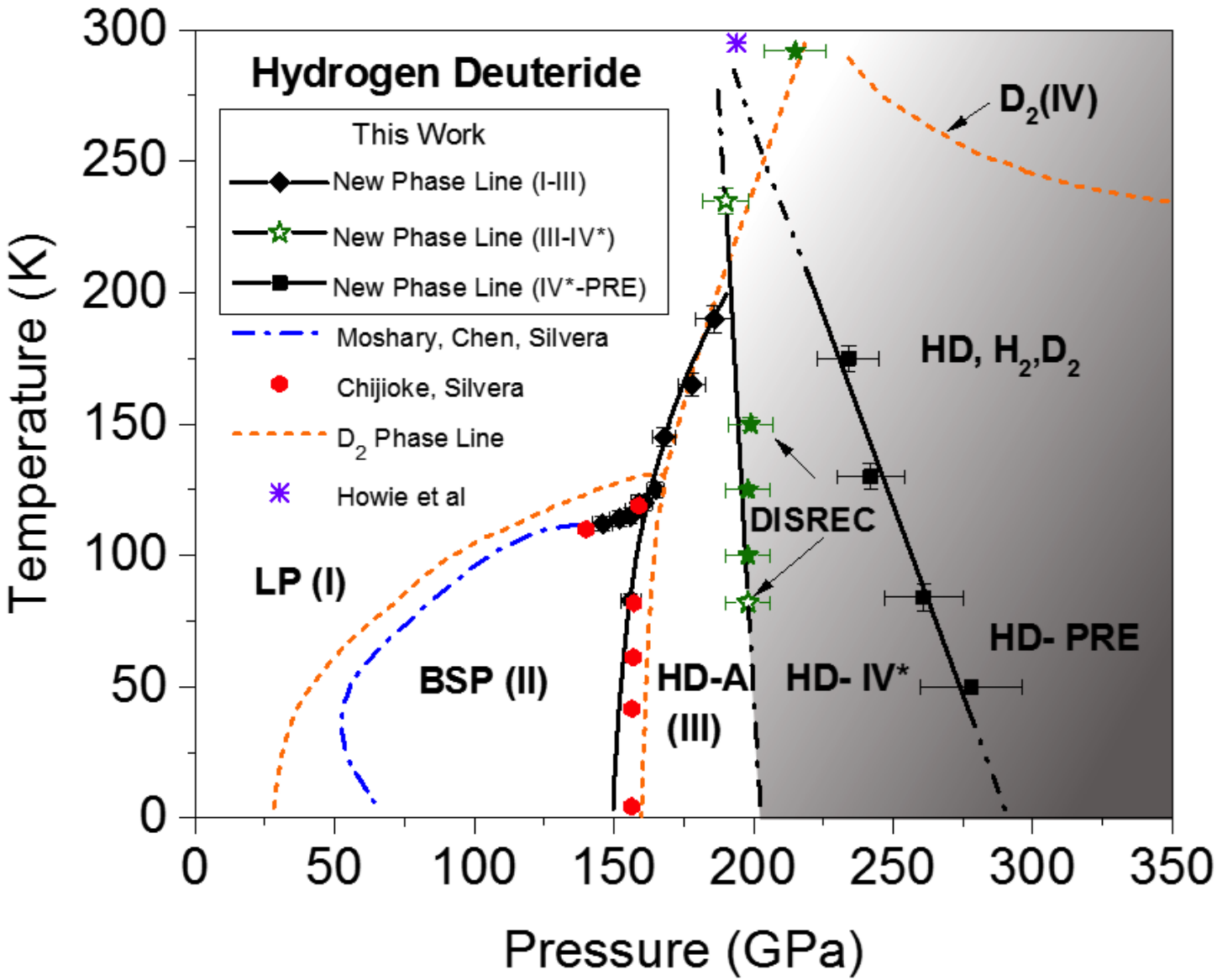
- **Phase line provides guidance to theorists**
- **Phase line may "soon" intersect melting line**

