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Western

Electrical Resistivity of Molten Ni at High Pressures and Preliminary Results for Liquid Fe

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Motivation

- Transport properties of liquid Ni and Fe at high P have important geophysical implications for terrestrial planetary interiors (e.g. dynamo, core heat flow, core evolution)

- Ni is a close electronic analogue of Fe (similarities of ferromagnetic and paramagnetic states)

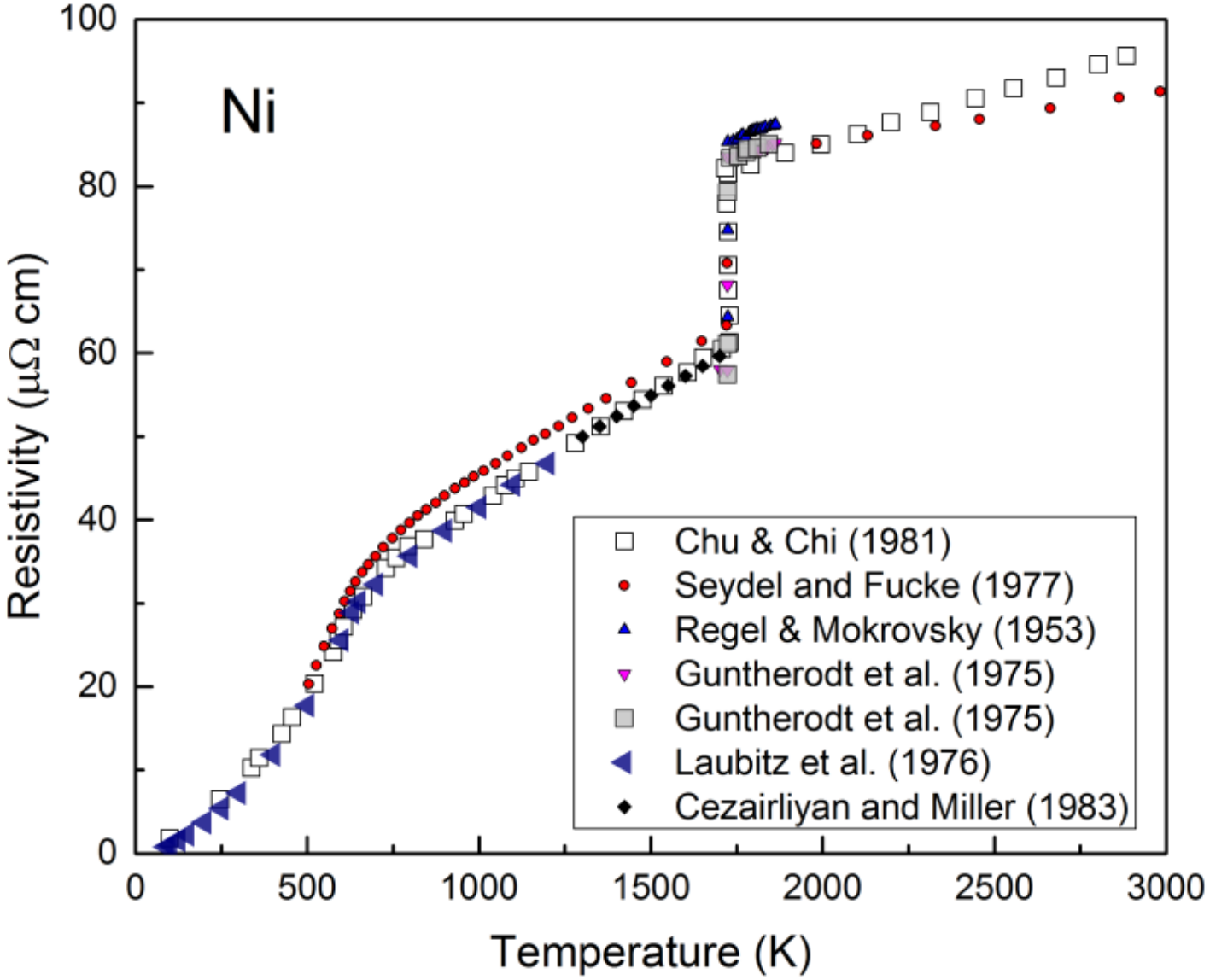
- The electrical resistivity of solids and liquids gives insight into electronic structure and properties of materials

Objective

- **The aim** of this study is to test experimentally if the **electrical resistivity of a pure Ni and Fe is constant along its P-dependent melting boundary** (Stacey and Anderson, 2001).

