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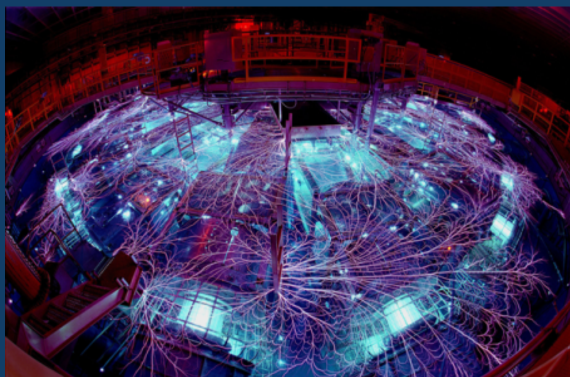

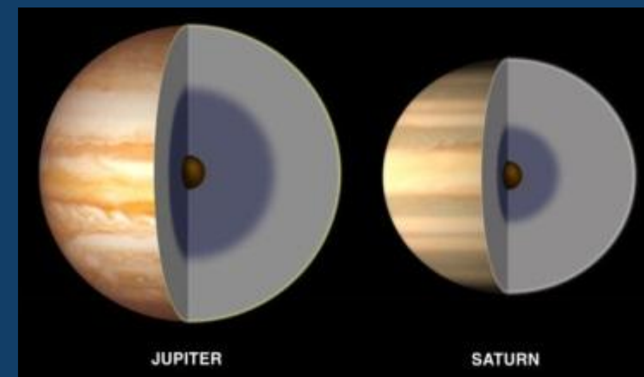

UNIVERSITÄT ROSTOCK
gegründet 1419
INSTITUT FÜR PHYSIK

Hydrogen at planetary conditions -
direct observation of an abrupt insulator-to-metal transition in dense liquid deuterium

Ronald Redmer & Marcus D. Knudson

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Albuquerque, NM

U.S. DEPARTMENT OF ENERGY
NNSA National Nuclear Security Administration

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Mike Desjarlais

Andreas Becker

Winfried Lorenzen

Ronald Redmer

Planetary Modeling

Nadine Nettelmann

Andreas Becker

Robert Püstow

Ronald Redmer

Pulse Shaping

Ray Lemke

Jean-Paul Davis

Mark Savage

Ken Struve

Keith LeChien

Brian Stoltzfus

Dave Hinshelwood

Entire Z crew

University of Rostock

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1. **Interior of giant planets: hydrogen phase diagram at high pressures**

2. **Search for metallic hydrogen and hypothetical plasma phase transition**

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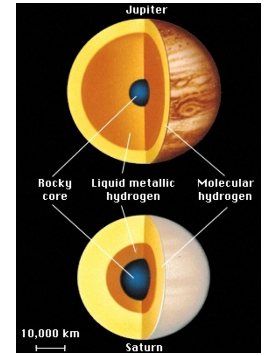
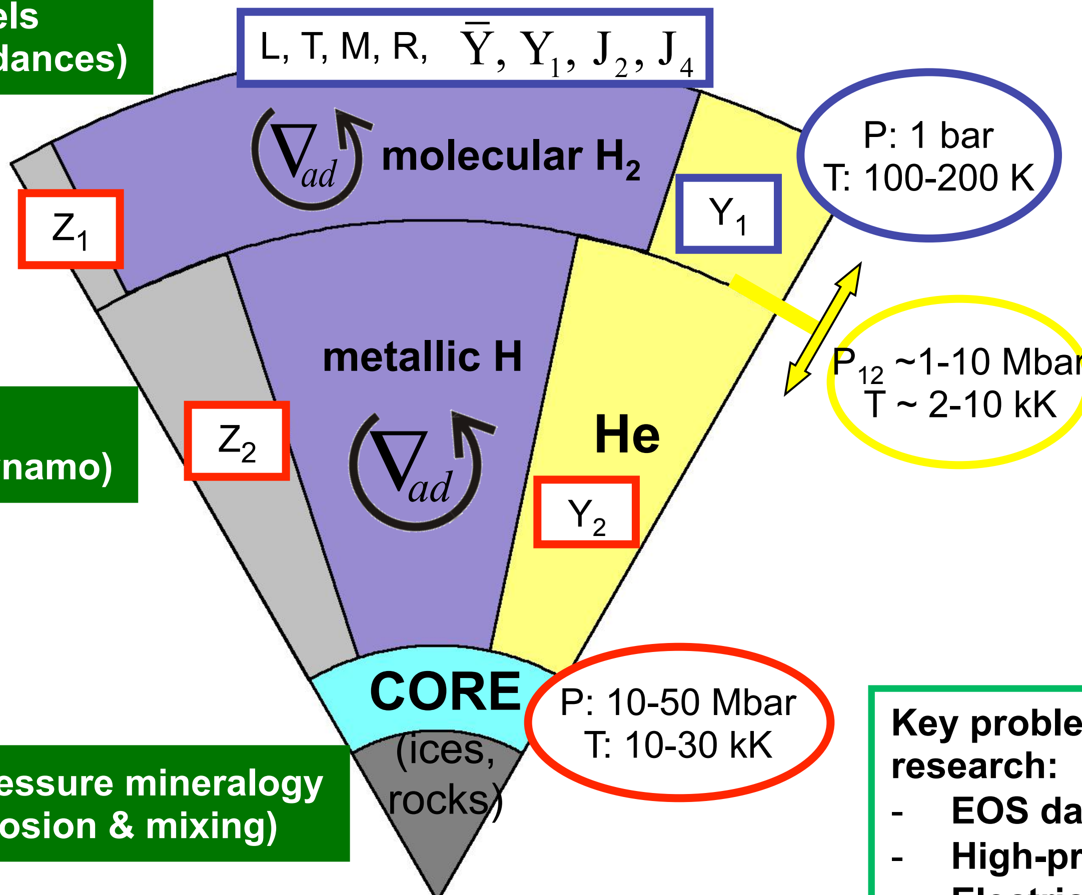
4. **Consequences for giant planets – structure and evolution of Jupiter and Saturn**

Interior of gas giants: H-He

three-layer model, input and constraints



Atmosphere models (luminosity, abundances)



Magnetic field generation (dynamo)

High-pressure mineralogy (core erosion & mixing)

Existence, origin and location of a layer boundary…
→ H metalization
→ H-He demixing

Key problems in warm dense matter research:

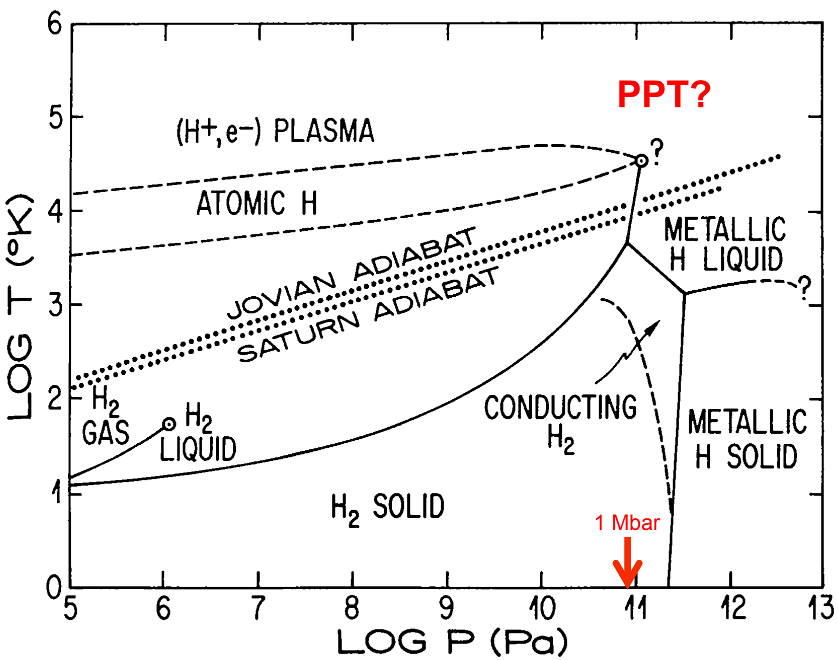
- **EOS data**
- **High-pressure phase diagram**
- **Electrical & thermal conductivity**
- **Diffusion & viscosity**

constraints
results from modeling
free parameter

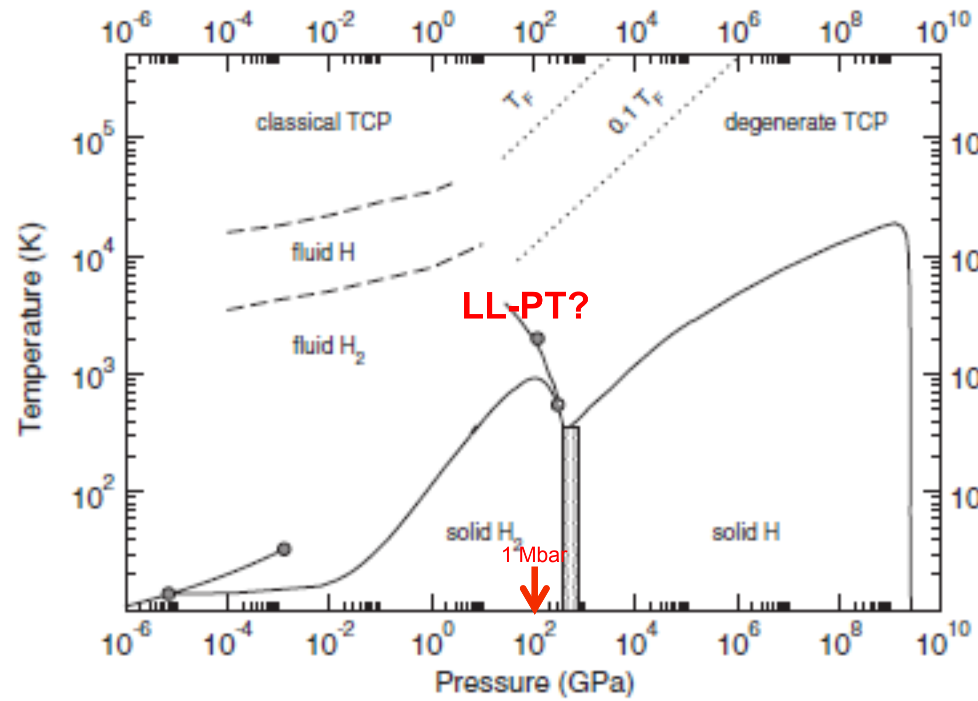
See e.g. D.J. Stevenson (1982), T. Guillot (1999), N. Nettelmann et al. (2008, 2012)

High-pressure phase diagrams - important for understanding planets

First-order phase transitions have been proposed:
Plasma Phase Transition (PPT)
Liquid-Liquid Phase Transition (LL-PT)

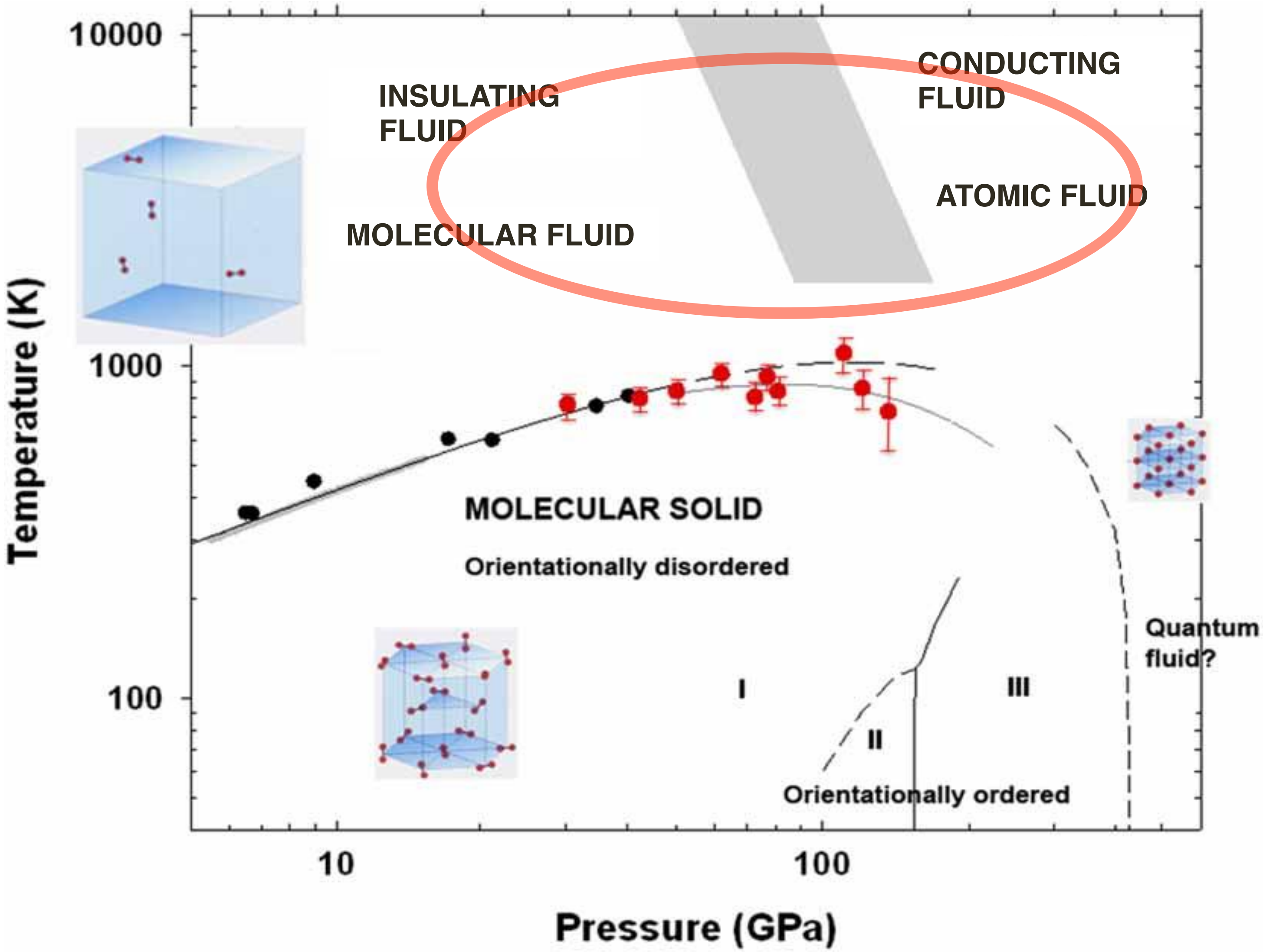


D.J. Stevenson, Ann. Rev. Earth Planet. Sci. **10**, 257 (1982)



J.M. McMahon et al., Rev. Mod. Phys. **84**, 1607 (2012)
See also R.J. Hemley, High Press. Res. **30**, 581 (2010)

Hydrogen at high pressures – the known phase diagram so far



R.J. Hemley, High Press Res. 30, 581 (2010)

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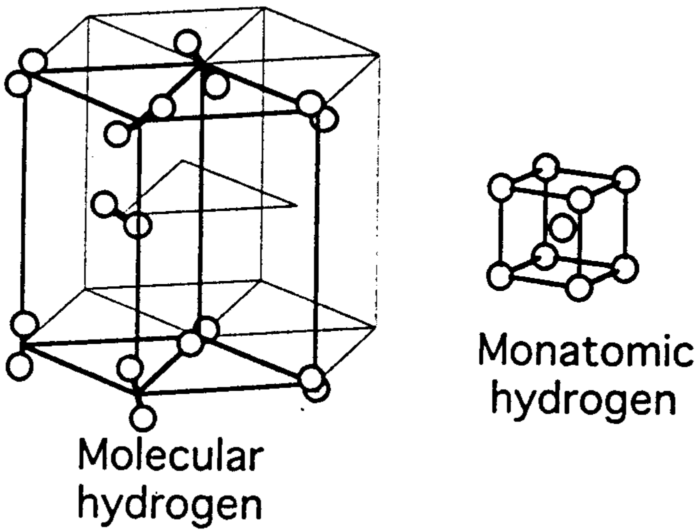
Solid metallic hydrogen at T=0 K?

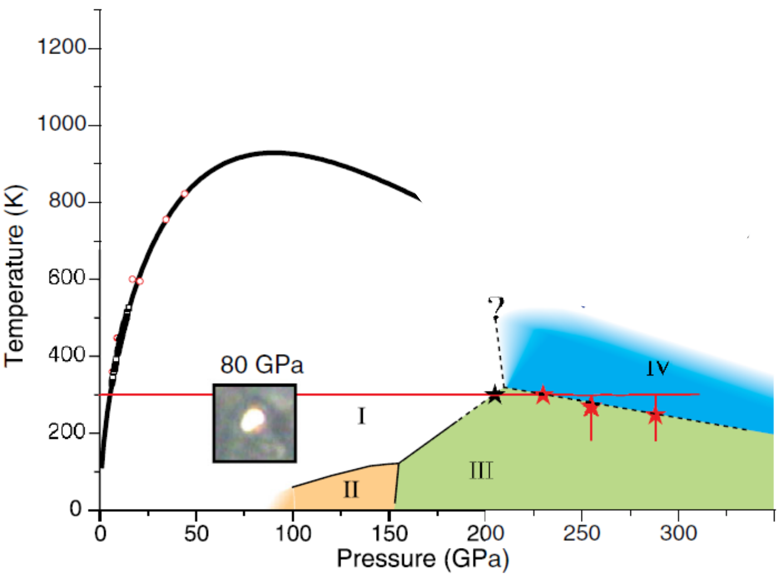
Proposed by Wigner and Huntington already in 1935 (at 25 GPa).

Not verified in **DAC** experiments up to 3.5 Mbar so far.

But rich phase diagram obtained in solid H: phases I, II, III, IV.



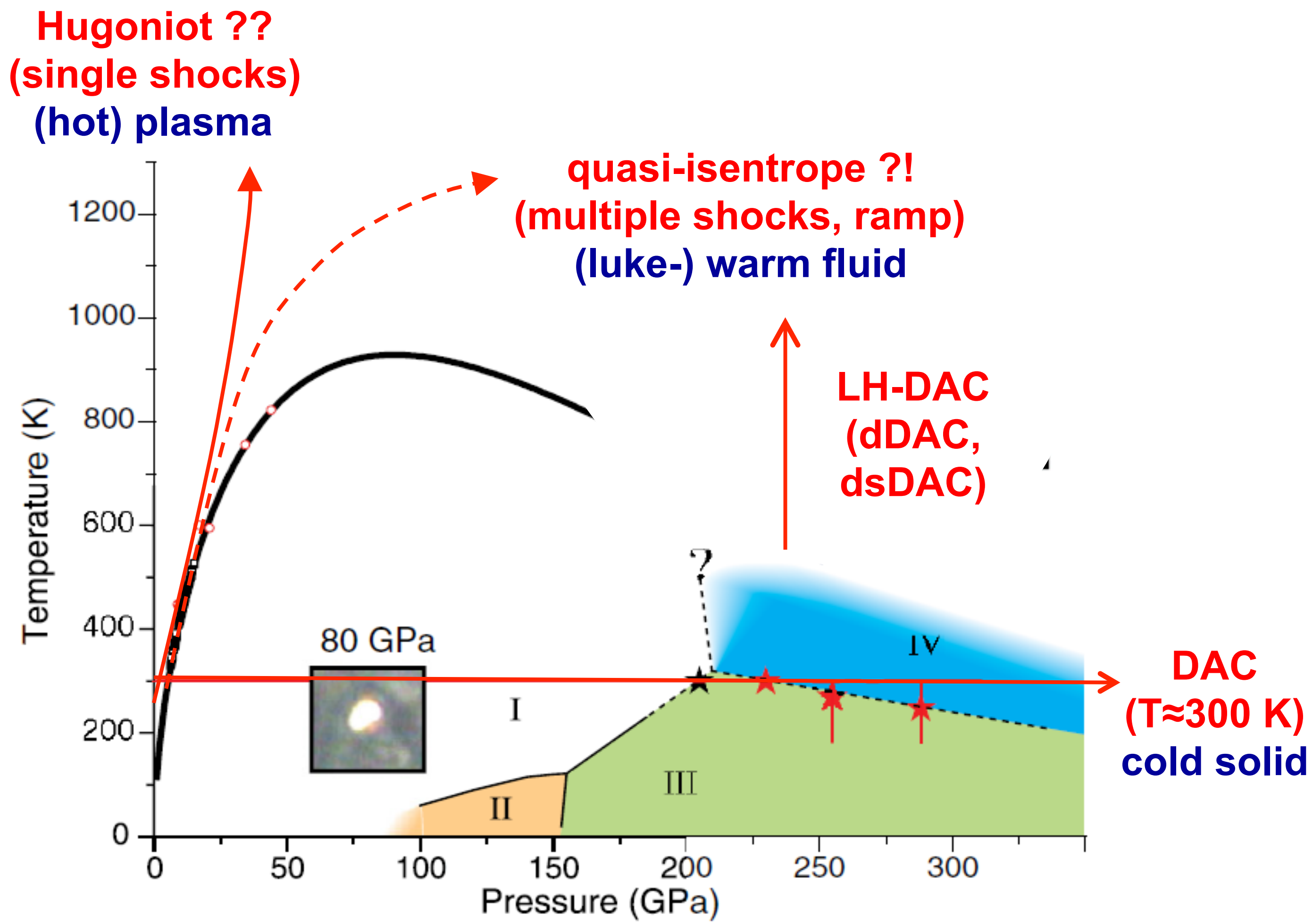
H.K. Mao & R.J. Hemley, RMP **66**, 671 (1994)



R.T. Howie et al., PRL **108**, 125501 (2012)

Metallization by band-gap closure:
From 15 eV in the hcp molecular insulator to zero in the bcc metal

Liquid metallic hydrogen at T>0 K?