- **Current temperatures too high to observe the predicted superconductivity or metastability**

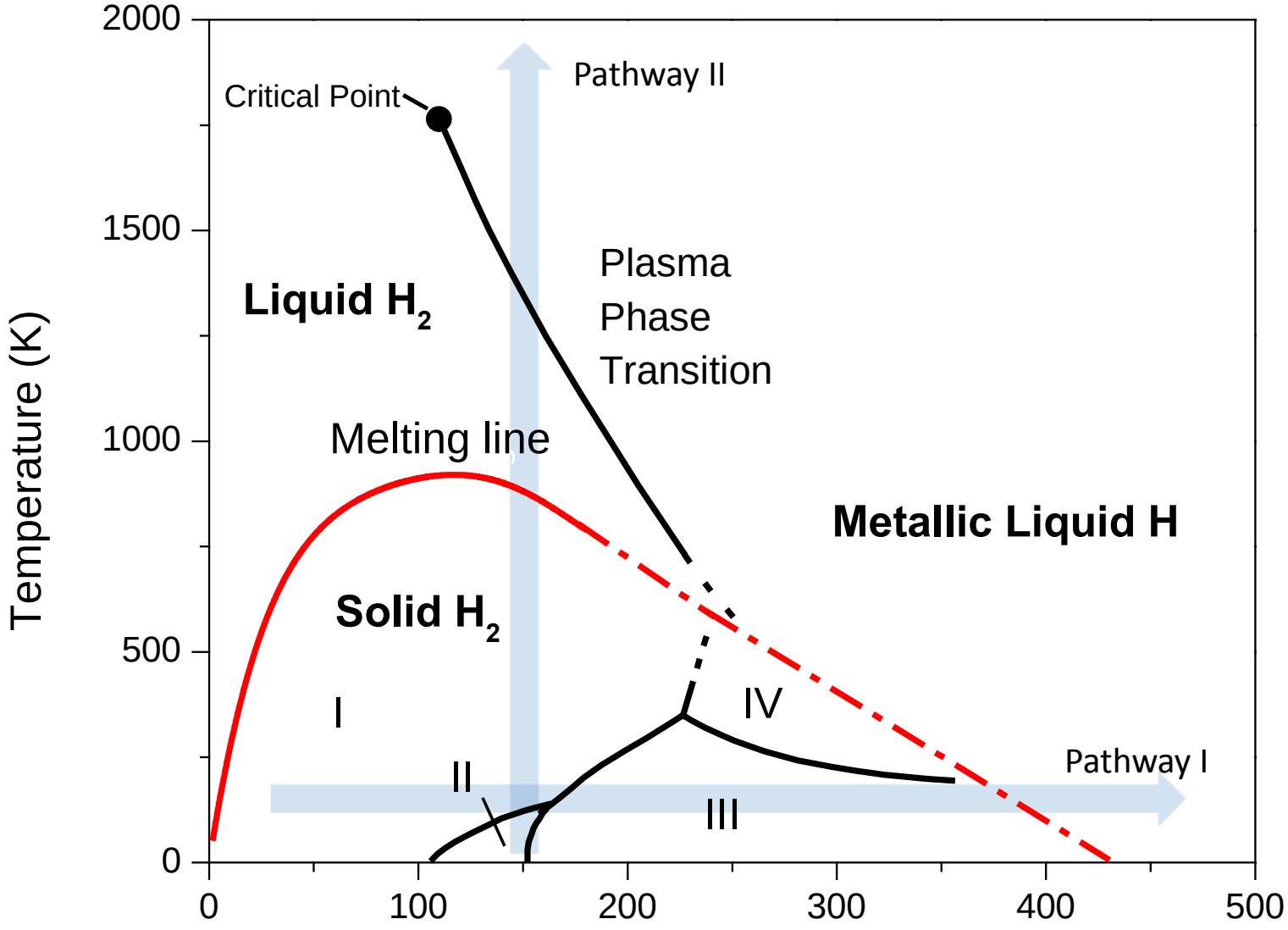
- **The first observation of a first-order phase transition to metallic hydrogen**

What about Pathway I?