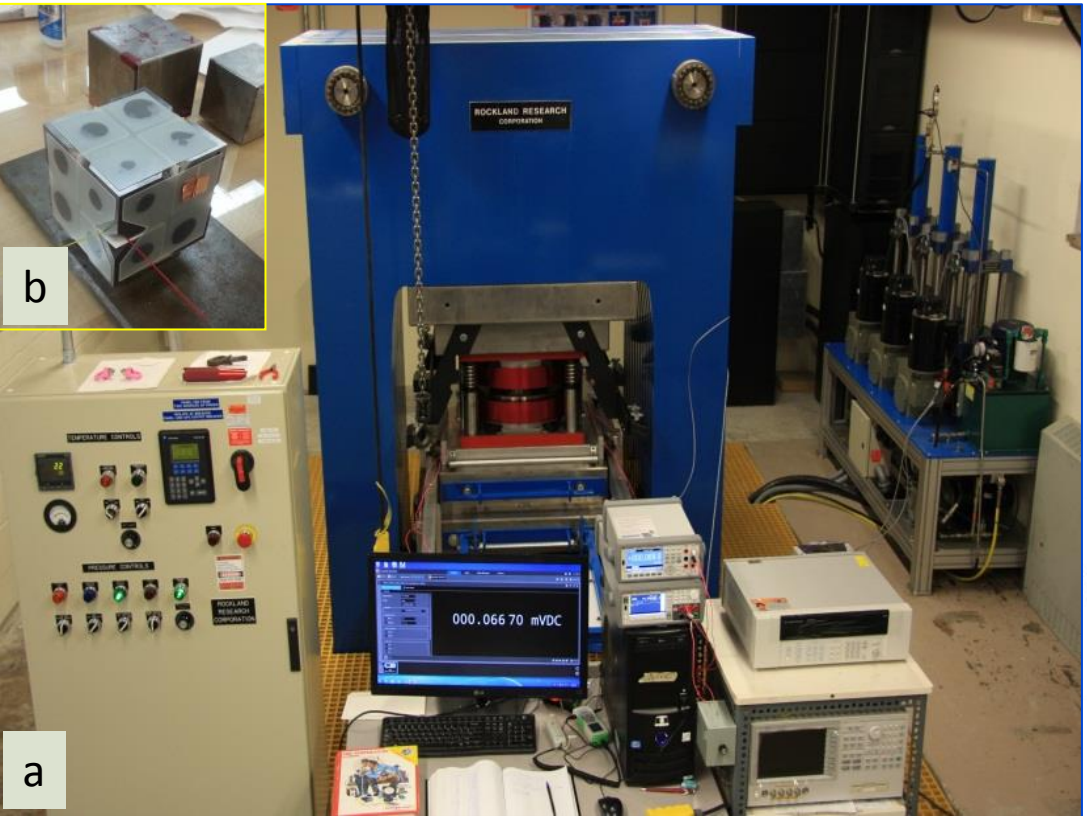
- If valid for 3d transition metals, then this behavior could serve as **a practical tool for low P studies to assess electrical resistivity of Earth's core** (Earth's inner core boundary is a melting boundary of Fe alloy).

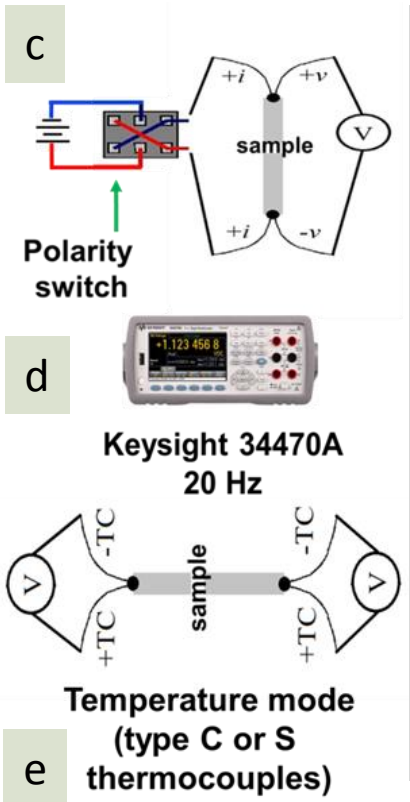
Review of Previous Ni Studies at 1atm



Methods and Experimental Challenges

- ✓ Control over the molten metal sample containment and geometry
- ✓ Reactivity with and diffusion of molten metal into its container
- ✓ Thermocouple - molten sample contamination