R.T. Howie et al., PRL **108**, 125501 (2012)

Higher temperatures: chemical models for the fluid and plasma region

$$F(T,V,\{N_c\}) = F_0 + F_\pm + F_{pol}$$

- $\mathbf{F_0}$ neutral fluid: H, H_2
- $\mathbf{F_\pm}$ plasma component: e, p, H^-, H_2^+
- $\mathbf{F_{pol}}$ polarization term: e-H, e-H_2
- multicomponent system: c = e, p, H, H_2
- chemical reactions: $H_2 \leftrightarrow 2H$, $H \leftrightarrow e+p$, …
- chemical equilibrium: $\mu_{H_2} = 2\mu_H$, $\mu_H = \mu_e + \mu_p$, …

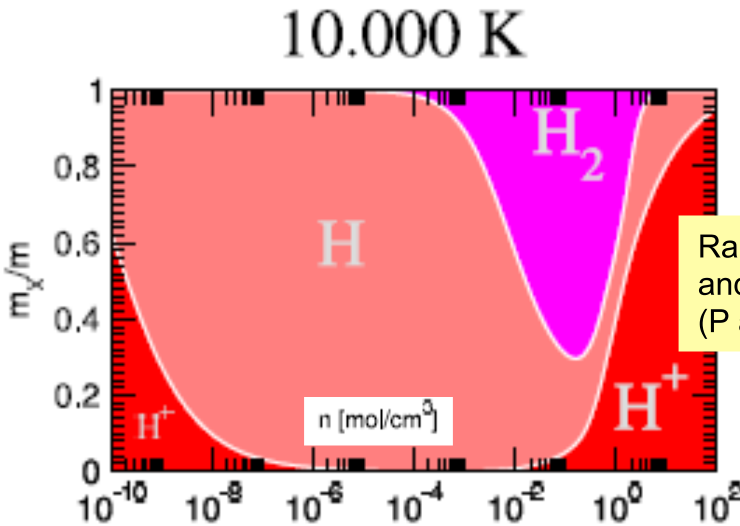
EOS: $F = U - TS$

$$p=-(dF/dV)_{T,N} \; , \; \mu_c = (dF/dN_c)_{T,V} \; , \; S=-(dF/dT)_{V,N}$$

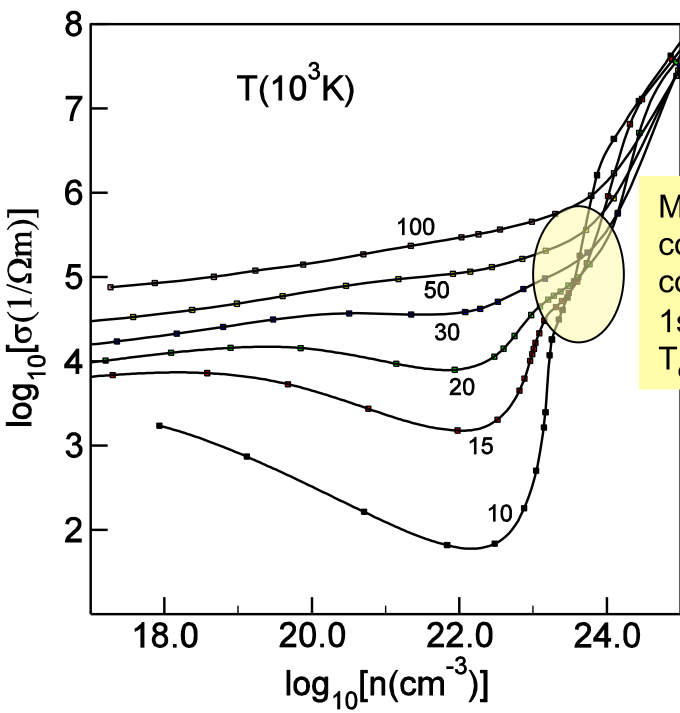
Landau & Zeldovich (1944) and Norman & Starostin (1968) …
Partially ionized plasma with pressure dissociation and ionization

W. Ebeling et al., 1970 – 2000 …
D. Saumon, G. Chabrier, H.M. Van Horn, ApJS **99**, 713 (1995)
H. Juranek et al., JCP **112**, 3780 (2000), **117**, 1768 (2002)

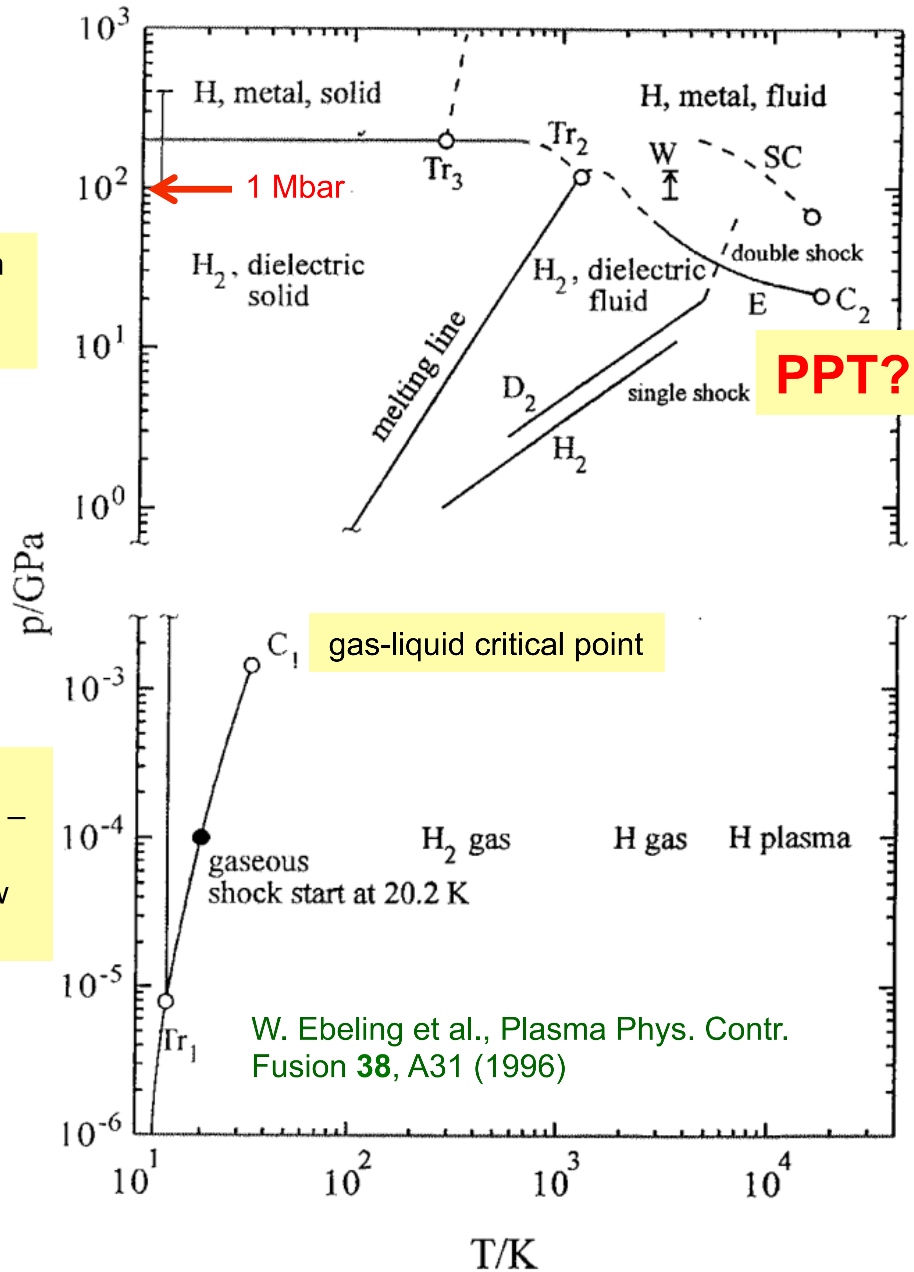
Phase diagram at high pressures - PPT?



Rapid dissociation and ionization (P and T driven)



Minimum metallic conductivity (Mott) – connected with a 1st-order PT below T_{cr}?



W. Ebeling et al., Plasma Phys. Contr. Fusion **38**, A31 (1996)

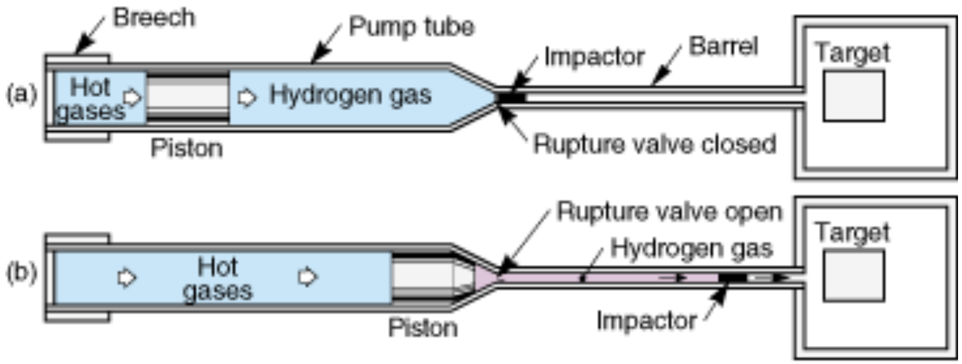
Critical point of the hypothetical PPT

Predicted by Zeldovich/Landau (1944) and Norman/Starostin (1968)

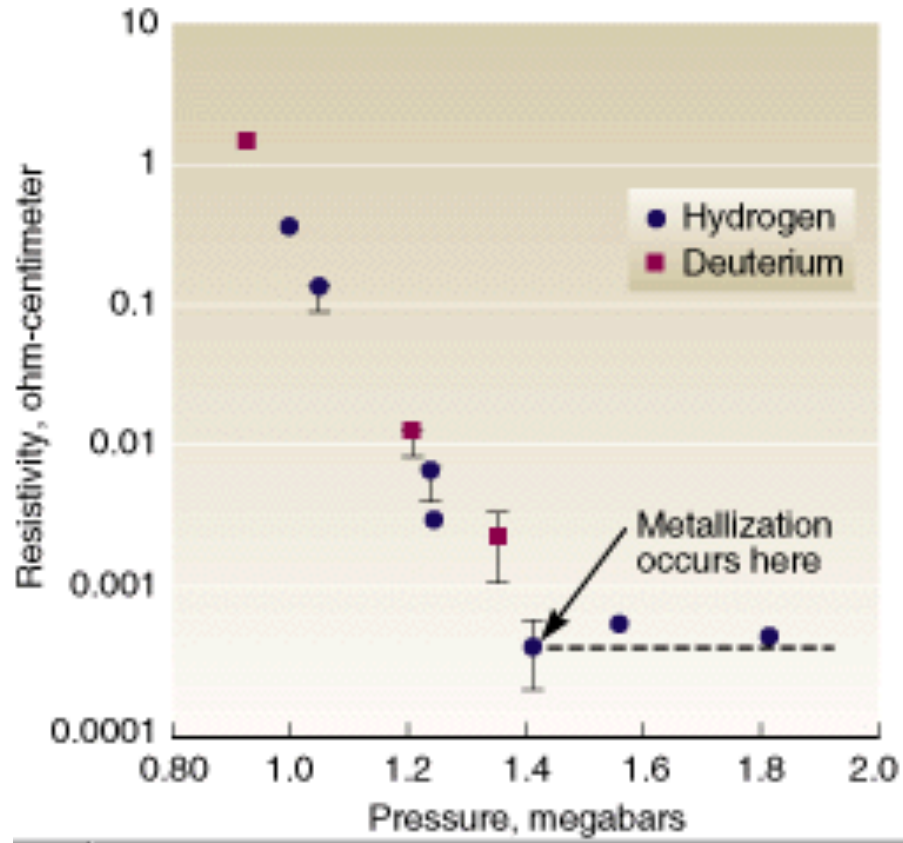
T_c (10^3 K)	P_c (GPa)	ρ_c (g/cm^3)	Method	Reference
12.6	95	0.95	PIP	Ebeling/Sändig (1973)
19	24	0.14	PIP	Robnik/Kundt (1983)
16.5	22.8	0.13	PIP	Ebeling/Richert (1985)
16.5	95	0.43	PIP	Haronska *et al.* (1987)
15.8	194	1.48	FIT	Hess *et al.* (1987)
15	64.6	0.36	PIP	Saumon/Chabrier (1991)
15.3	61.4	0.35	PIP	Saumon/Chabrier (1992)
14.9	72.3	0.29	PIP	Schlanges *et al.* (1995)
16.5	57	0.42	PIP	Reinholz *et al.* (1995)
11	55	0.25	PIMC	Magro *et al.* (1996)
20.9	0.3	0.002	FE model	Kitamura/Ichimaru (1998)
10	50	0.17	spin pol. H	Xu/Hansen (1999)
for 10^4 K between 0.1-1 g/cm^3			DPIMC	Filinov *et al.* (2004)
16.2		0.13	REMC	Kremp *et al.* (2005)
10-20	≈ 50	0.1-0.5	Exp.	PPT exists?

Almost all based on chemical models including pressure dissociation and ionization.

Gas gun experiments: fluid metallic hydrogen



(a) In the first stage of the gas gun (blue shading), hot-burning gases from gunpowder drive a piston, which in turn compresses hydrogen gas. (b) In the second stage (pink shading), the high-pressure gas eventually ruptures a second-stage valve, accelerating the impactor down the barrel toward its target.



Reverberating shock waves in sandwich target
→ quasi-isentropic process
→ low temperatures

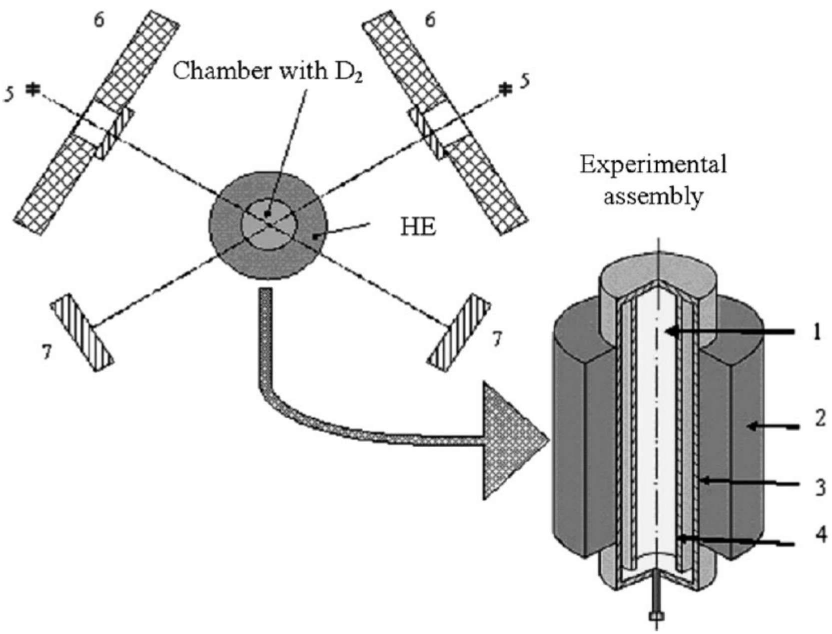
No indication of a first-order phase transition!

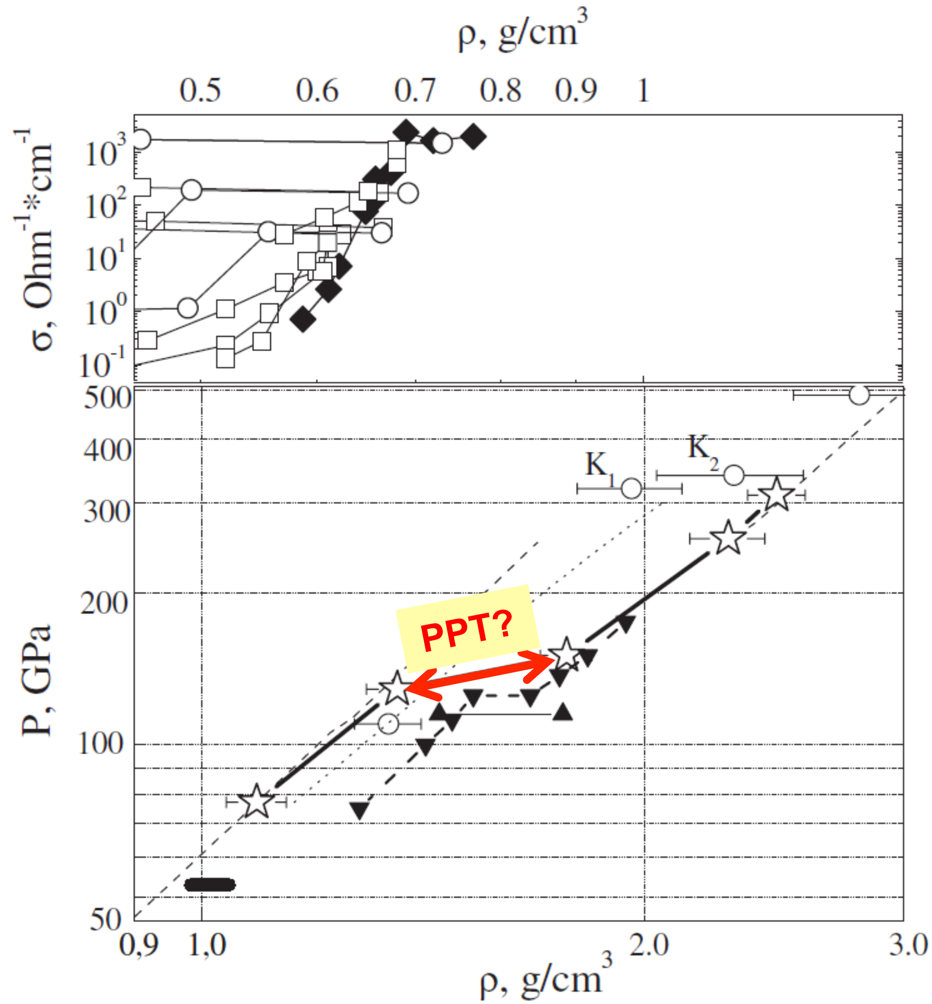
Metallic conductivity at ~3000 K and 1.4 Mbar (Wigner & Huntington 1935)

W.J. Nellis et al., PRL **68**, 2937 (1992)
S.T. Weir et al., PRL **76**, 1860 (1996)

Quasi-isentropic shock compression experiments using high explosives

V.E. Fortov et al., PRL **99**, 185001 (2007)





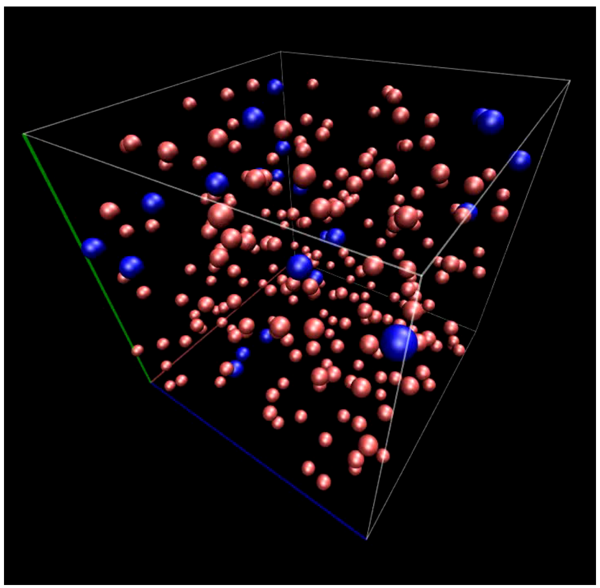
Direct experimental indication of a first-order PPT?

Chemical model SAHA-D used to evaluate the data – correct?

But strong increase in electrical conductivity describes clearly an insulator-to-metal transition

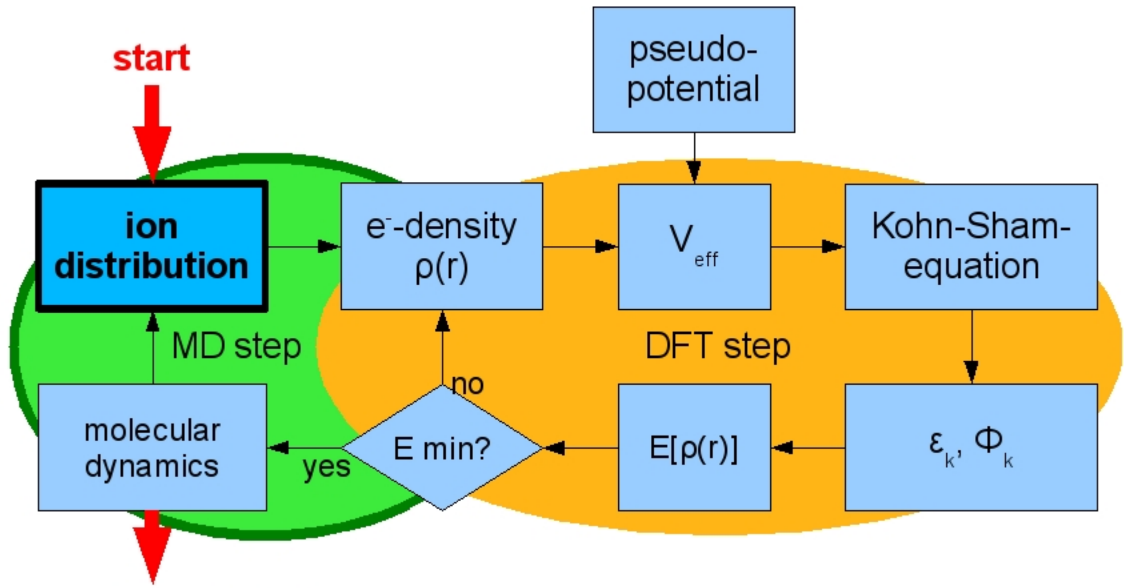
DFT-MD simulations: strict physical picture

No effective pair potentials or assuming bound states (H, H2) – nuclei and electrons!
Born-Oppenheimer approximation: combination of (quantum) DFT (e) and (classical) MD (nuclei).
Warm Dense Matter: finite-temperature DFT-MD simulations based on
N.D. Mermin, Phys. Rev. **137,** A1441 (1965)
Implemented e.g. in the Vienna Ab-initio Simulation Package (VASP) or Abinit …
G. Kresse and J. Hafner, PRB **47**, 558 (1993), ibid. **49**, 14251 (1994)
G. Kresse and J. Furthmüller, Comput. Mat. Sci. **6**, 15 (1996), PRB **54**, 11169 (1996)



H-He (8.6%) @ 1 Mbar, 4000 K

box length ~ 10^{-9} m

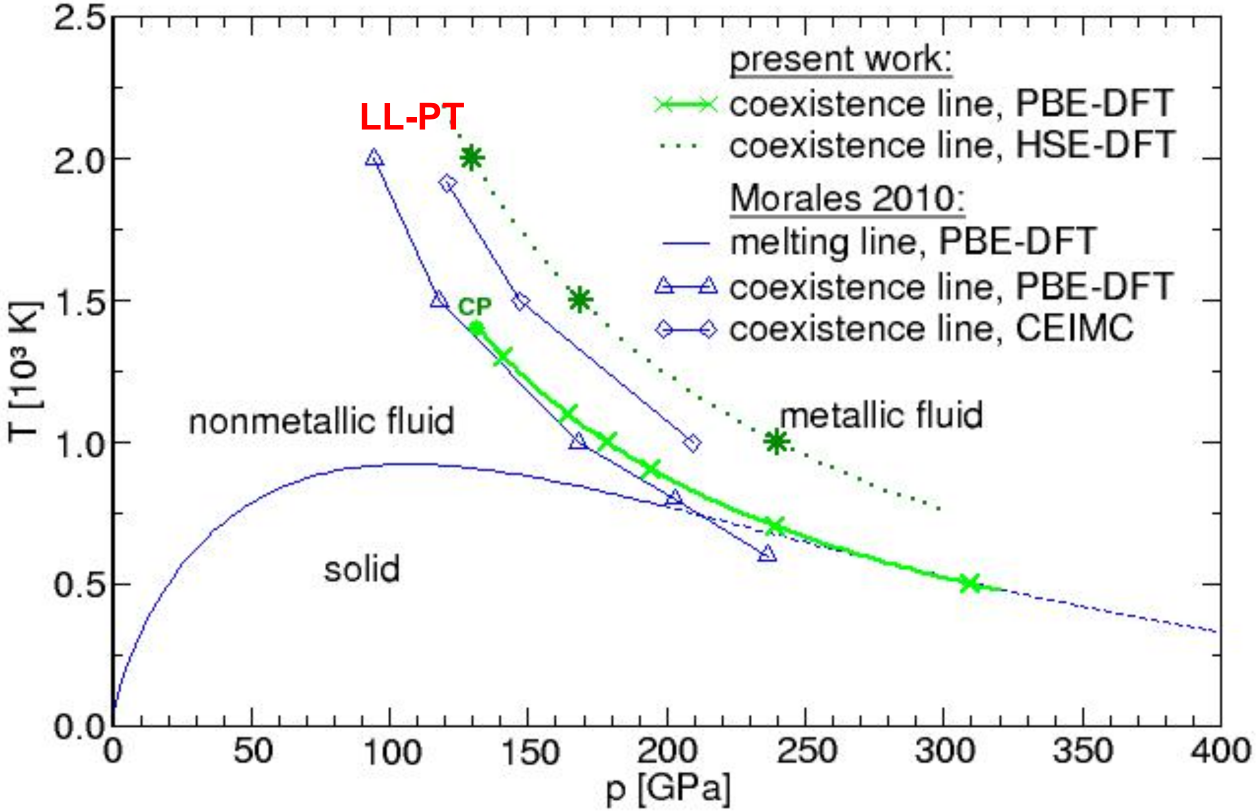


thermodynamic data
high-pressure phase diagram
pair correlation functions
electrical & thermal conductivity
diffusion coefficient
viscosity, opacity