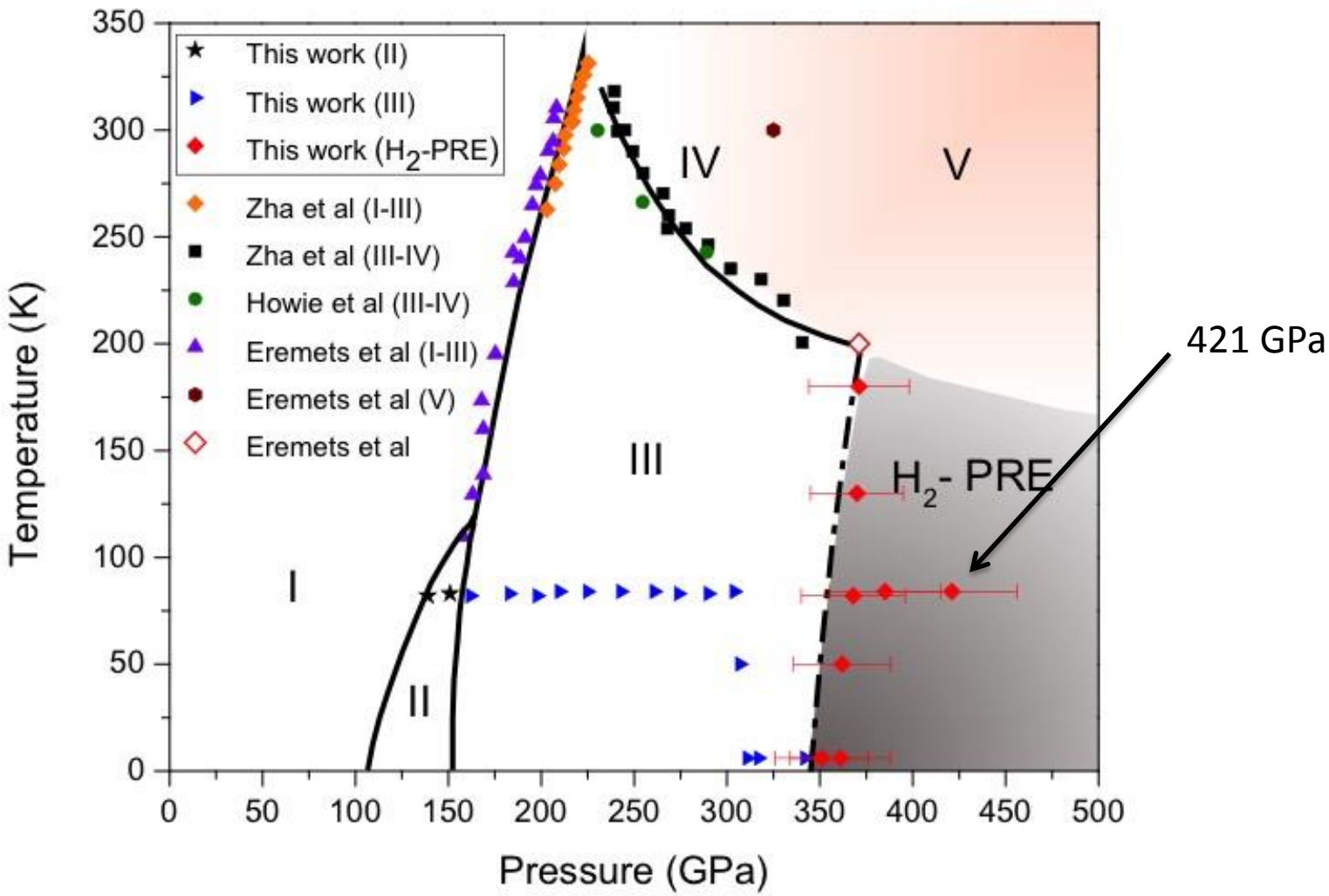

Temperature (K)
2000
1500
1000
500
0
0
100
200
300
400
500
Critical Point
Pathway II
Liquid H_2
Plasma
Phase
Transition
Melting line
Metallic Liquid H
Solid H_2
I
II
III
IV
Pathway I