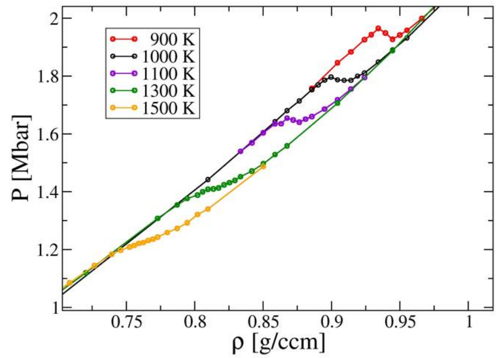
GP size ~ 10^8 m

H phase diagram at high pressure

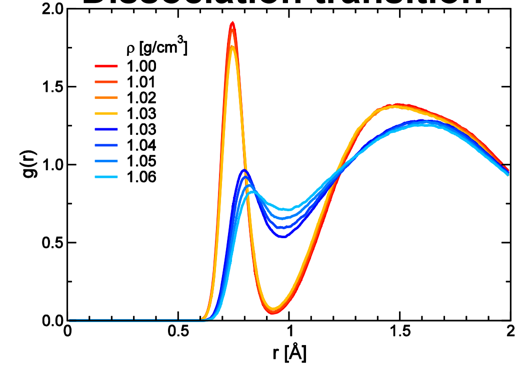
1st-order liquid-liquid phase transition predicted at much lower T (Scandolo 2003)



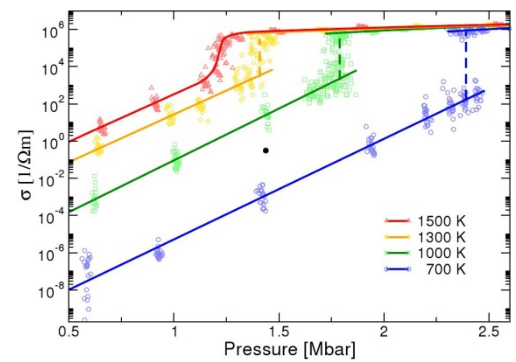
First-order phase transition



Dissociation transition



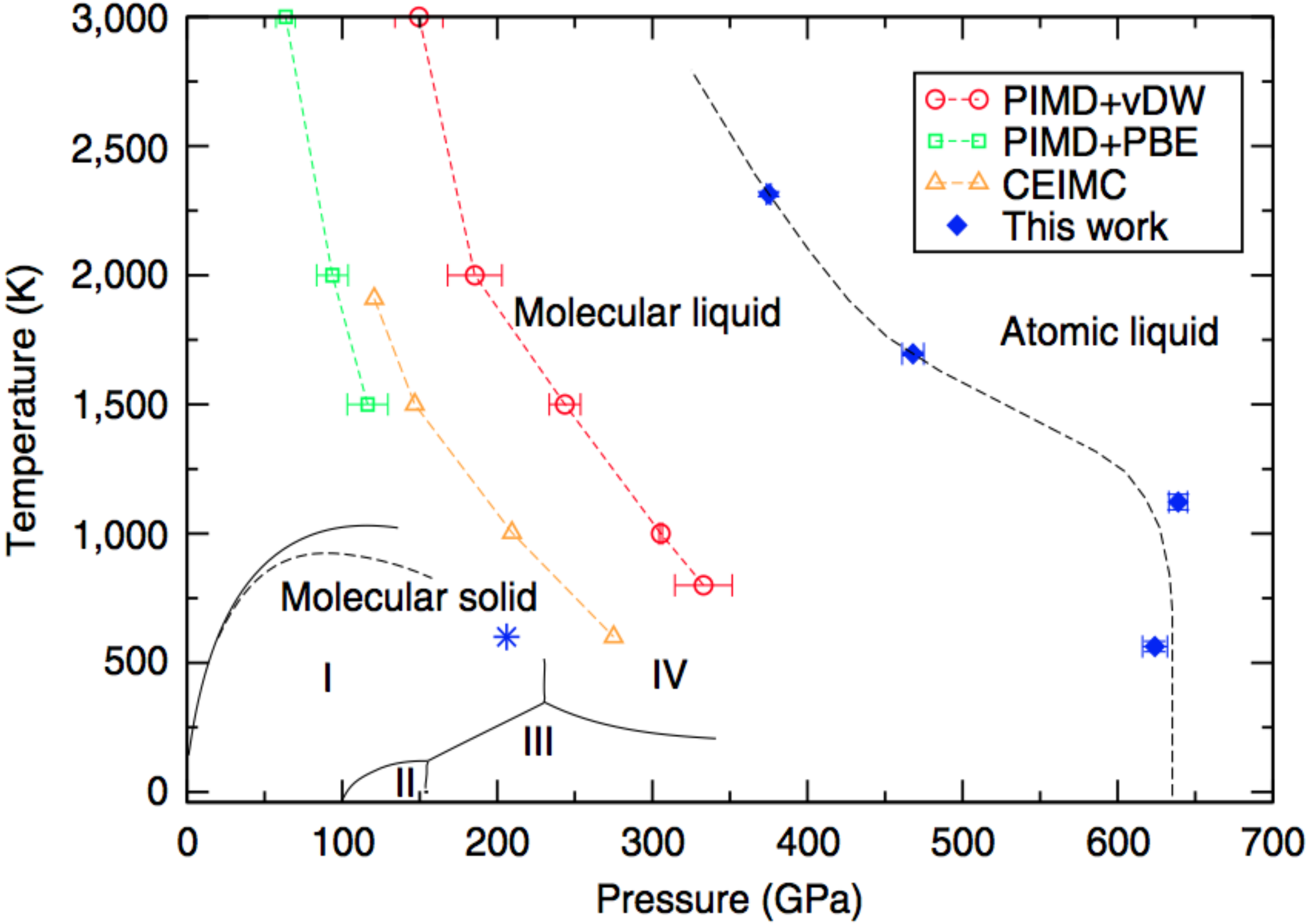
Nonmetal-to-metal transition



W. Lorenzen, B. Holst, R. Redmer, PRB **82**, 195107 (2010)
M.A. Morales et al., PNAS **107**, 12799 (2010)

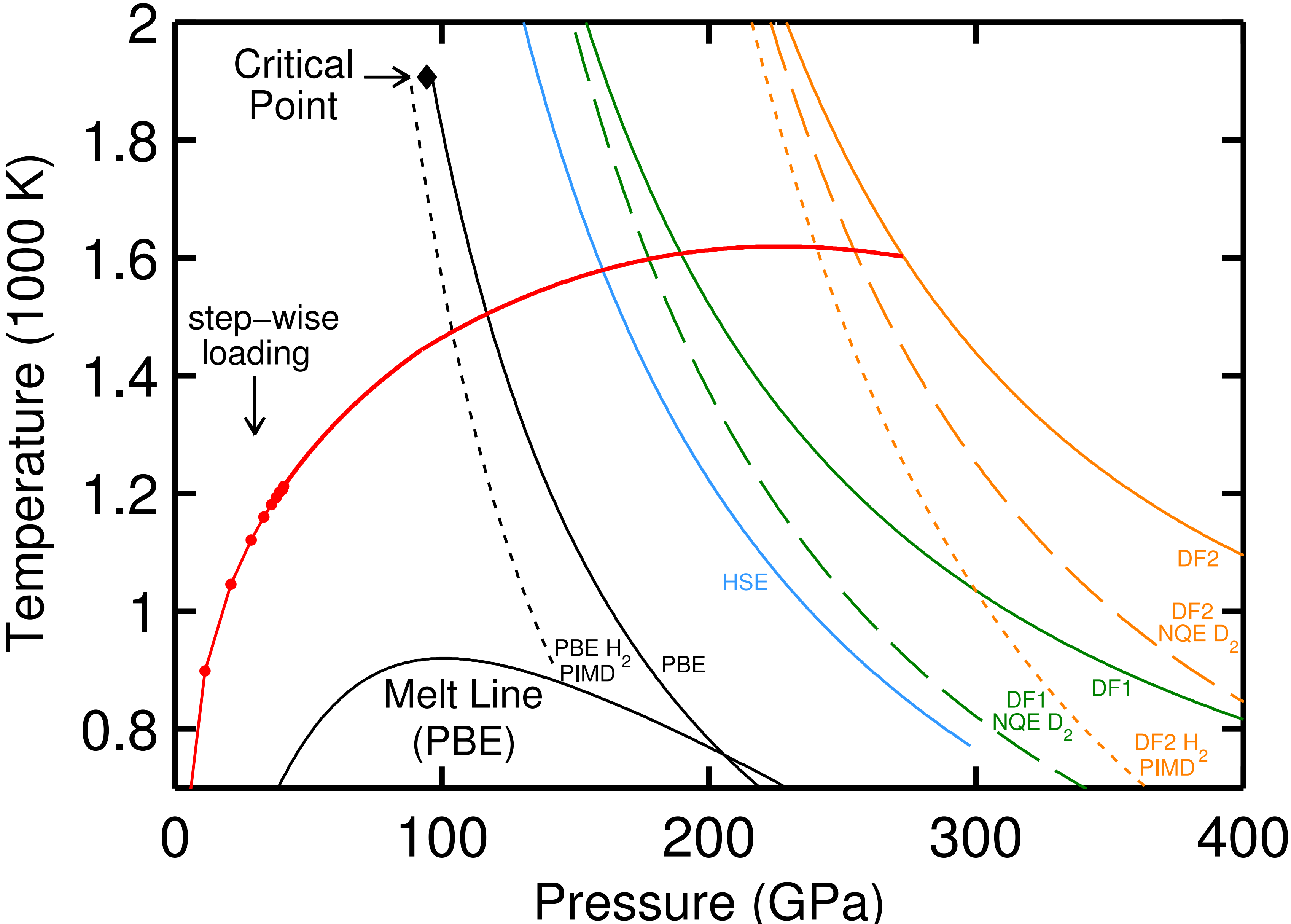
Problems: NQE, non-local XC functionals, vdW corrections … shift the transition, see M.A. Morales et al., PRL **110**, 065702 (2013)

Recent predictions of the LL-IMT in hydrogen



[illegible] atter (2014)

Proposed Experiment: Shock - Ramp



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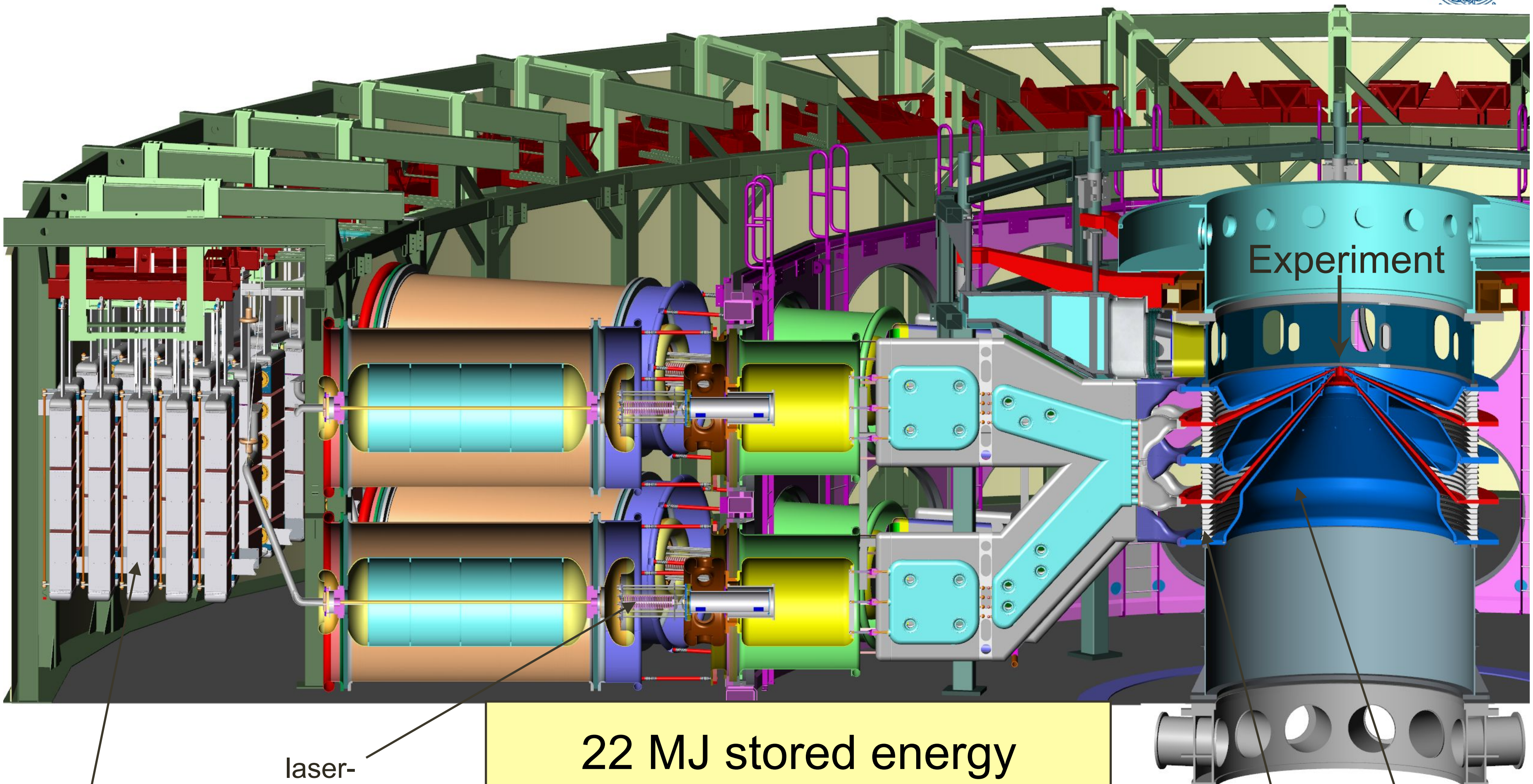
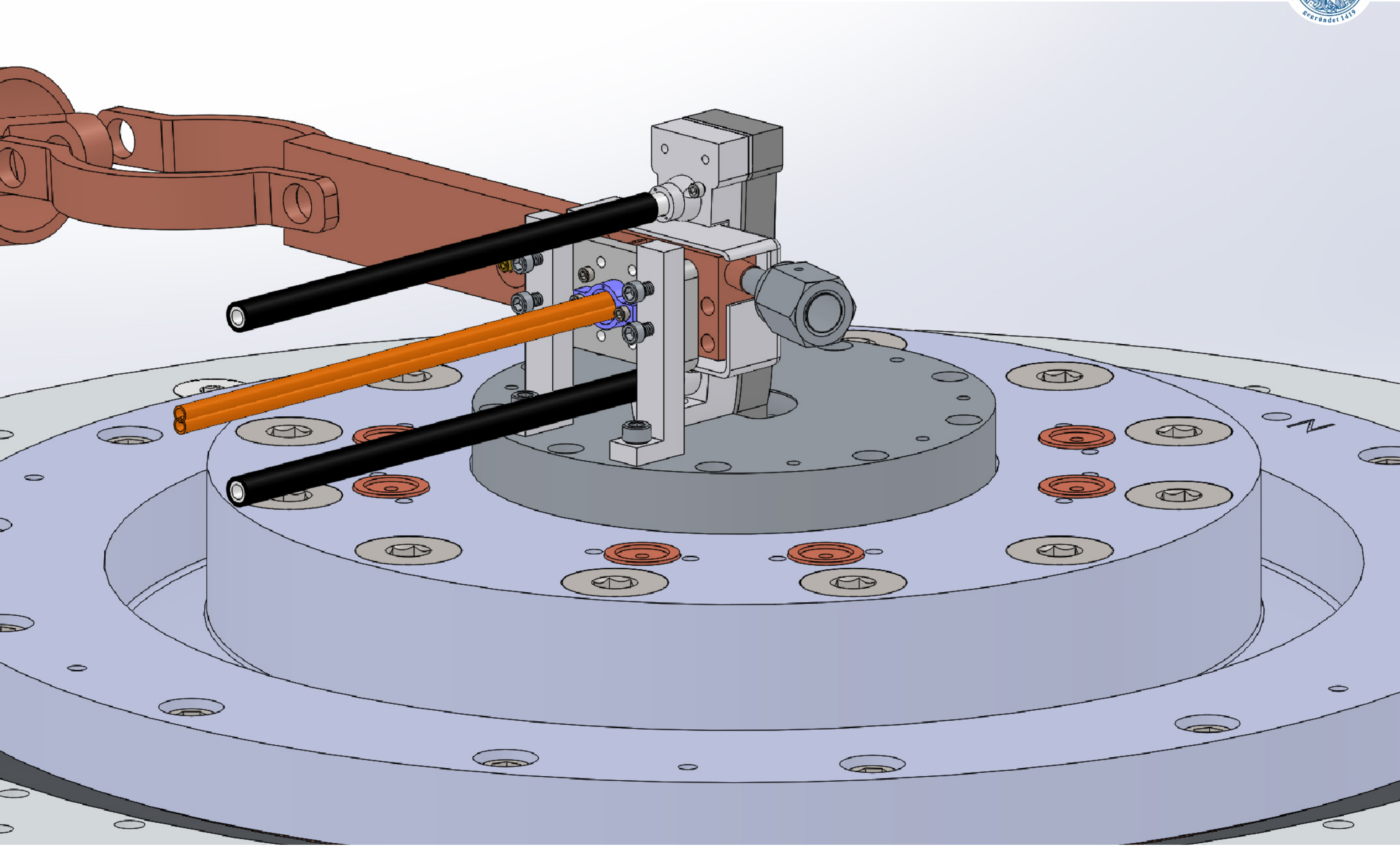
Sandia Z Machine

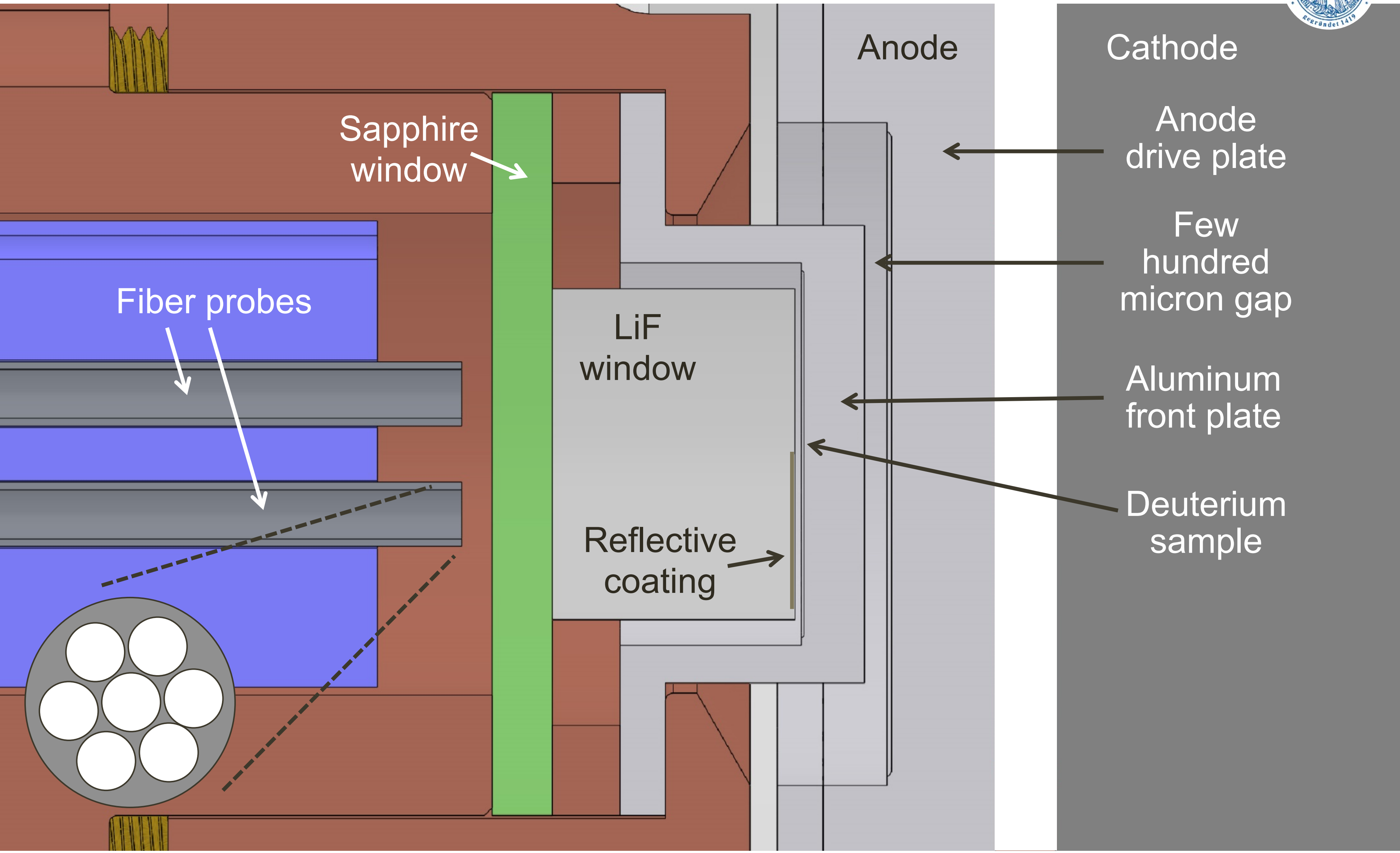


22 MJ stored energy
~25 MA peak current
~200-600 ns rise time

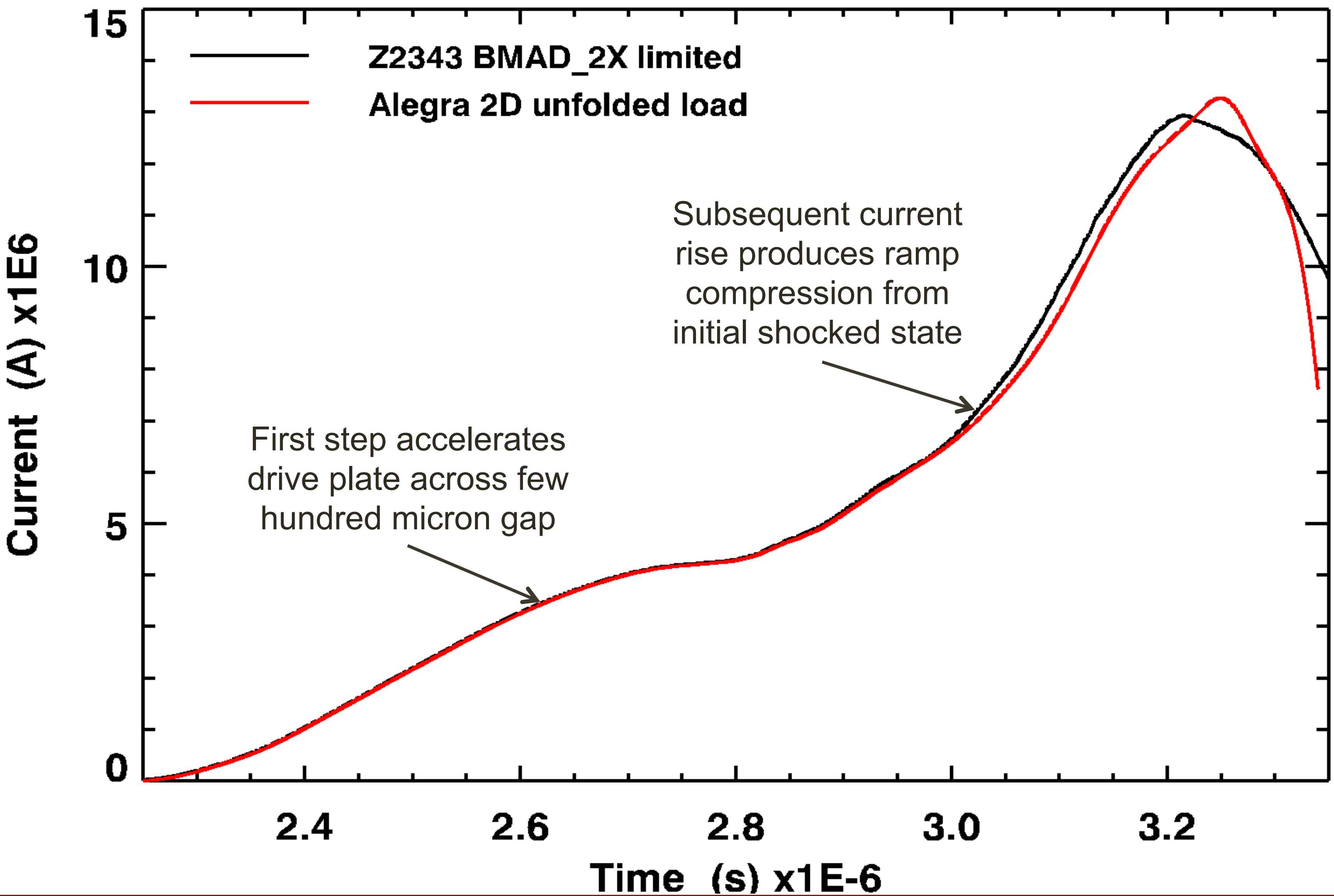
Stripline experimental configuration



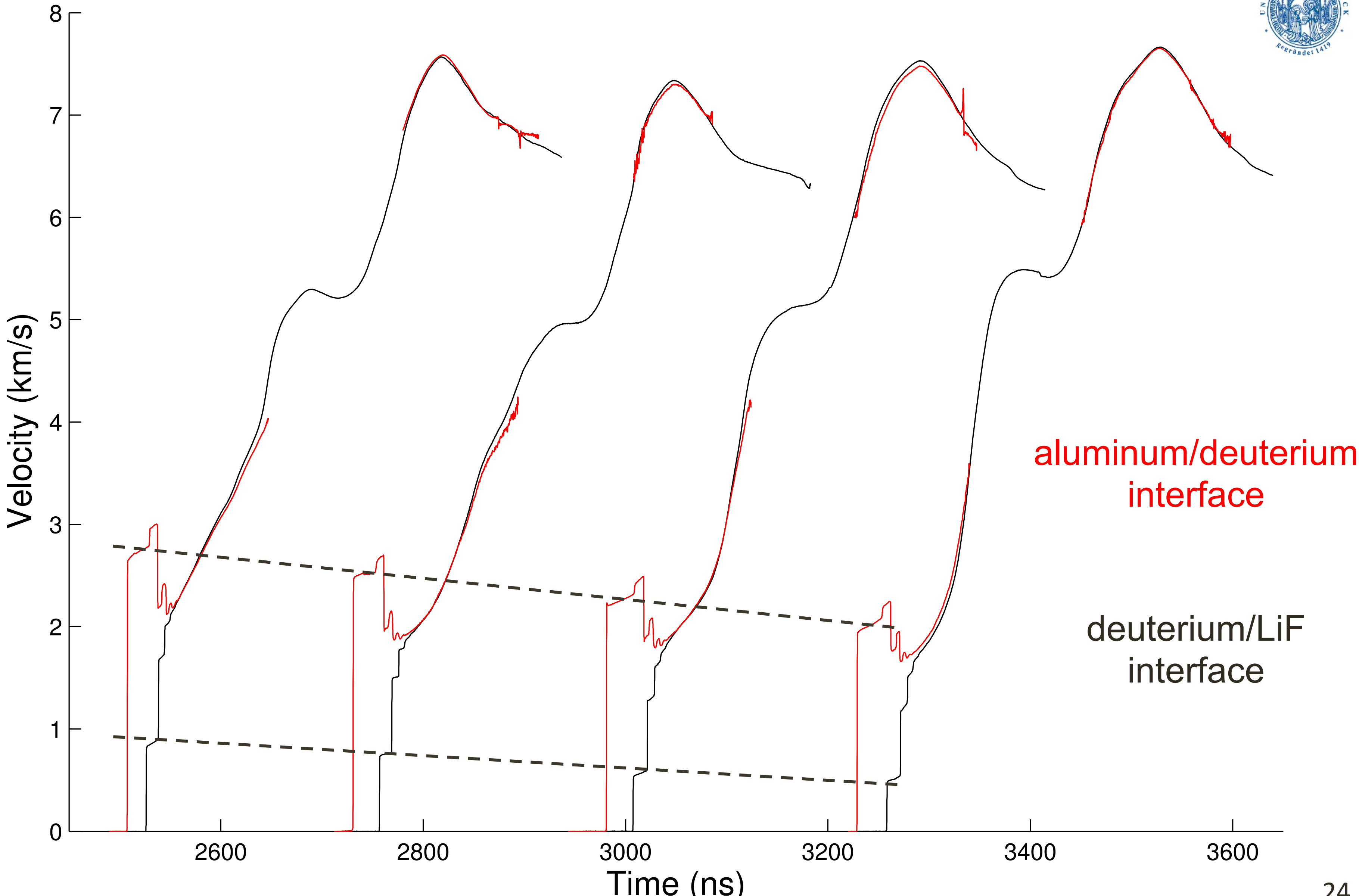
Experimental configuration



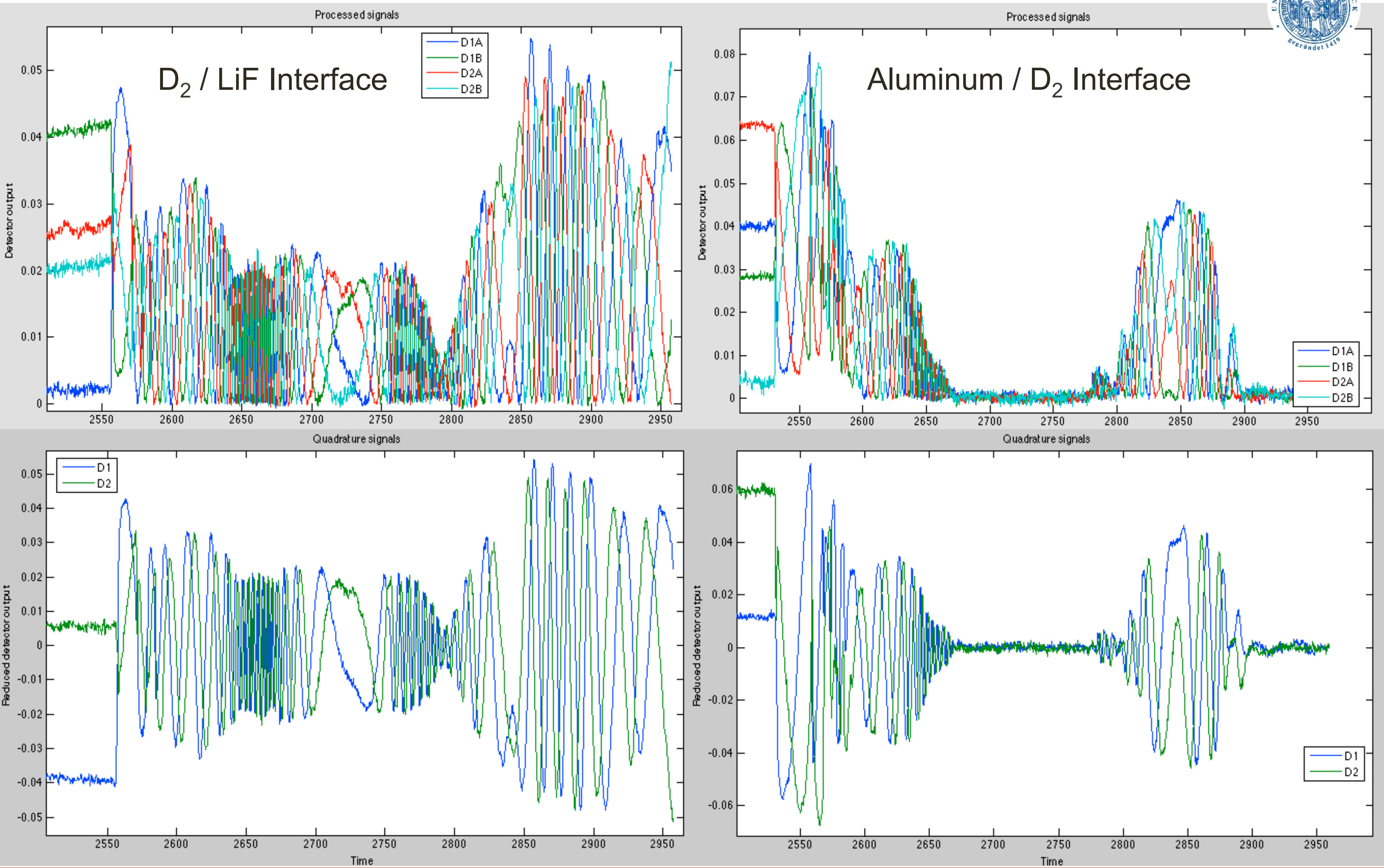
Two-step pulse shape provides shock-ramp profile