Dias, Noked, Silvera; PRL accepted for publication

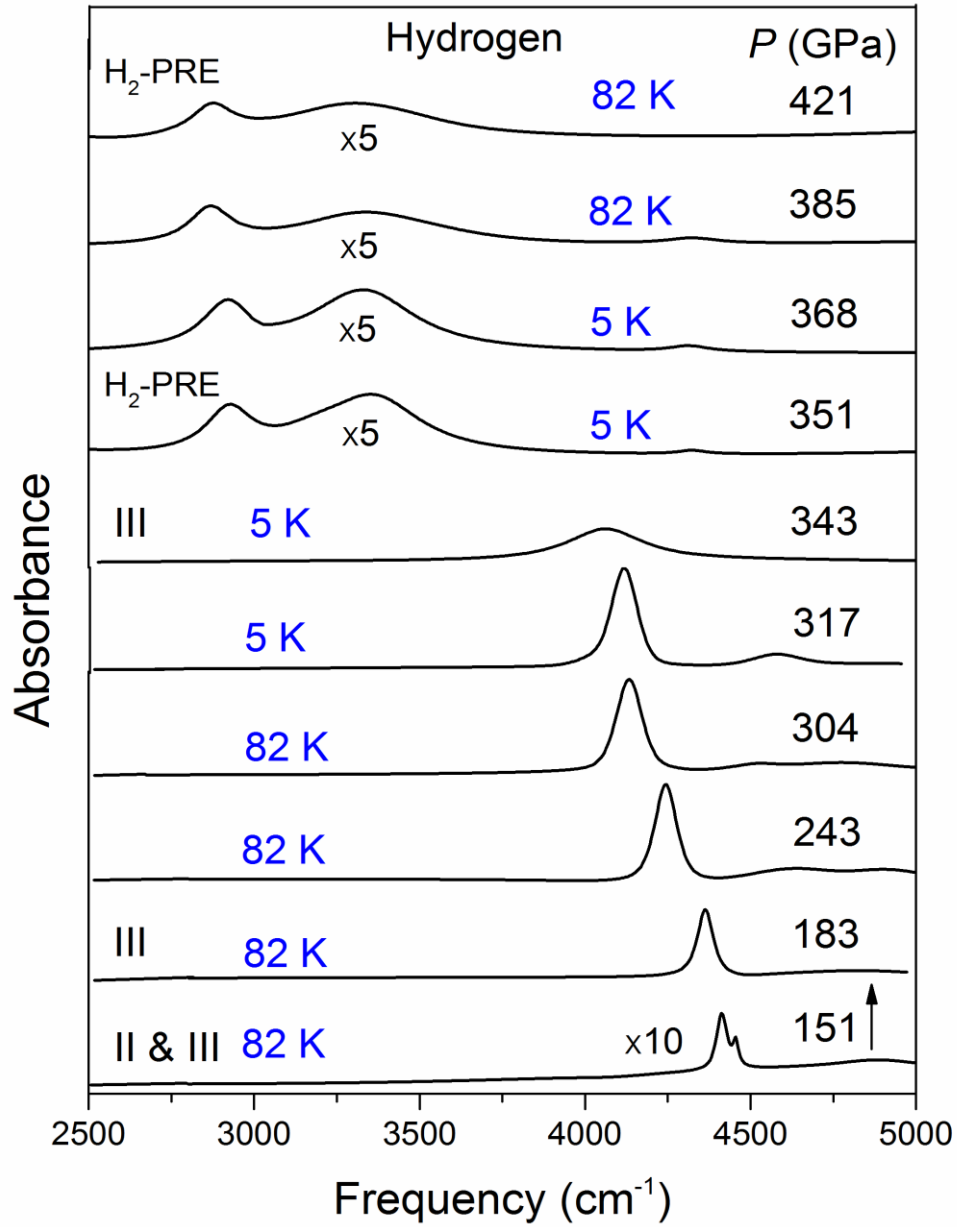

Temperature (K)
2000
1500
1000
500
0
0
100
200
300
400
500
Critical Point
Pathway II
Liquid H_2
Plasma
Phase
Transition
Melting line
Metallic Liquid H
Solid H_2
I
II
III
IV
Pathway I