Stripline experimental profiles



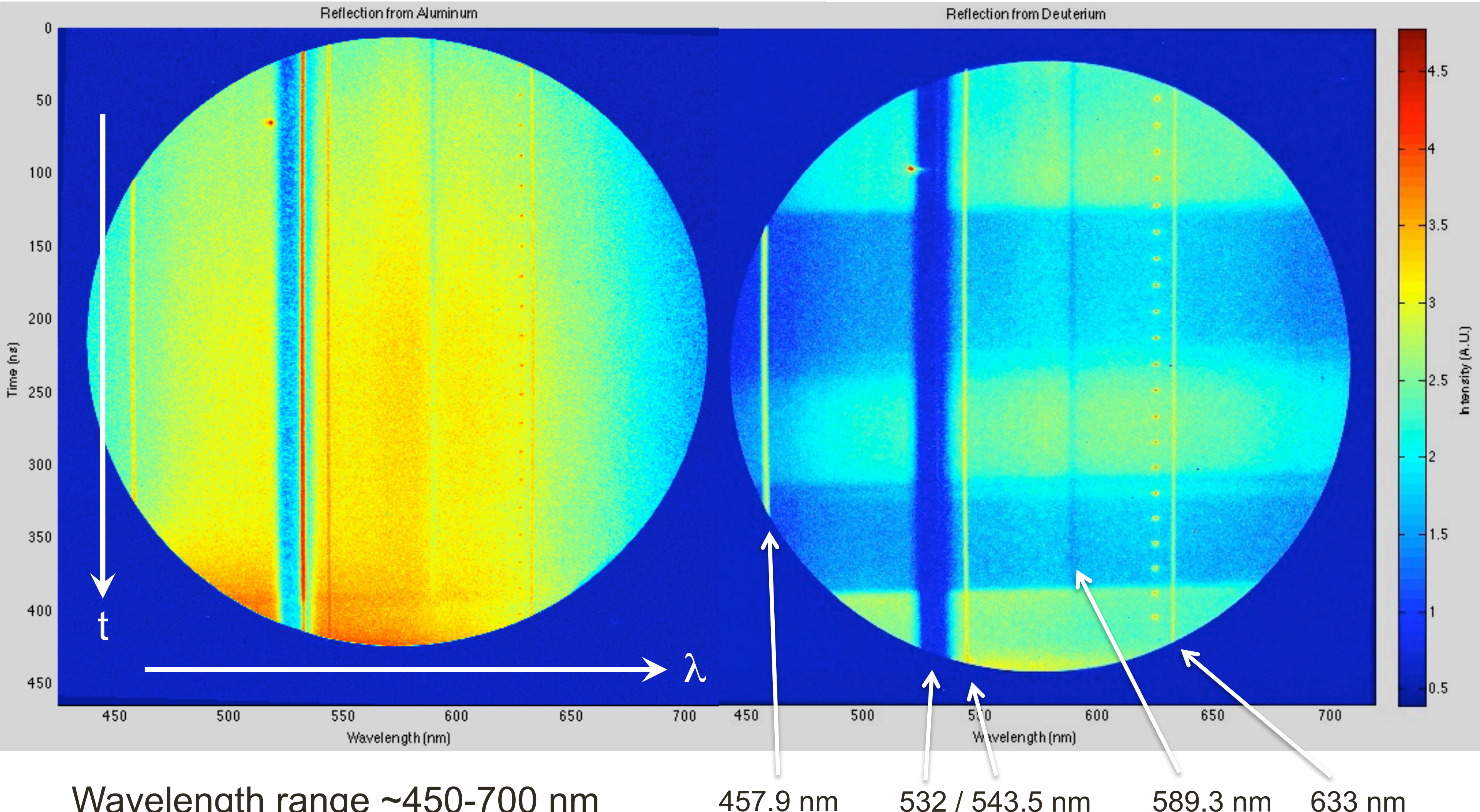
Processed VISAR signals



SVS system provides data to infer reflectivity

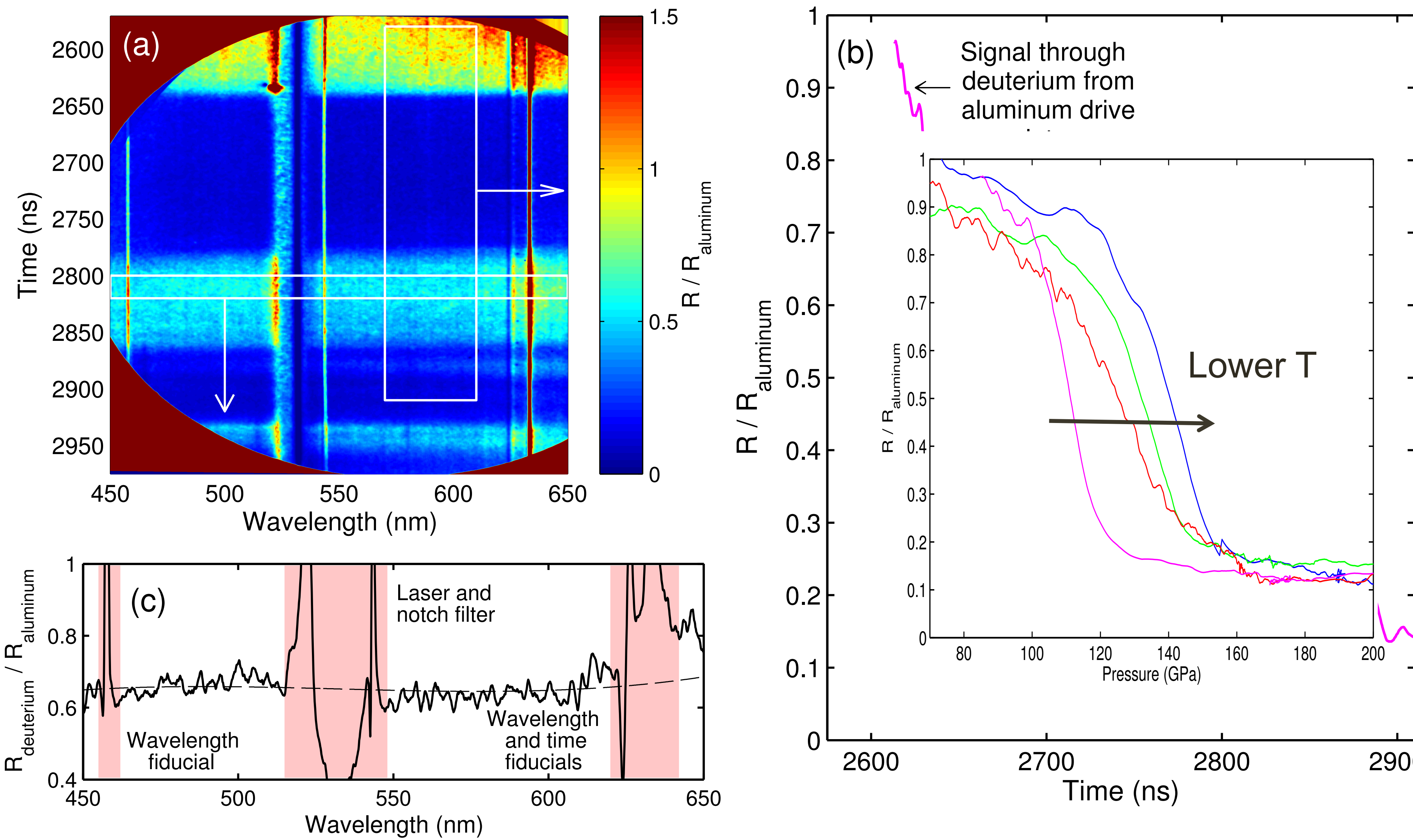
Reflection from aluminum coating

Reflection from deuterium

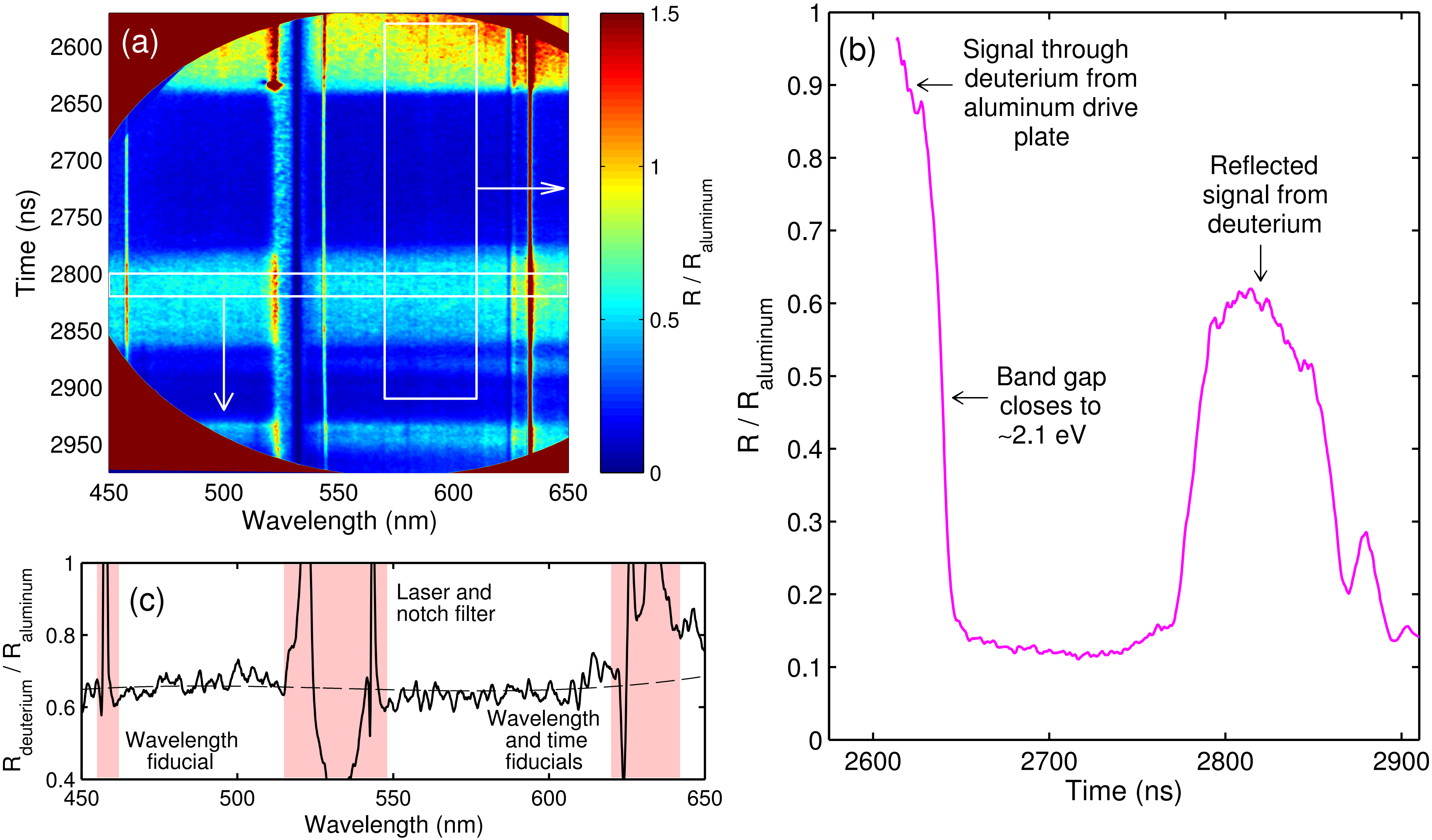


Wavelength range ~450-700 nm

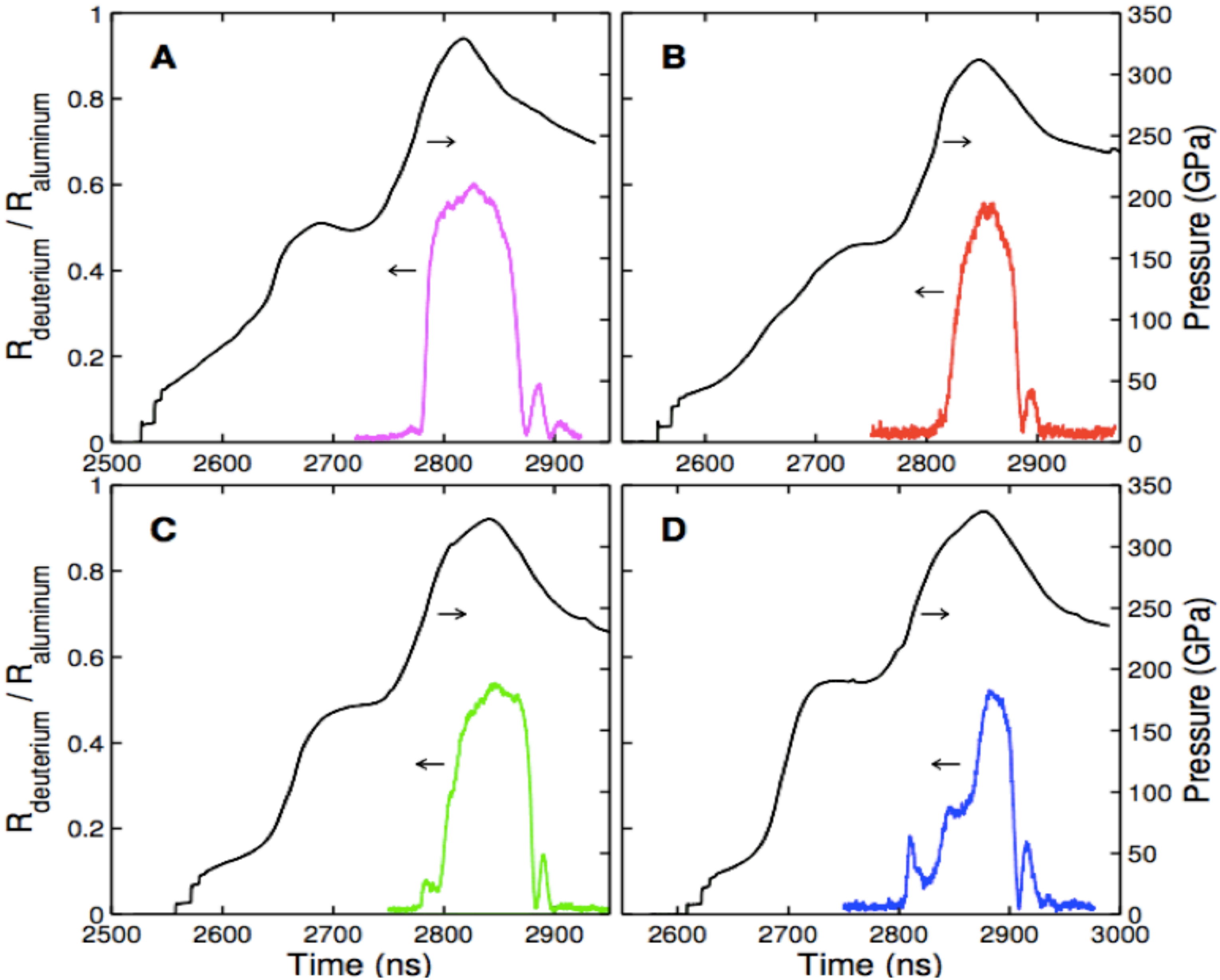
SVS system provides data to infer reflectivity



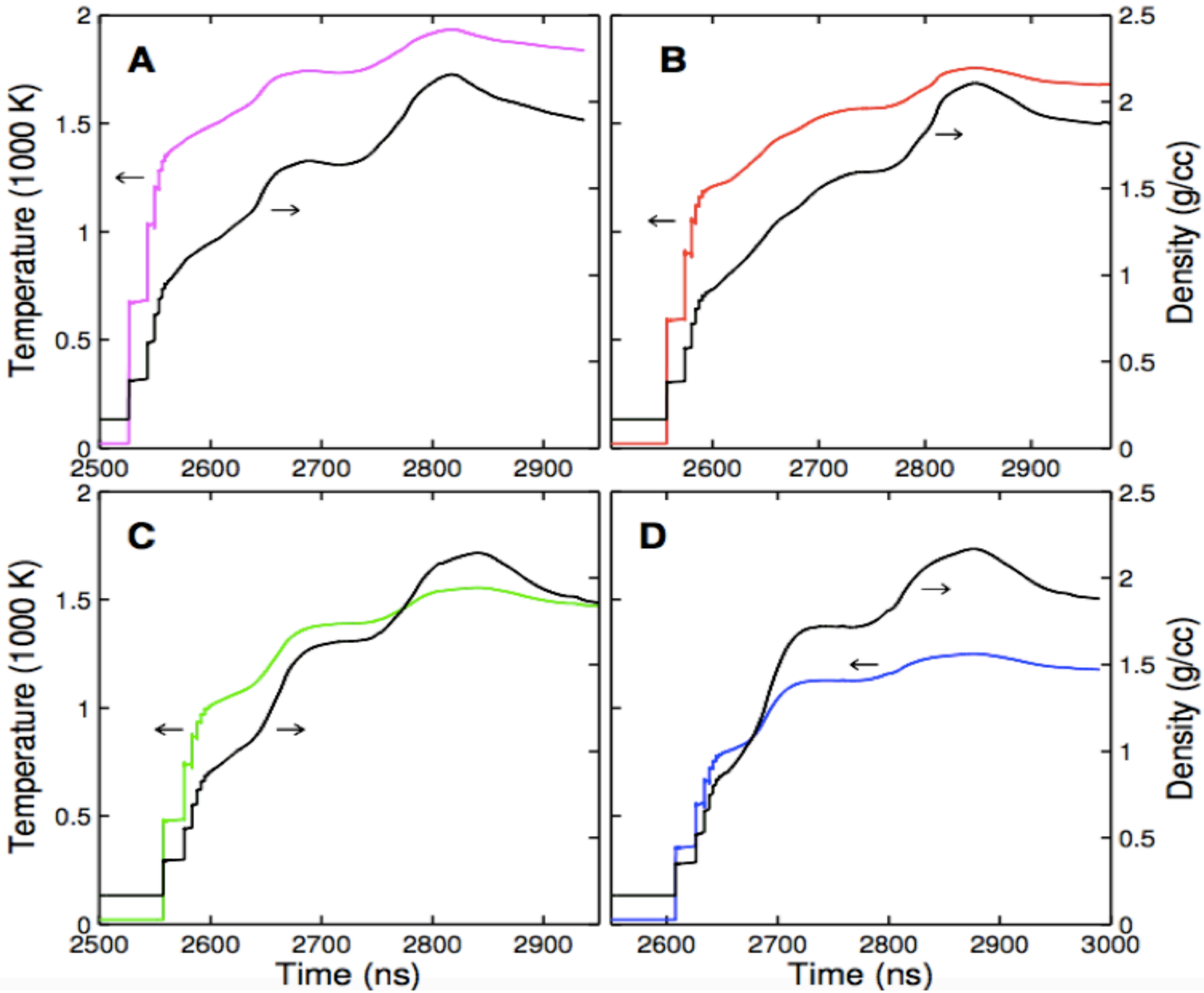
SVS system provides data to infer reflectivity



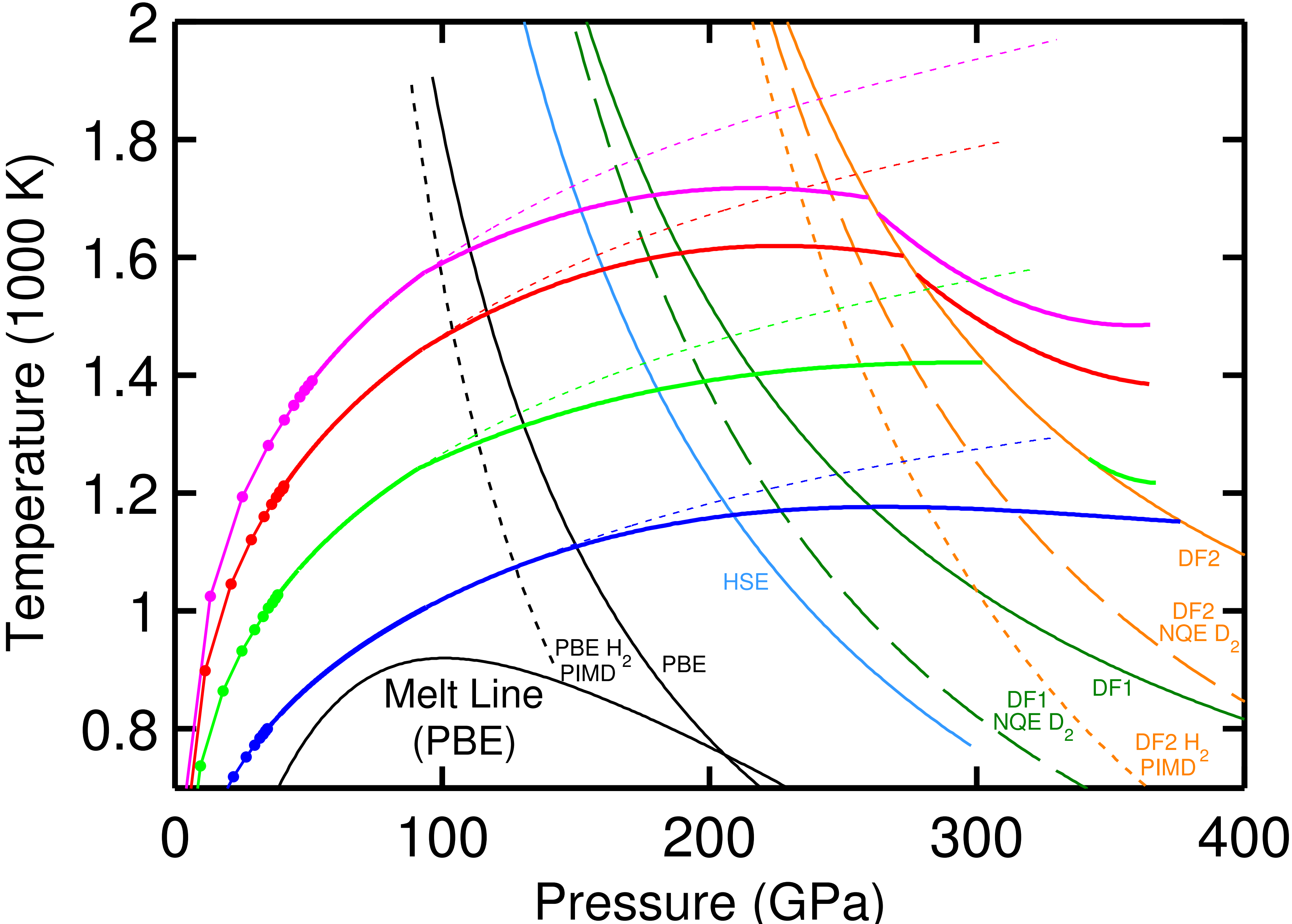
Reflectivity and pressure vs. time from VISAR



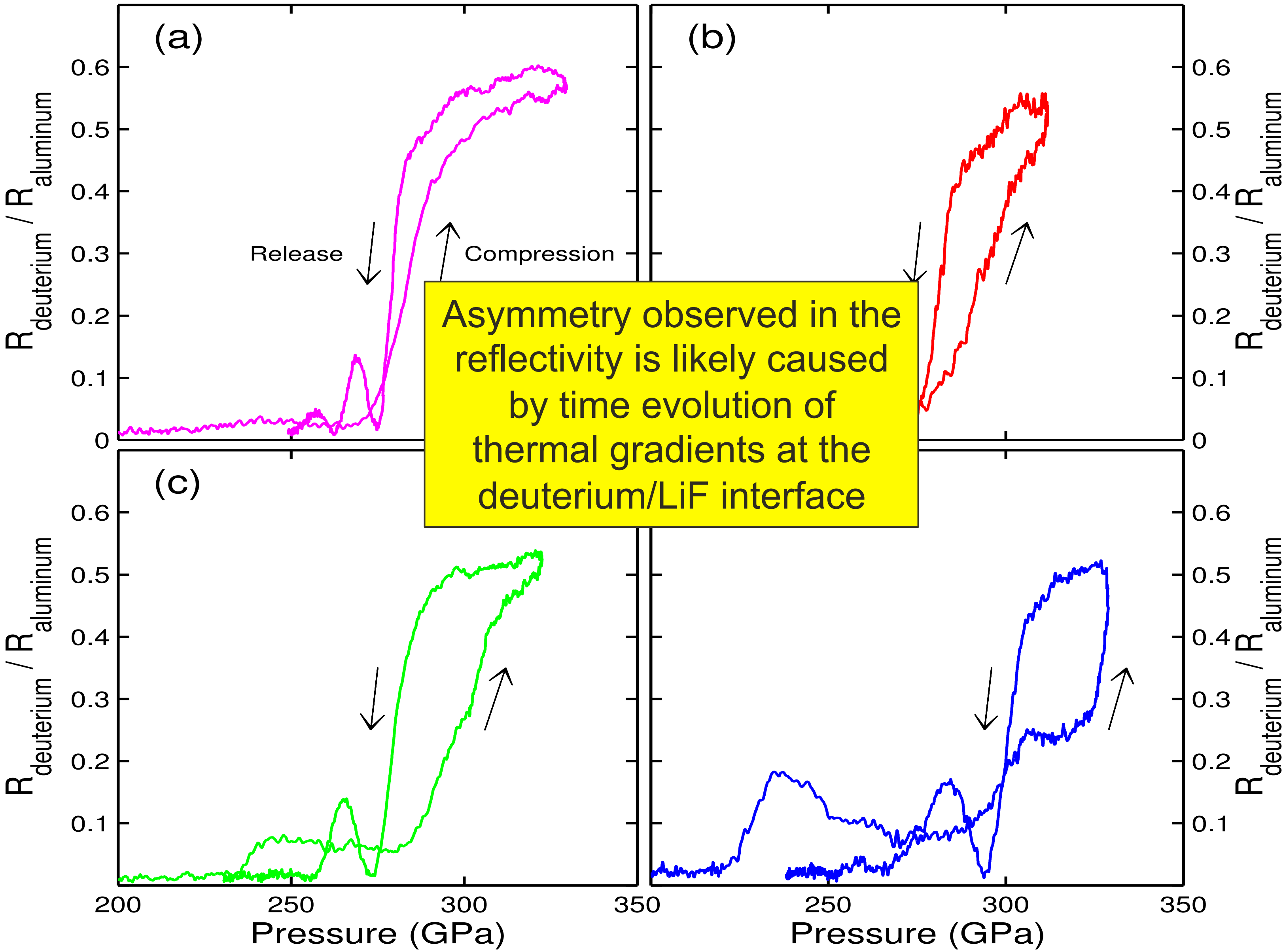
Temperature and density from Kerley03 EOS



Experimental PT Paths

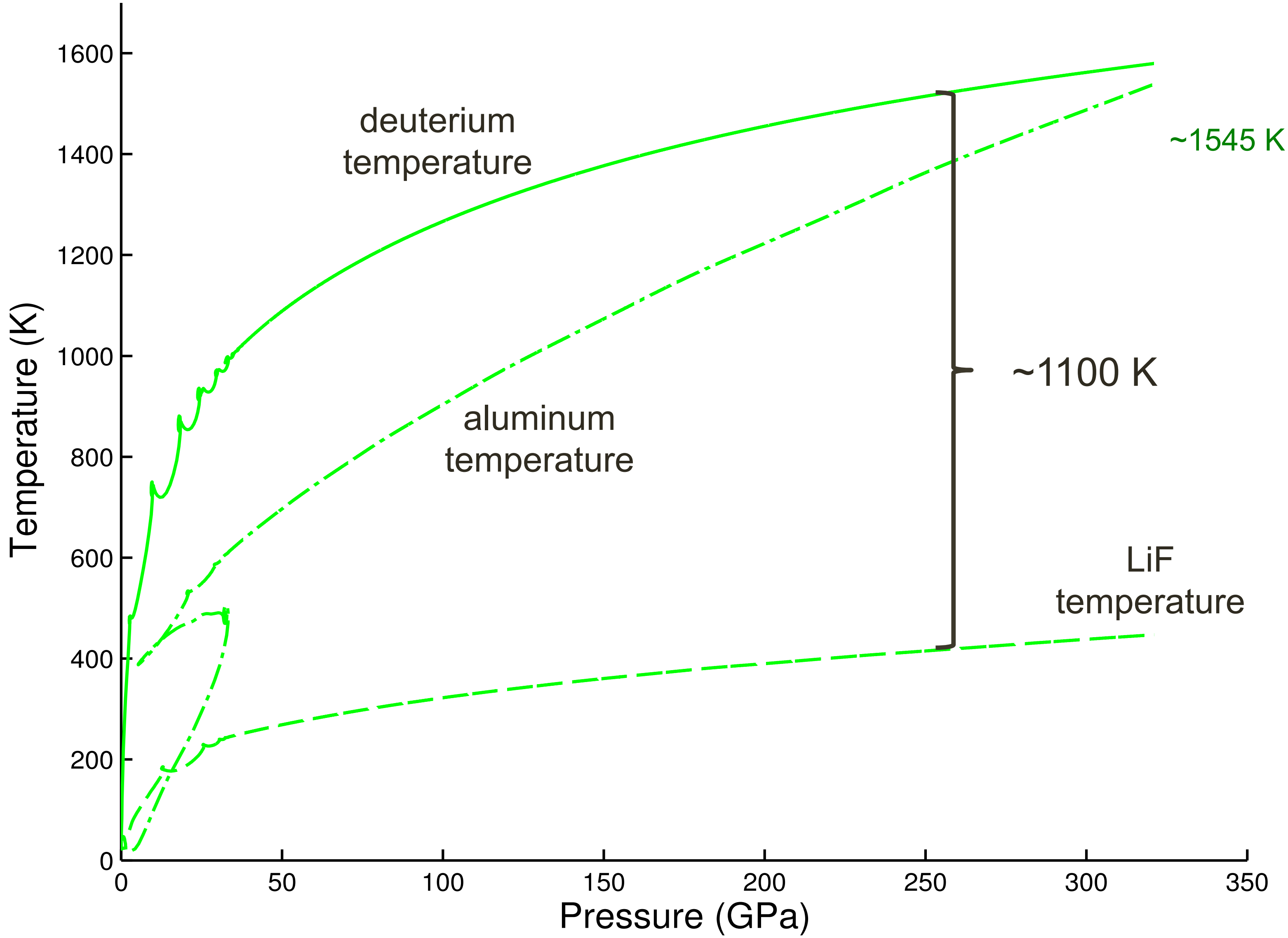


Reflectivity signals mapped to pressure

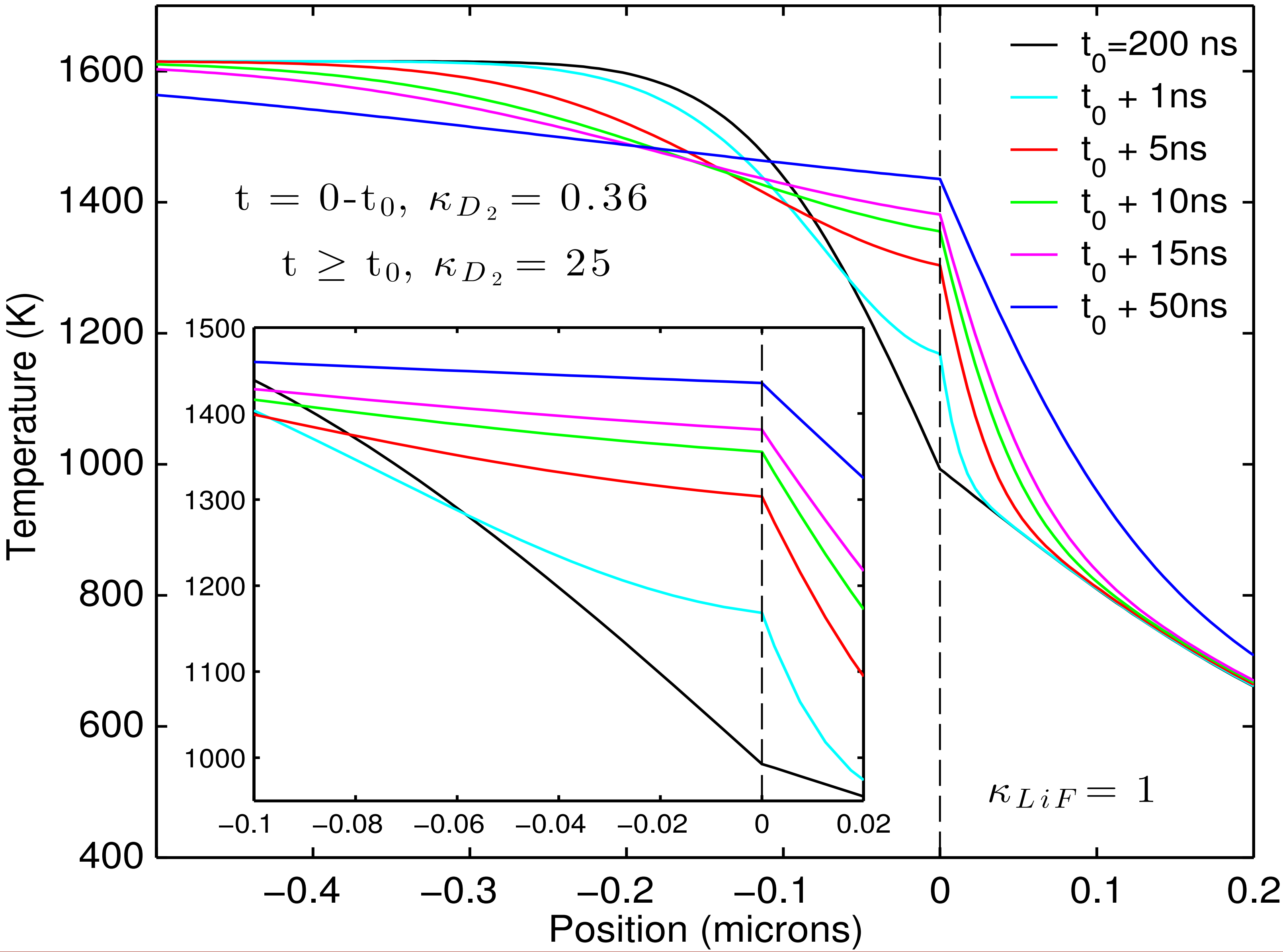


Asymmetry observed in the reflectivity is likely caused by time evolution of thermal gradients at the deuterium/LiF interface