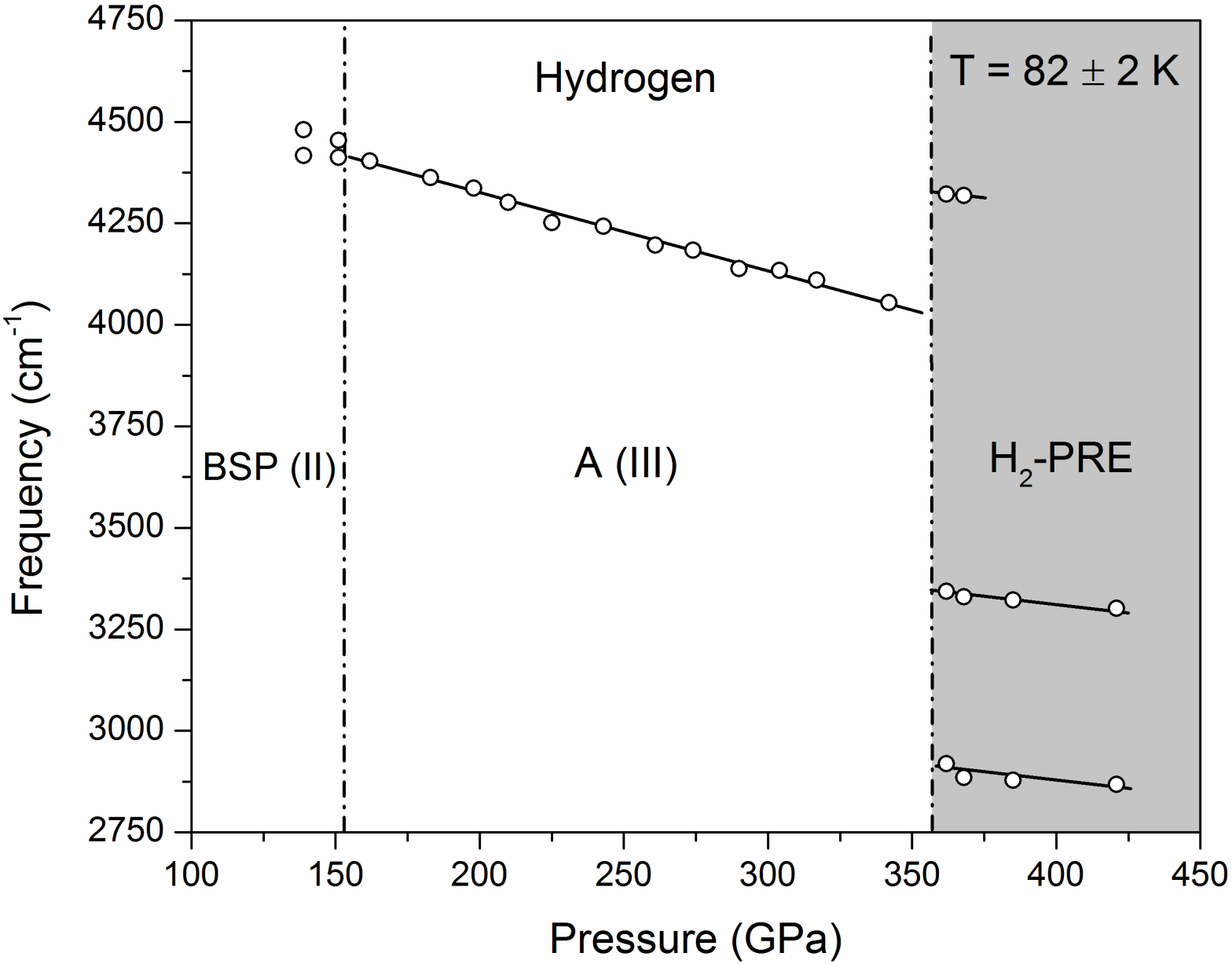
Hydrogen: the Stubborn Insulator