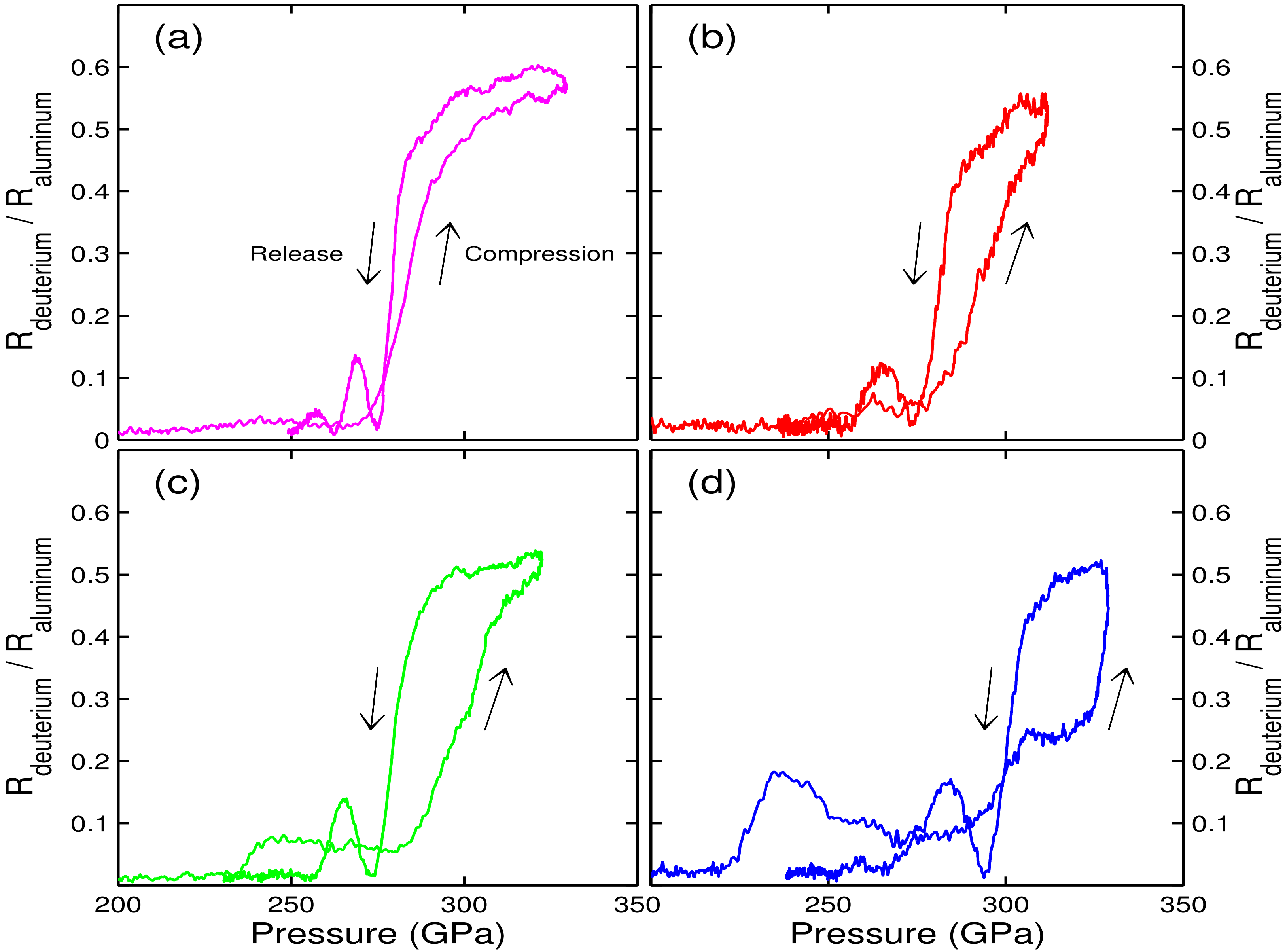
There is a significant temperature difference at the deuterium/LiF interface



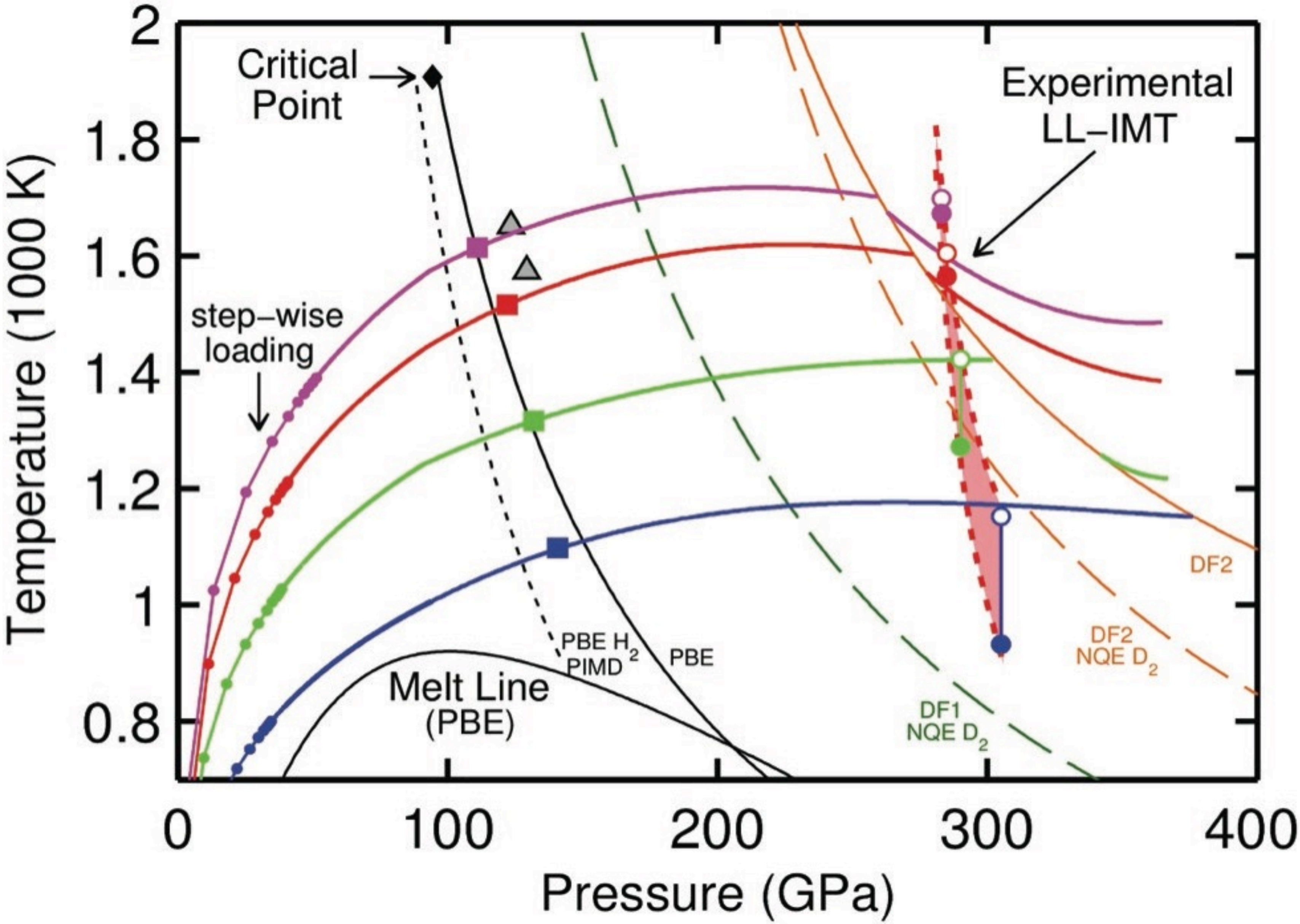
Thermal conduction simulations



Reflectivity signals mapped to pressure



Location of the LL-IMT in deuterium



HIGH-PRESSURE PHYSICS

Direct observation of an abrupt insulator-to-metal transition in dense liquid deuterium

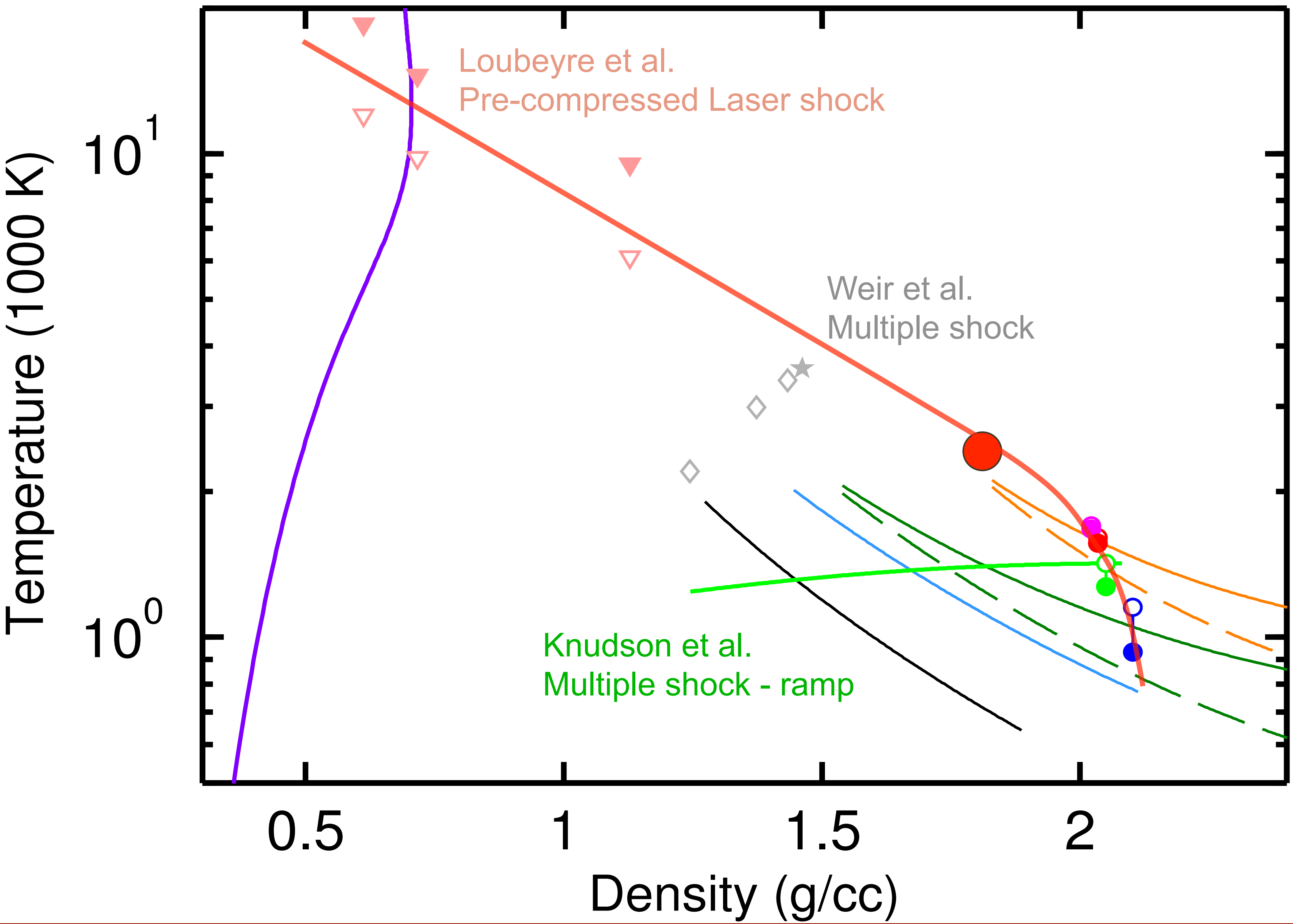
M. D. Knudson,[1*] M. P. Desjarlais,[1] A. Becker,[2] R. W. Lemke,[1] K. R. Cochrane,[1] M. E. Savage,[1] D. E. Bliss,[1] T. R. Mattsson,[1] R. Redmer[2]

Eighty years ago, it was proposed that solid hydrogen would become metallic at sufficiently high density. Despite numerous investigations, this transition has not yet been experimentally observed. More recently, there has been much interest in the analog of this predicted metallic transition in the dense liquid, due to its relevance to planetary science. Here, we show direct observation of an abrupt insulator-to-metal transition in dense liquid deuterium. Experimental determination of the location of this transition provides a much-needed benchmark for theory and may constrain the region of hydrogen-helium immiscibility and the boundary-layer pressure in standard models of the internal structure of gas-giant planets.

In 1935, Wigner and Huntington (*1*) were the first to predict that when squeezed to sufficiently high density (ρ) and pressure (*P*), hydrogen would undergo a density-driven transition from an insulating, molecular solid to a conducting, atomic solid. Subsequently, this fundamental question of precisely how and at what *P* hydrogen metallizes at low temperature (*T*) has become one of the longest-standing open questions of high-pressure physics (*2*). More recently, there has been much interest in the analogous molecular insulator to atomic metal transition in the liquid at low *T* just above the melt line, largely due to its relevance to planetary science (*3*, *4*). A metallization transition in this region could provide a constraint for the

[1]Sandia National Laboratories, Albuquerque, NM, USA. [2]Institute of Physics, University of Rostock, Rostock, Germany.
*Corresponding author. E-mail: mdknuds@sandia.gov

Dynamic compression of deuterium



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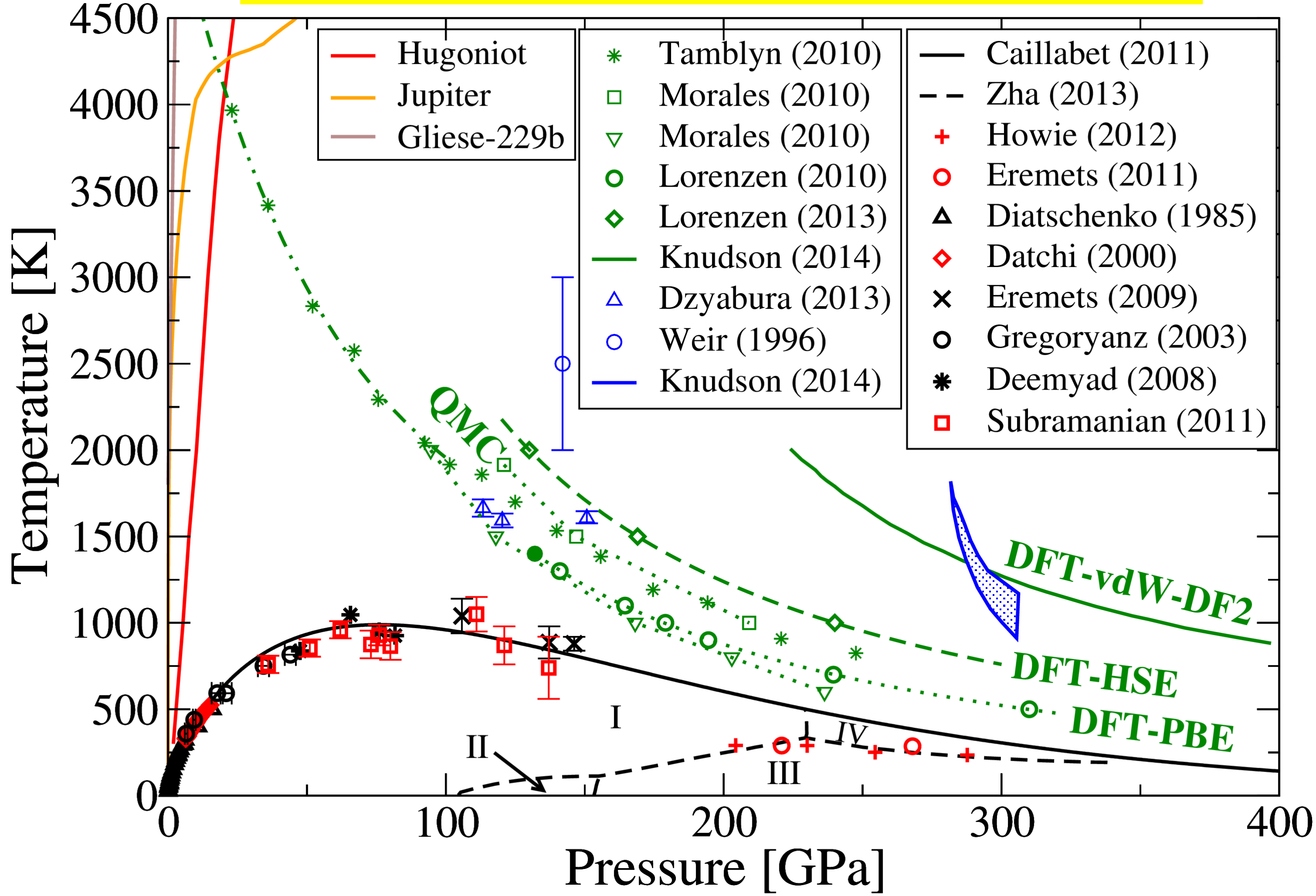
New H phase diagram at high pressure

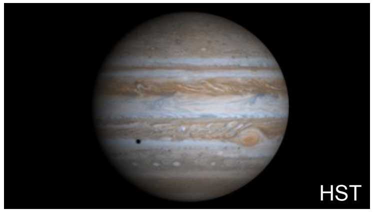
Location of the 1st-order liquid-liquid phase transition?
Obviously not relevant for the layer boundary, or ?

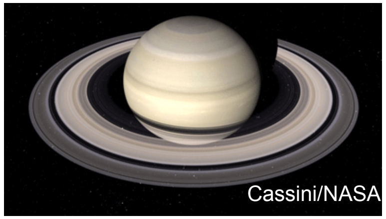


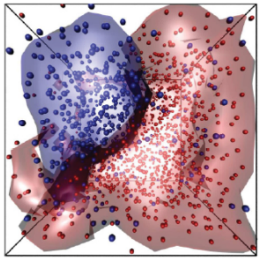
Demixing of H/He and evolution of Jupiter and Saturn

R. Püstow & N. Nettelmann (U Rostock),

J.J. Fortney (UCSC), **S. Hamel** (LLNL), **W. Lorenzen**







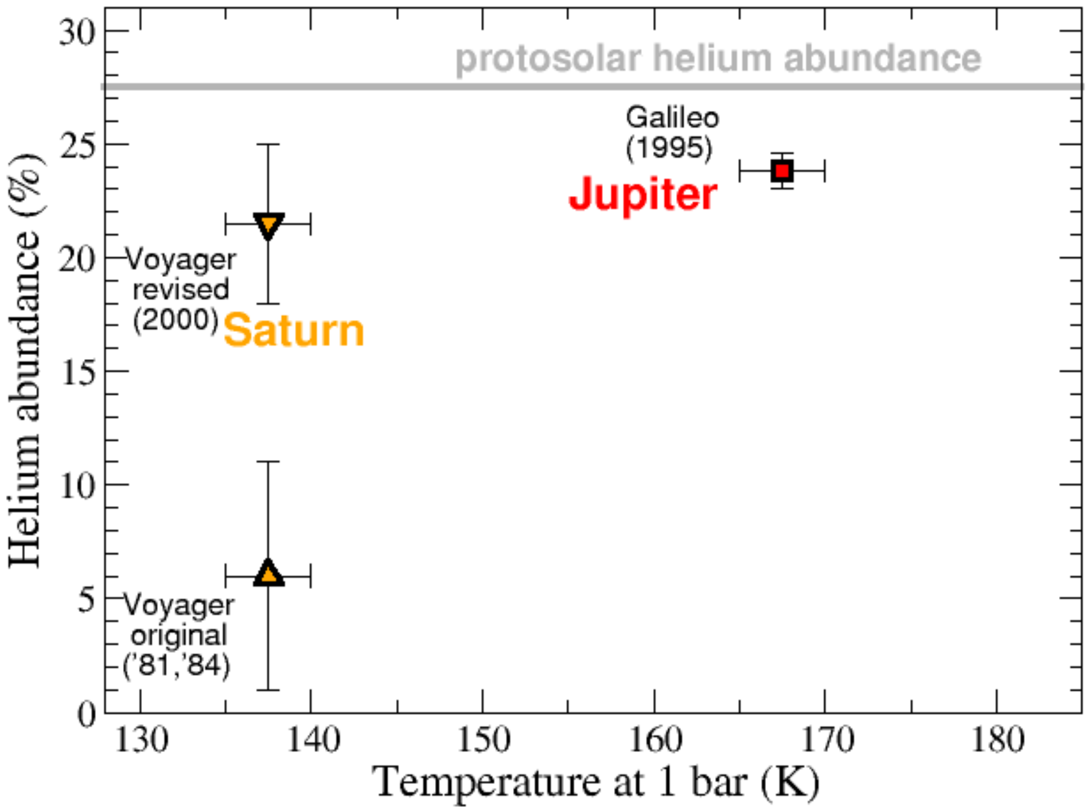
	Jupiter	Saturn	
Mass	**318 M_{Earth}**	**95 M_{Earth}**	**P > ~ 1 Mbar**
Radius	**11 R_{Earth}**	**9 R_{Earth}**	**T < ~ 10,000 K**
Luminosity	**1.7 x L_{inc}**	**1.9 x L_{inc}**	**x_{He} < 0.95**
T at 1-bar	**170 K**	**140 K**	
T, P at center	**~20,000 K, ~70 Mbar**	**~10,000 K, ~40 Mbar**	
H/He (M_{planet})	**~0.9**	**~0.8**	

- **Lorenzen et al (Rostock)**
- **Morales et al (LLNL)**

By courtesy of N. Nettelmann.

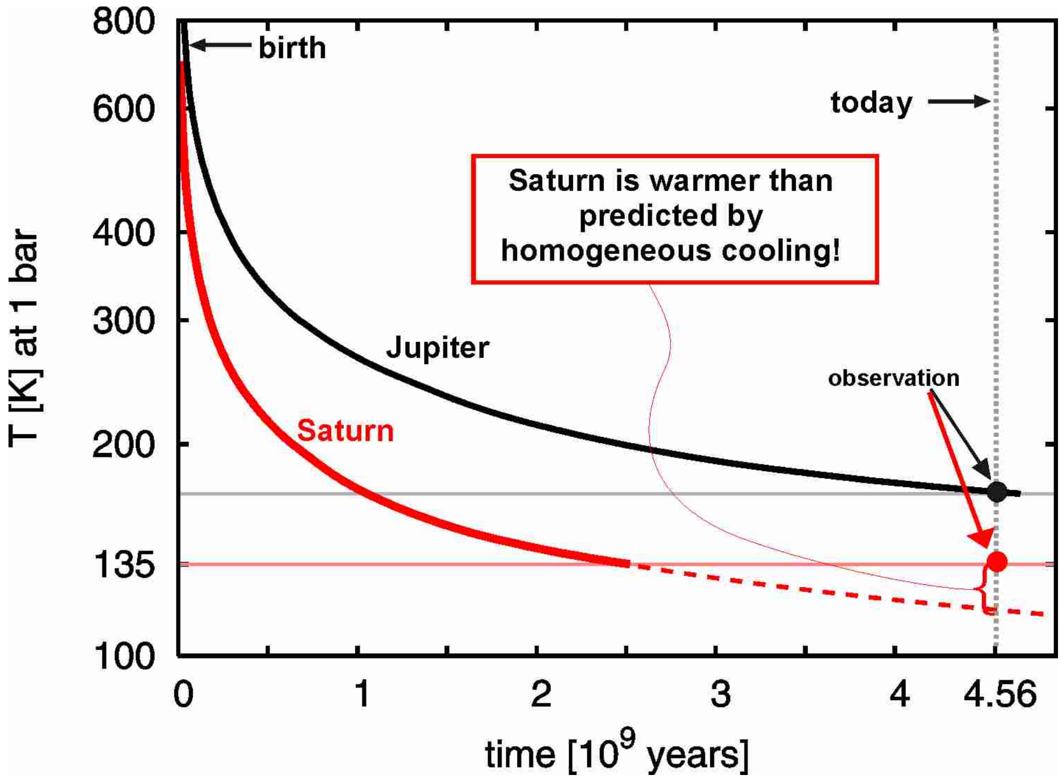
Jupiter & Saturn: atmospheric helium depletion

The atmospheres are observed to be depleted in helium compared to the protosolar value (y_{He}=0.275).