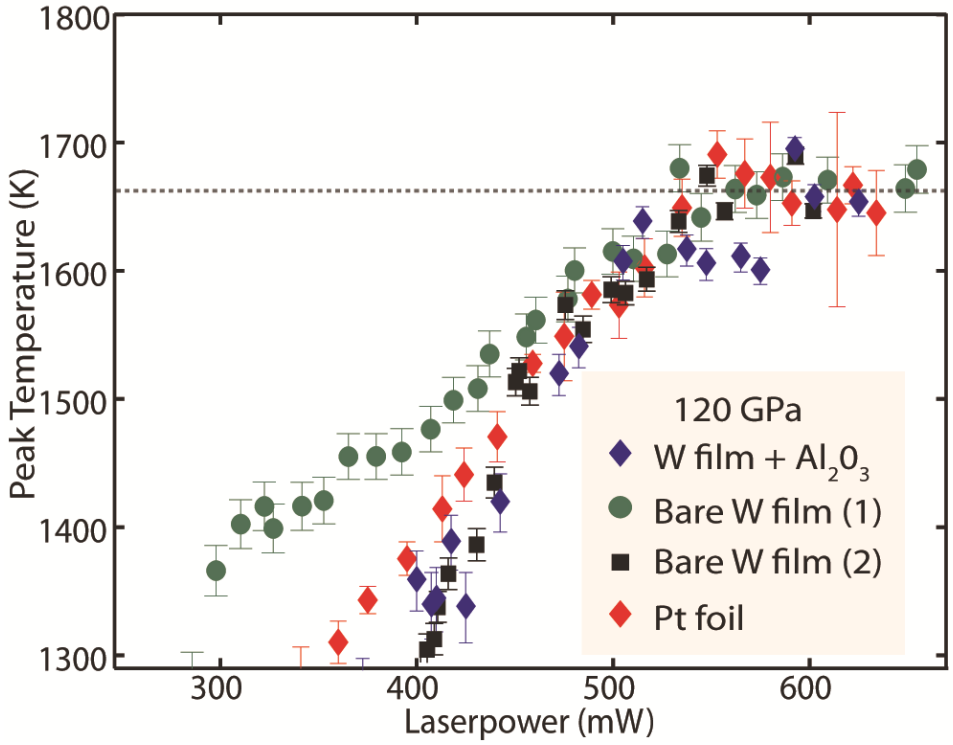


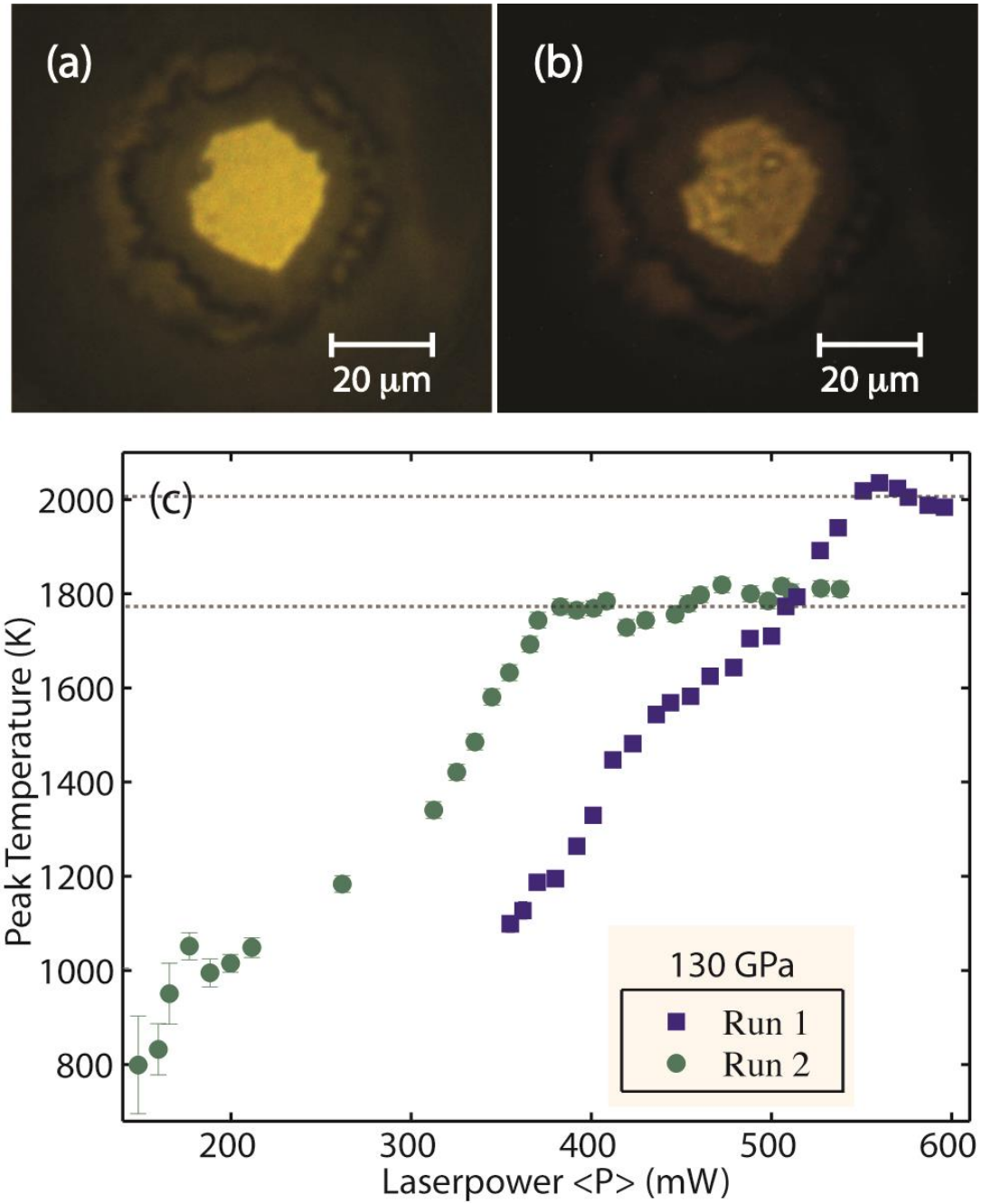
Dias, Noked, Silvera: ready for submission