By courtesy of N. Nettelmann.

Saturn: excess luminosity

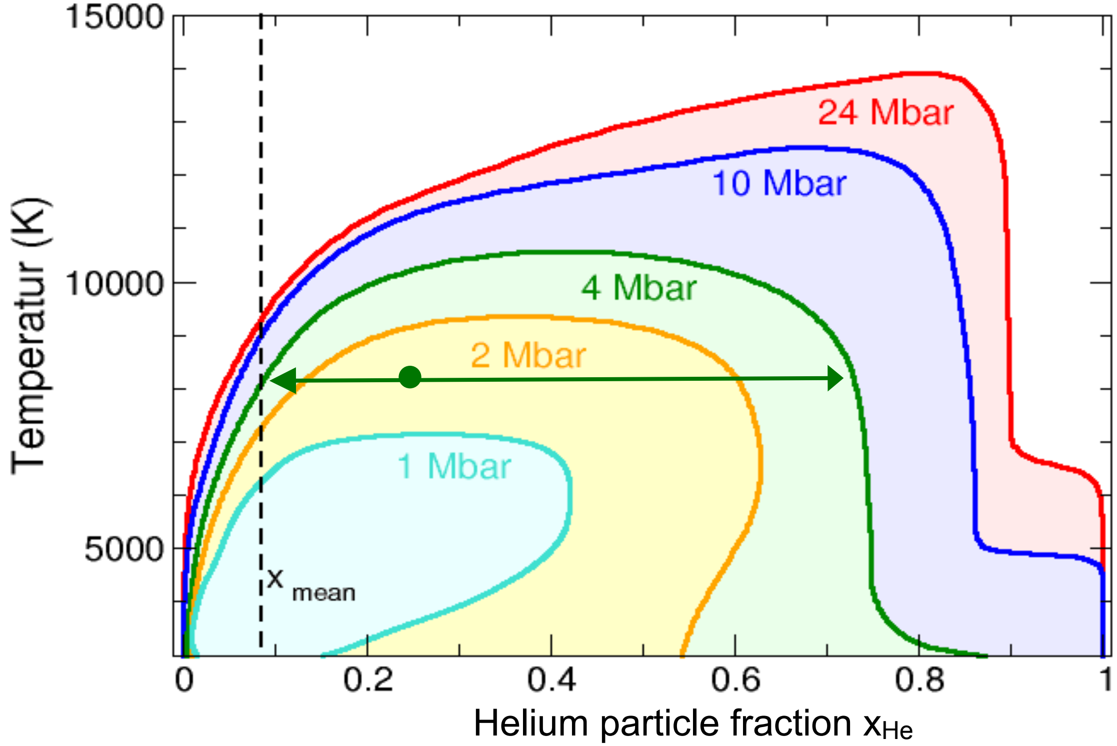


Jupiter: standard models reproduce all observational constraints

Saturn: standard models predict too low luminosity – why?

H/He phase diagram: T (x_{He}), P={1,2,4,10,24} Mbar

A demixing H-He system will separate into two phases on the phase boundary.

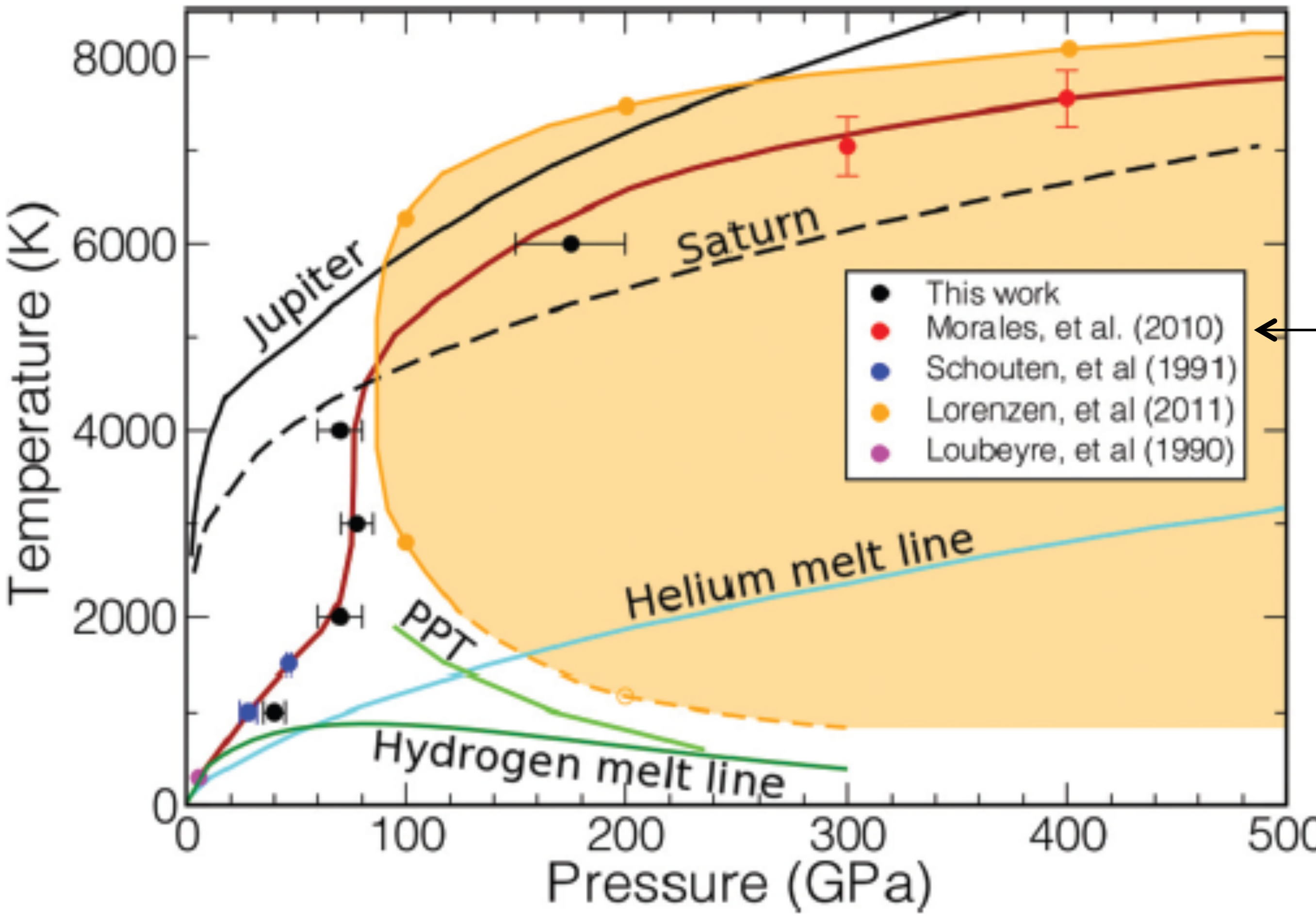


Lorenzen, Holst, Redmer (2009, 2011) via DFT-MD simulations using the PBE XC functional – imapct of non-ideal entropy of mixing, NQE, nonlocal XC functionals, vdW corrections?

By courtesy of N. Nettelmann.

H-He demixing in gas giants

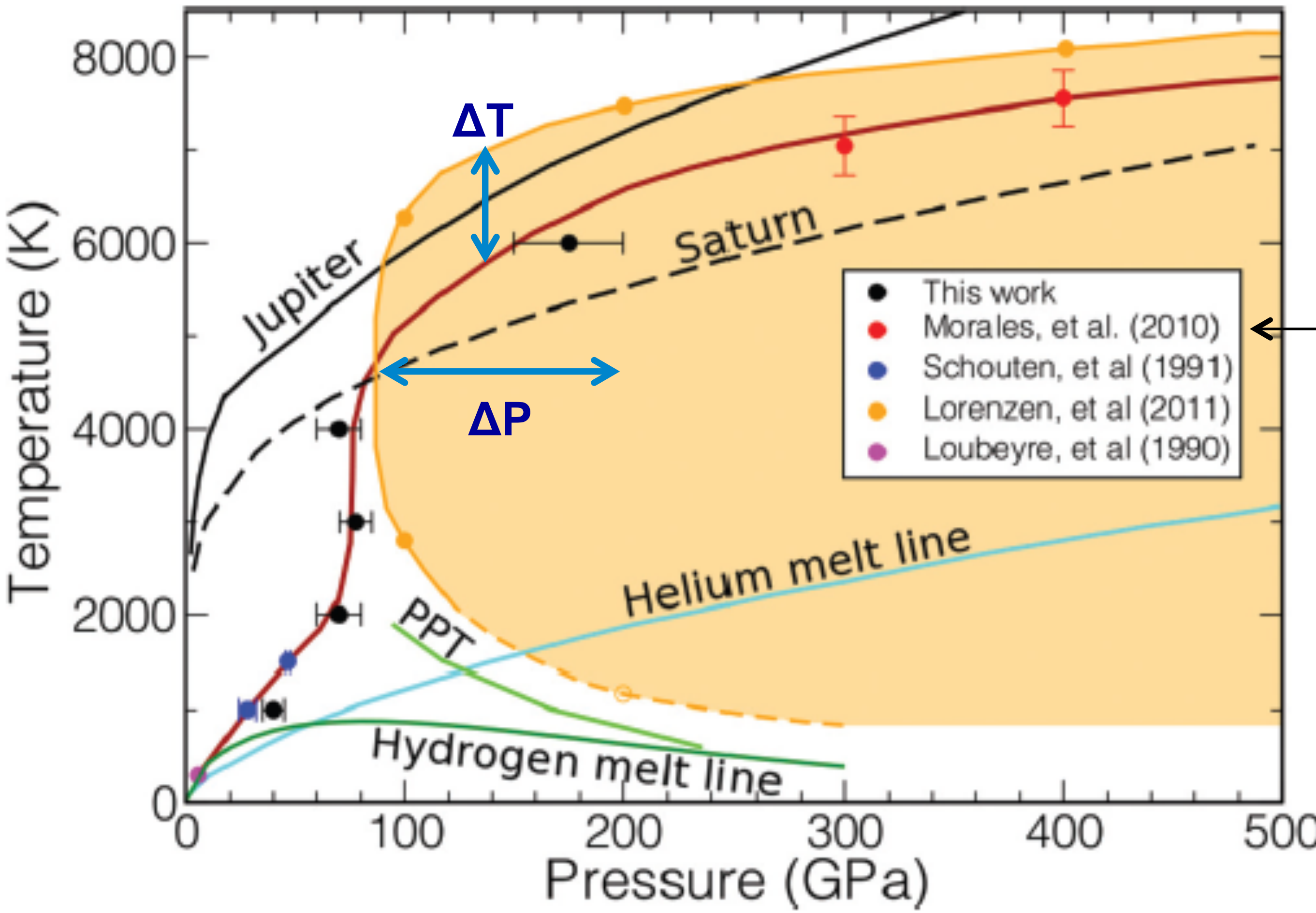
Metallic hydrogen acts as a "catalyst" for H/He demixing.
Effect maybe relevant for Jupiter but clearly is for Saturn.



W. Lorenzen et al., PRL **102**, 115701 (2009), PRB **84**, 235109 (2011)
M.A. Morales et al., PNAS **106**, 1324 (2009) , PRB **87**, 174105 (2013).

H-He demixing in gas giants

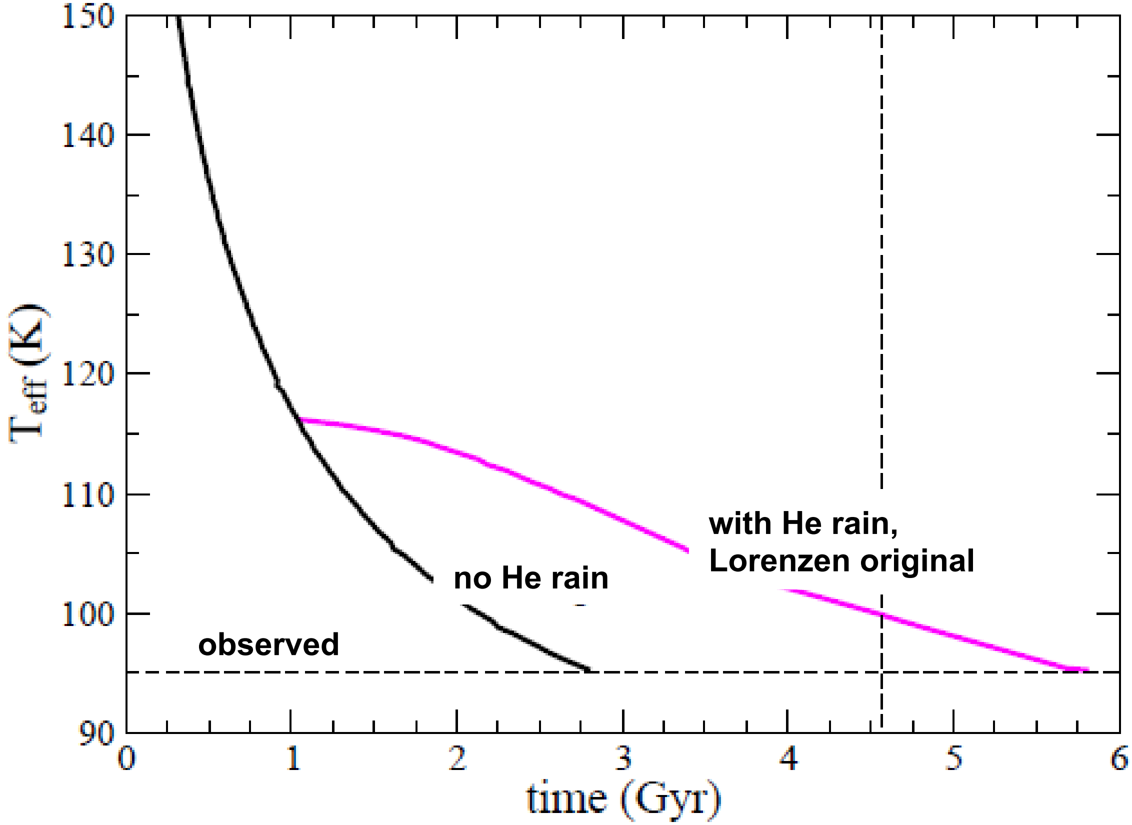
Correct location of demixing region?
Study shifts in P and T!

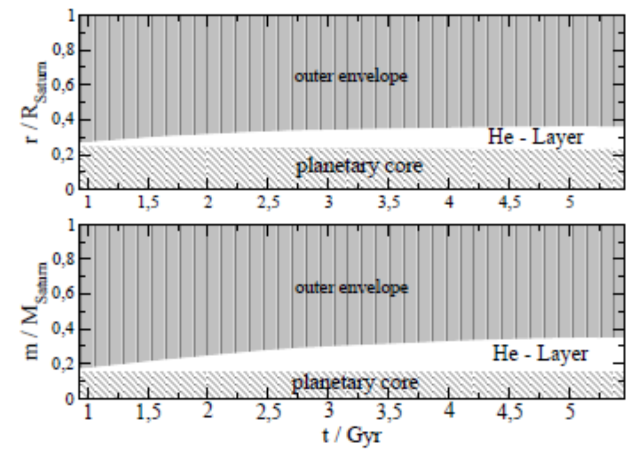


W. Lorenzen et al., PRL **102**, 115701 (2009), PRB **84**, 235109 (2011)
M.A. Morales et al., PNAS **106**, 1324 (2009) , PRB **87**, 174105 (2013).

Results for Saturn: new cooling curve

Application of the original Lorenzen (2009, 2011) phase diagram leads to over-estimation of Saturn‘s excess luminosity.

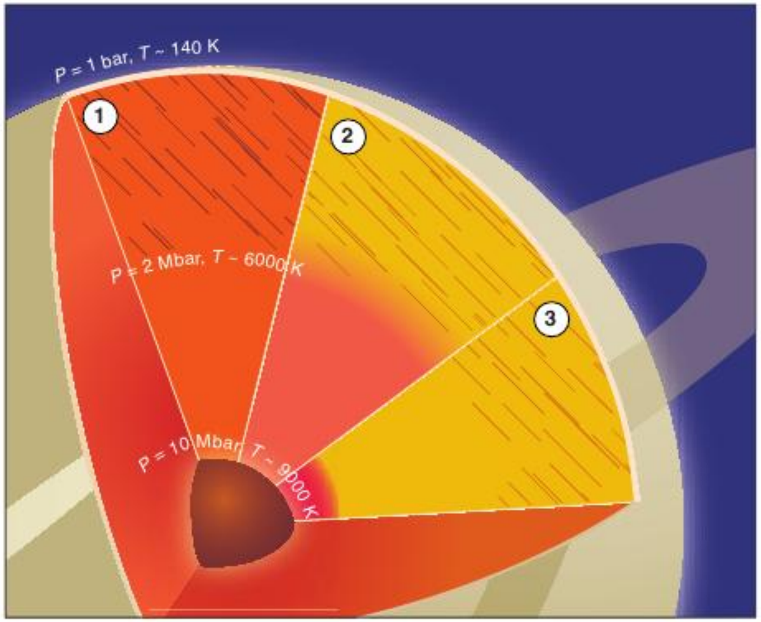




- **Püstow, Nettelmann, Redmer, submitted**
- **Lorenzen, Holst, Redmer (2009, 2011)**

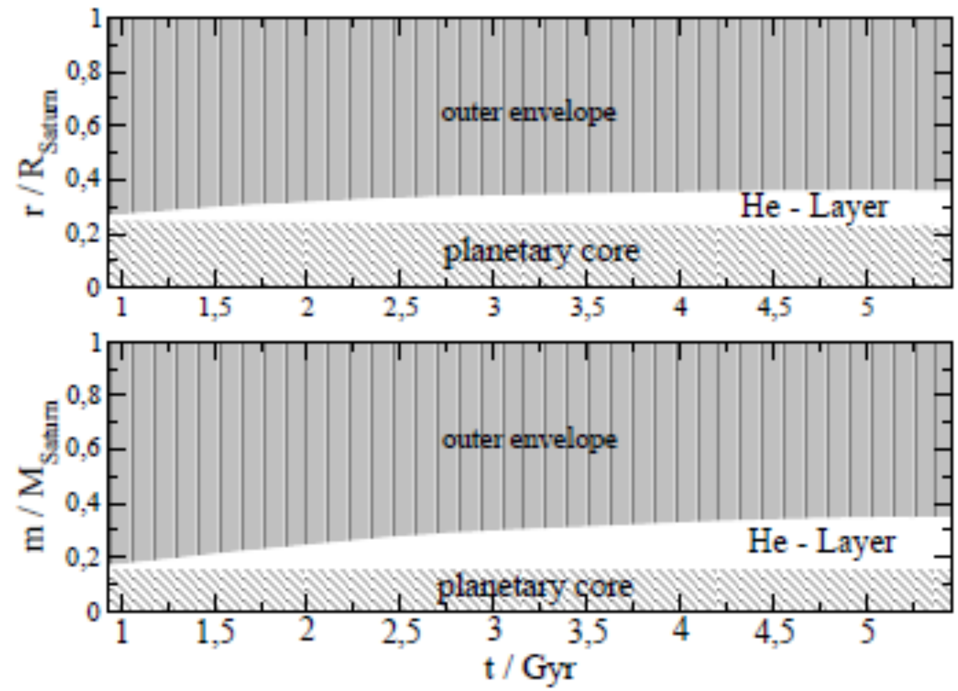
Evolution and structure of Saturn: He layer

J.J. Fortney, Science **305**, 1414 (2004)

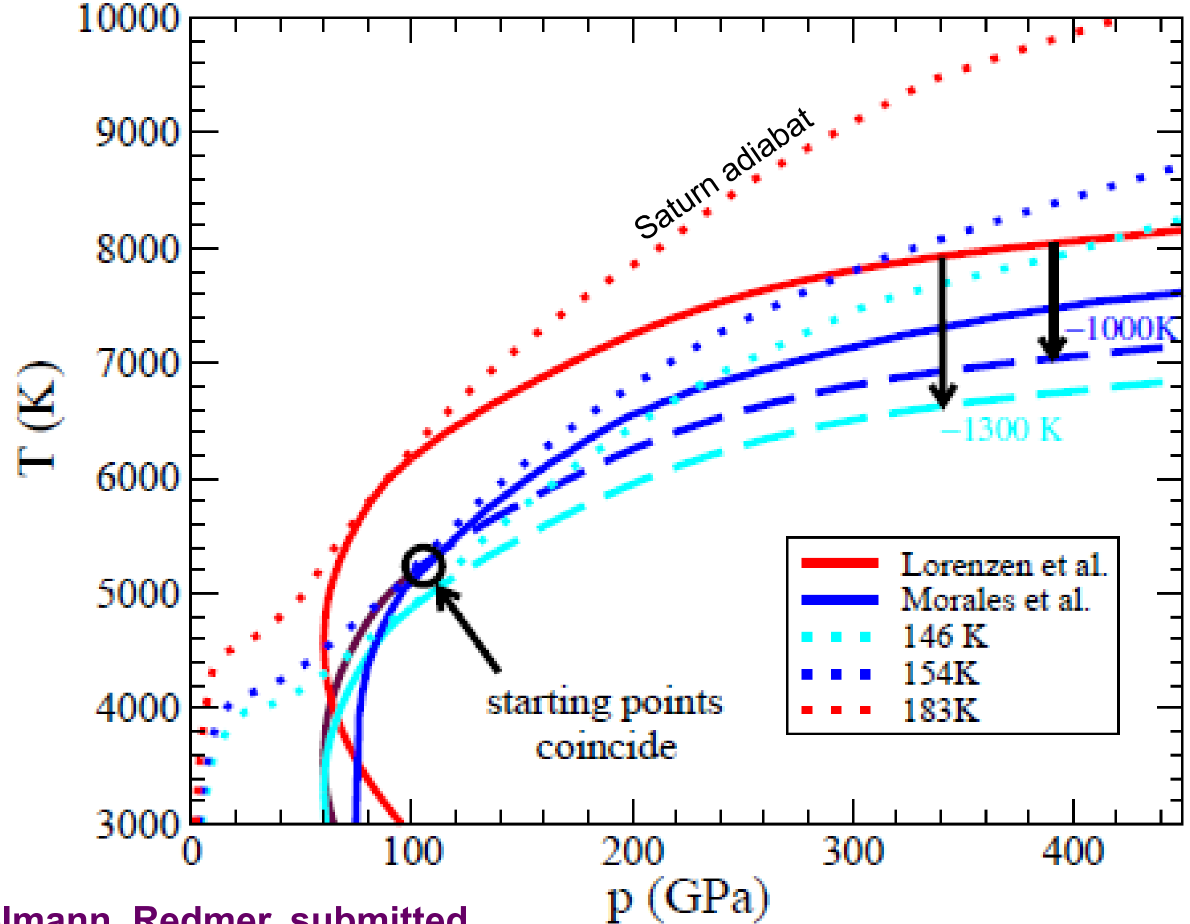


Three views of the interior of Saturn. Orange represents the protosolar He/H ratio. A yellower orange indicates less He, and a redder orange more He. Brown is the ice/rock core. The hashed regions indicate that H is liquid molecular, whereas in the unhashed regions it is liquid metallic. (**1**) Saturn at an age of ~1.5 billion years, before the onset of He phase separation. (**2**) The current Saturn according to a previously proposed H-He phase diagram (*11*). (**3**) The current Saturn according to a phase diagram derived from new evolutionary models (*2*).

R. Püstow et al. (2015)



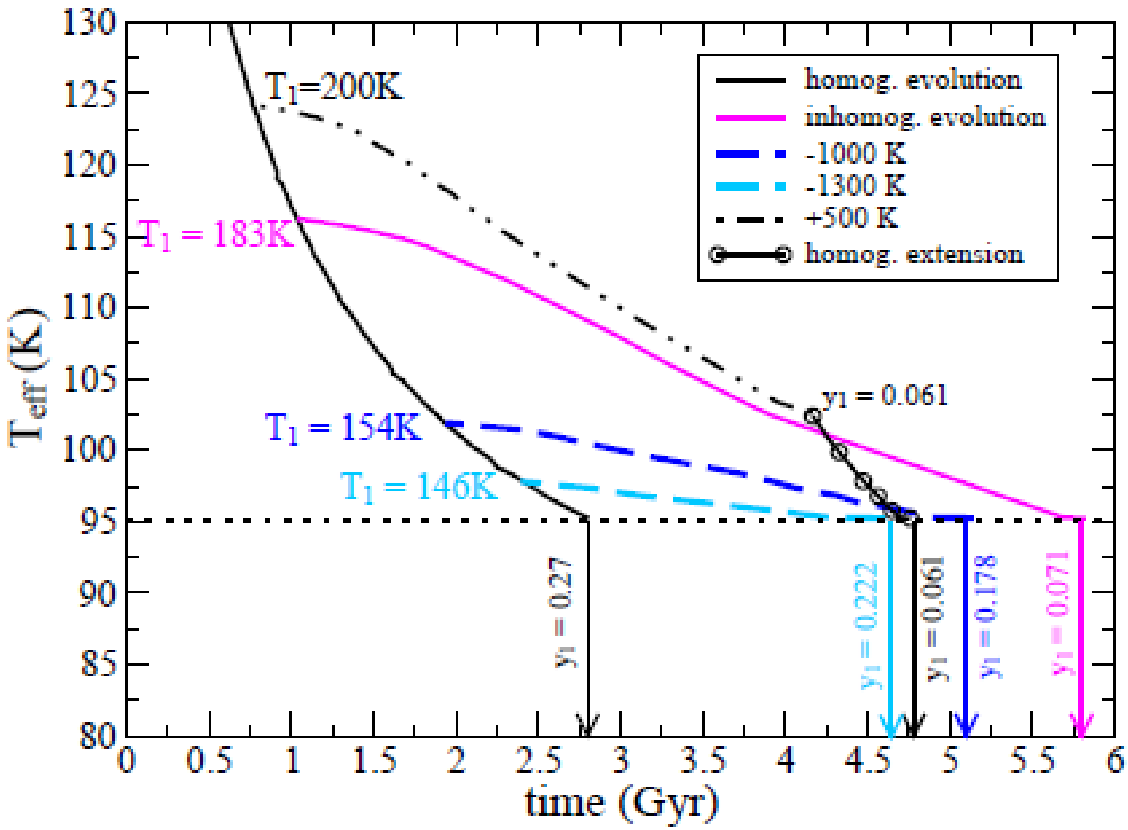
Modifications of the Lorenzen H/He phase diagram ($\Delta T = -1000$ K , -1300 K, +500 K)

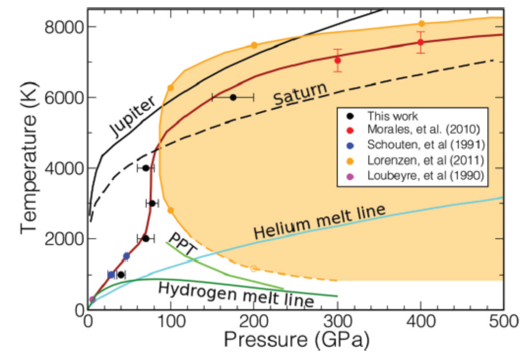


- Püstow, Nettelmann, Redmer, submitted
- Morales et al (2009, 2013)
- Lorenzen, Holst, Redmer (2009, 2011)

Saturn cooling curves for modified Lorenzen H/He phase diagrams

Lorenzen H-He EOS with demixing available for all He concentrations.
Shifts by $\Delta T = -1300$ K and $\Delta T = +500$ K yield the correct cooling time.

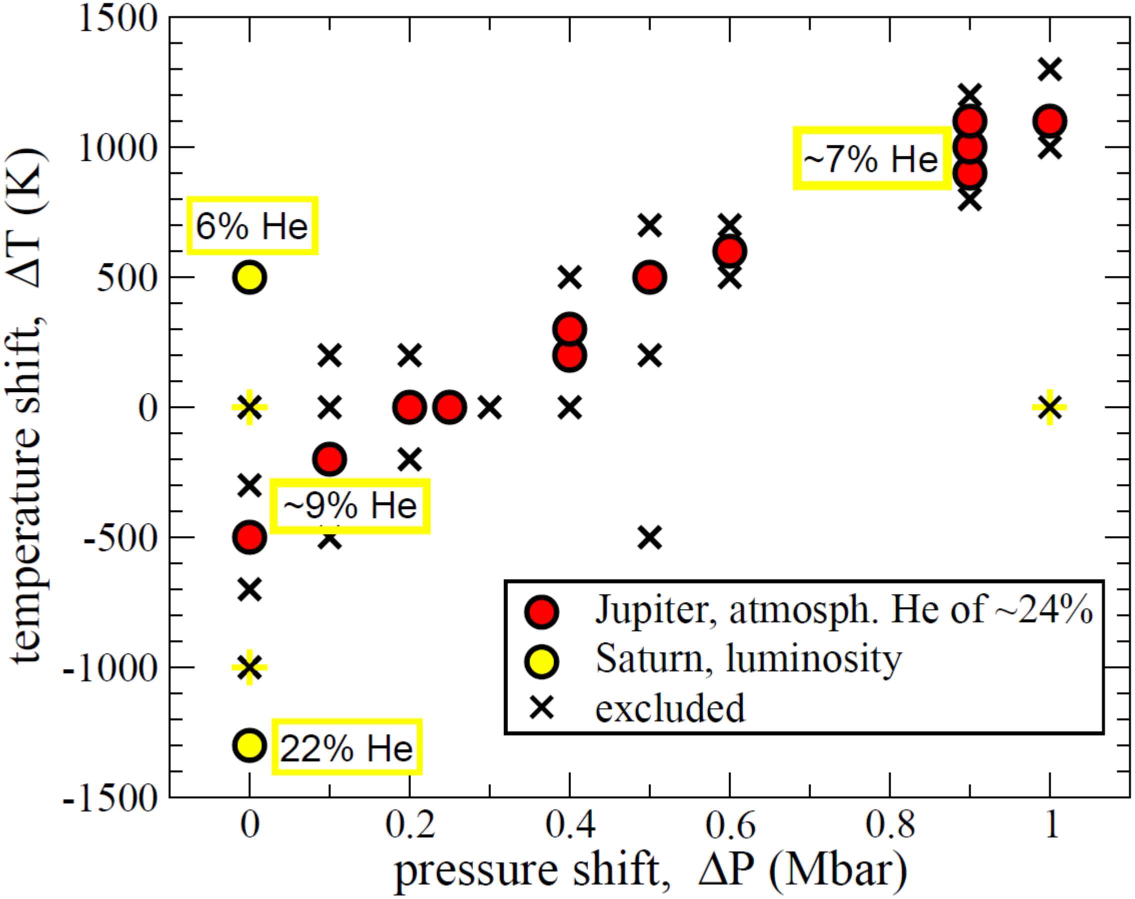


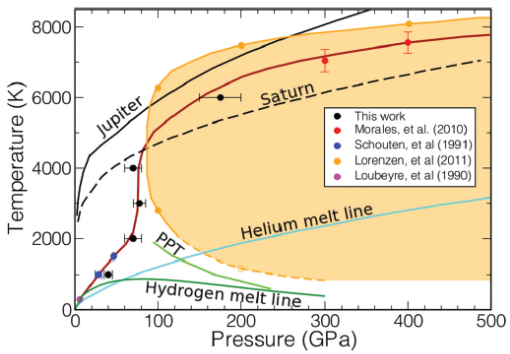


- Püstow, Nettelmann, Redmer, submitted

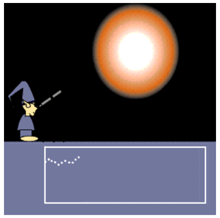
Which H/He phase diagrams reproduce observations for Jupiter and Saturn?

We use Jupiter's measured atmosph. He abundance to predict corrections to the H/He phase diagram



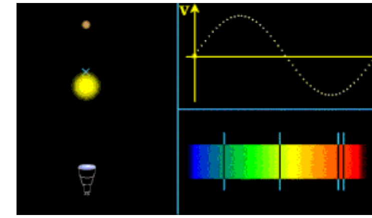


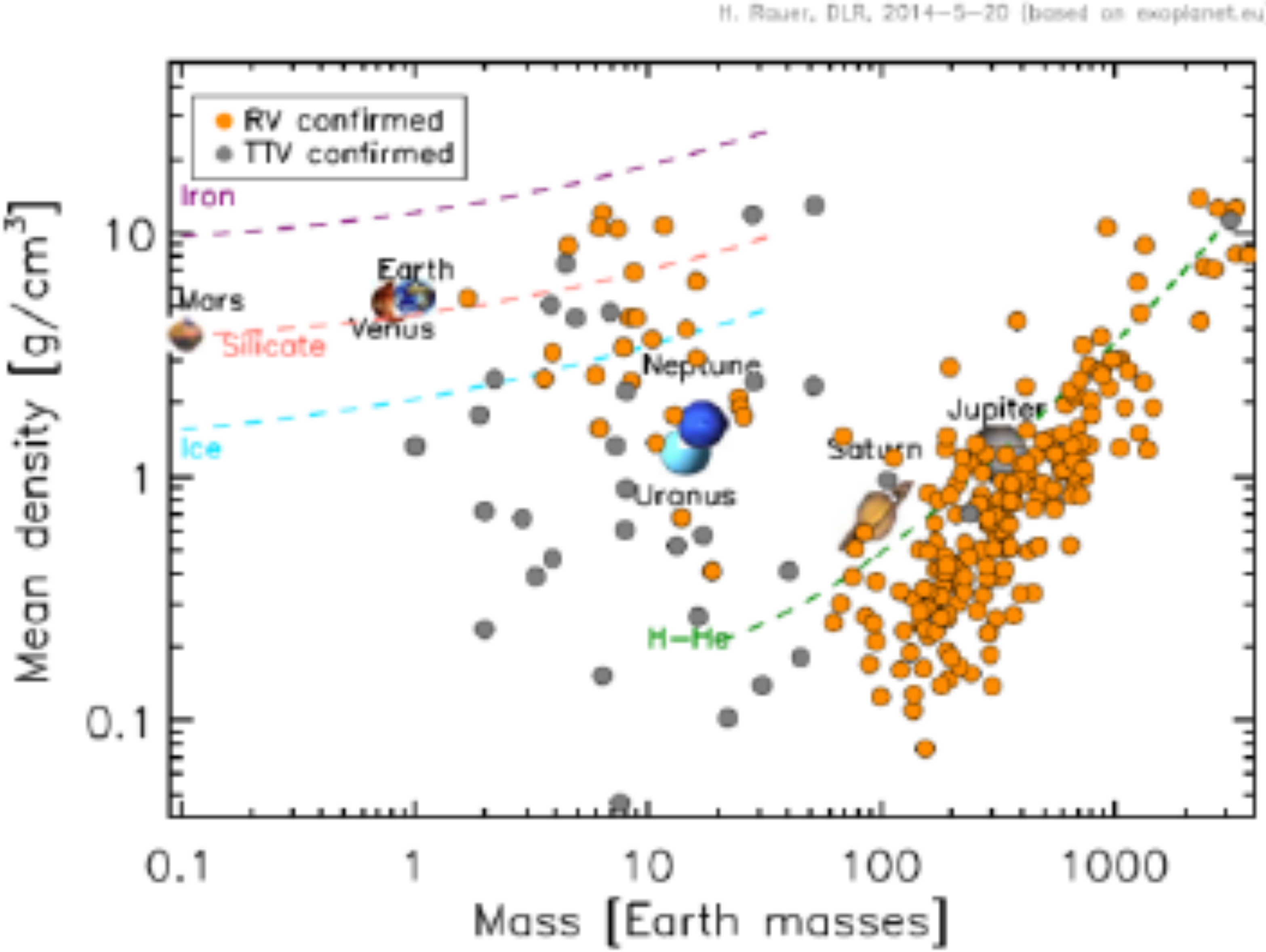
ZFSP experiments are an important step towards a coherent understanding of interior structure, evolution and magnetic field of the gas giants.

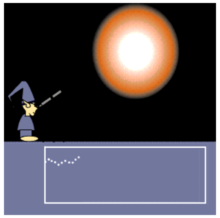


Mean planet density vs. mass

H. Rauer et al., Exp. Astron. **38**, 249 (2014)

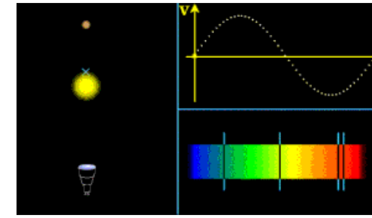




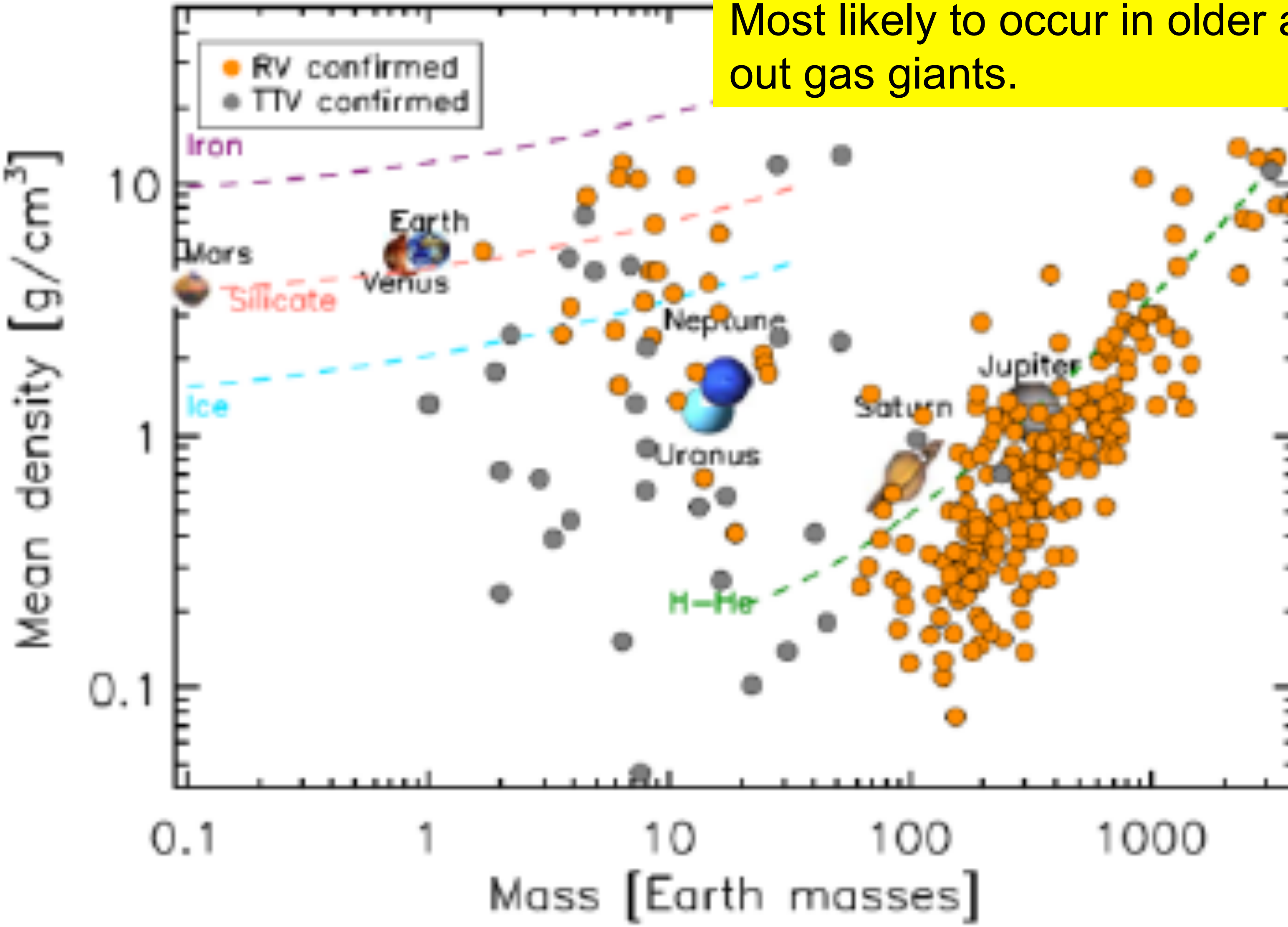


Mean planet density vs. mass

H. Rauer et al., Exp. Astron. **38**, 249 (2014)



H/He demixing in Saturn-like planets? Most likely to occur in older and far-out gas giants.



Conclusions

- Shock-ramp technique enables experimental access to the region of phase space where the liquid-liquid, insulator-metal transition (LL-IMT) has been proposed for hydrogen
- Experiments above ~250 GPa show clear evidence of metallization of deuterium
  - Very abrupt increase in reflectivity to ~40-50%
  - Pressure state well above numerous first principles predictions
  - Indications suggest that the density-driven transition is first order
- Impact on planetary physics due to a first constraint for the
  - Region of metallic (conductive) hydrogen → magnetic field, interior
  - Region of H/He demixing → evolution, interior

Acknowledgements

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Devon Dalton

Anthony Romero

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Alan Carlson

QMD Calculations

Mike Desjarlais

Andreas Becker

Winfried Lorenzen

Ronald Redmer

Planetary Modeling

Nadine Nettelmann

Andreas Becker

Robert Püstow

Ronald Redmer

Pulse Shaping

Ray Lemke

Jean-Paul Davis

Mark Savage

Ken Struve

Keith LeChien

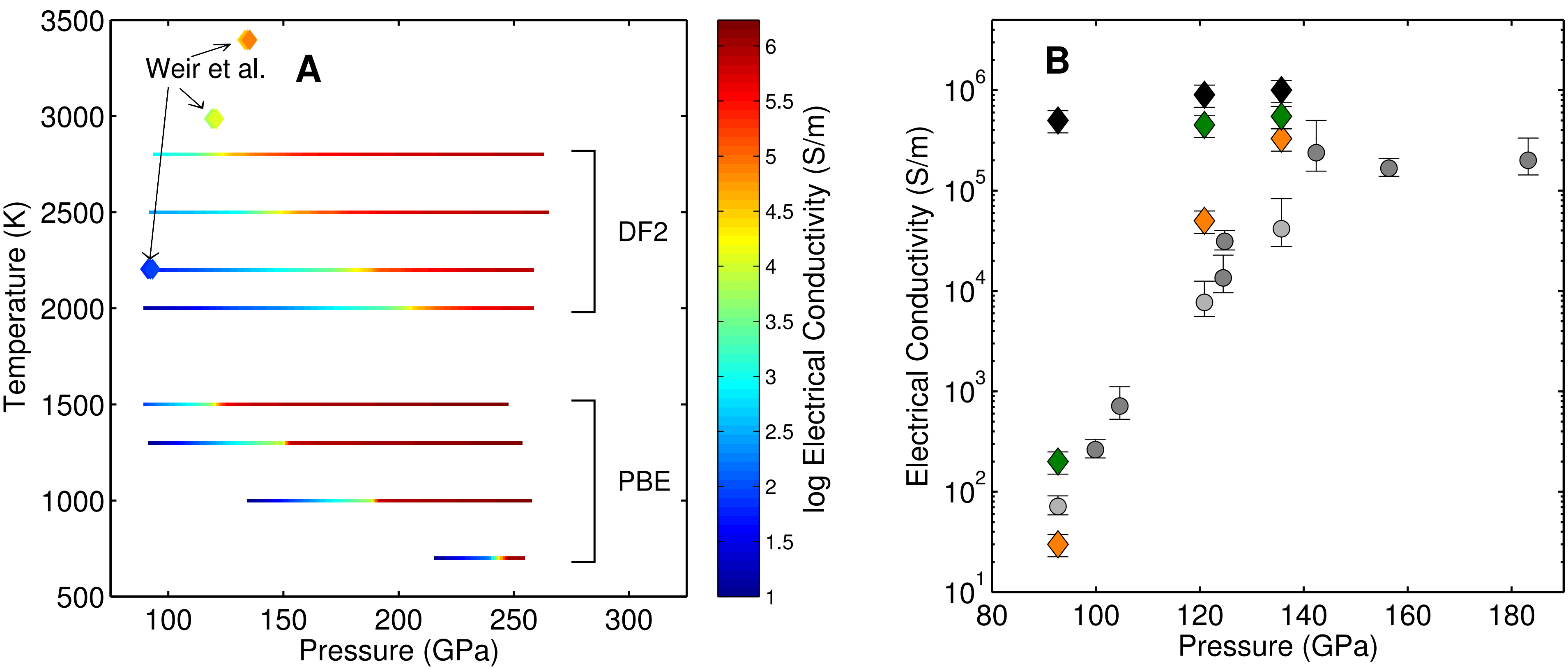
Brian Stoltzfus

Dave Hinshelwood

Entire Z crew

University of Rostock

Comparison of Weir et al. data with DFT



Reanalysis of Weir et al. data