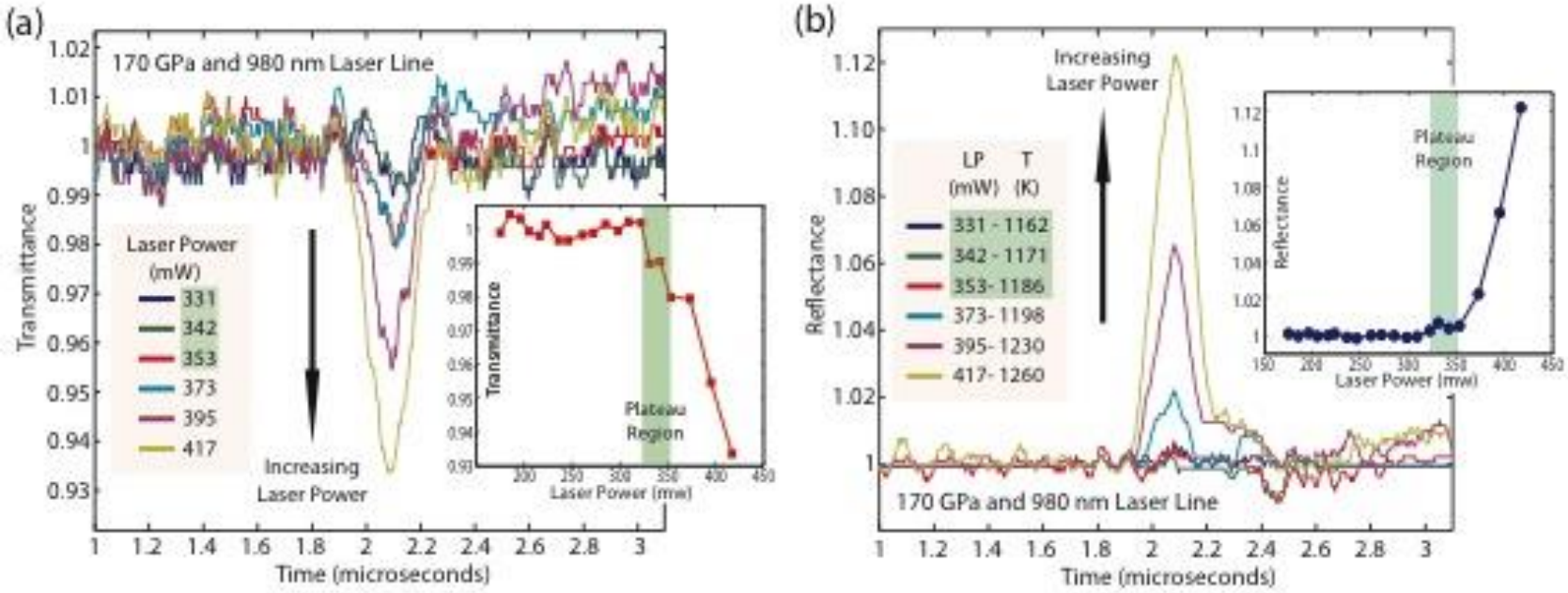
Thank you


Hydrogen
P (GPa)
H2-PRE
82 K
421
x5
82 K
385
x5
5 K
368
x5
H2-PRE
5 K
351
x5
III
5 K
343
5 K
317
82 K
304
82 K
243
III
82 K
183
II & III
82 K
x10
151
Absorbance
2500
3000
3500
4000
4500
5000
Frequency (cm-1)


Hydrogen
T = 82 ± 2 K
BSP (II)
A (III)
H2-PRE
Frequency (cm-1)
Pressure (GPa)
4750
4500
4250
4000
3750
3500
3250
3000
2750
100
150
200
250
300
350
400
450


1800
1700
1600
1500
1400
1300
Peak Temperature (K)
300
400
500
600
Laserpower (mW)
120 GPa
W film + Al_2O_3
Bare W film (1)
Bare W film (2)
Pt foil


(a)
20 μm
(b)
20 μm
(c)
Peak Temperature (K)
2000
1800
1600
1400
1200
1000
800
200
300
400
500
600
Laserpower <P> (mW)
130 GPa
Run 1
Run 2


(a)
170 GPa and 980 nm Laser Line
Transmittance
Laser Power (mW)
331
342
353
373
395
417
Increasing Laser Power
Time (microseconds)
Transmittance
Plateau Region
Laser Power (mW)
(b)
Increasing Laser Power
Reflectance
LP (mW)
T (K)
331 - 1162
342 - 1171
353- 1186
373- 1198
395- 1230
417- 1260
170 GPa and 980 nm Laser Line
Time (microseconds)
Reflectance
Plateau Region
Laser Power (mW)