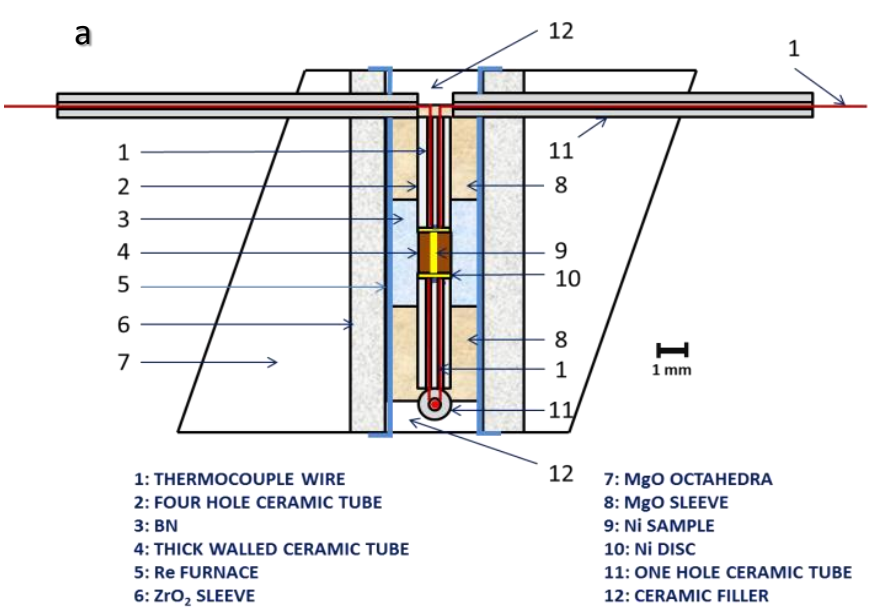


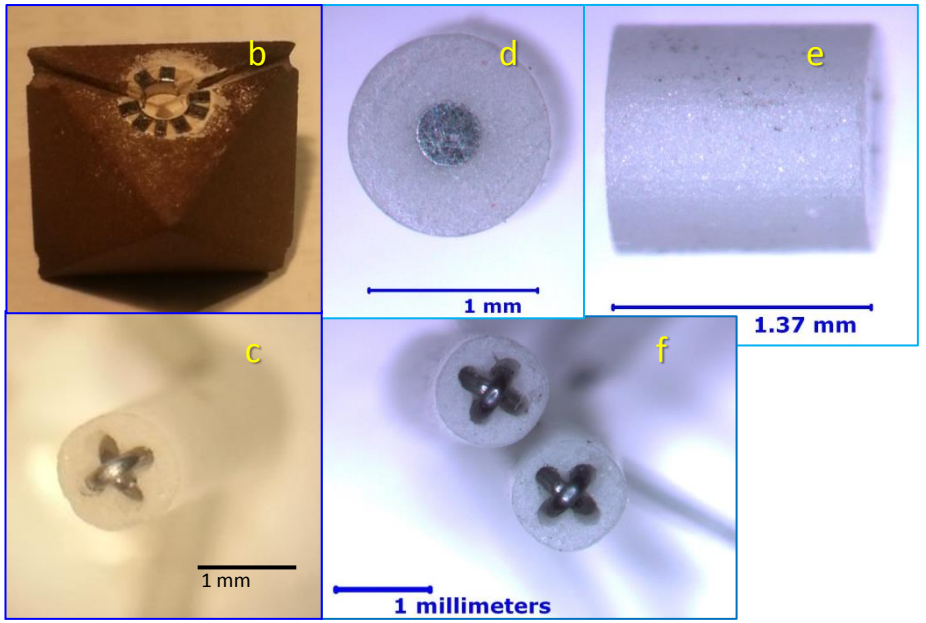


(a) Large volume 3000 ton press.
(b) Assembled WC cubes containing the sample cell.
(c) (c-e) Schematic of polarity switch (c), depicting the voltage (c) and temperature mode (e).
(d) fast acquisition meter

Methods – Cont'd

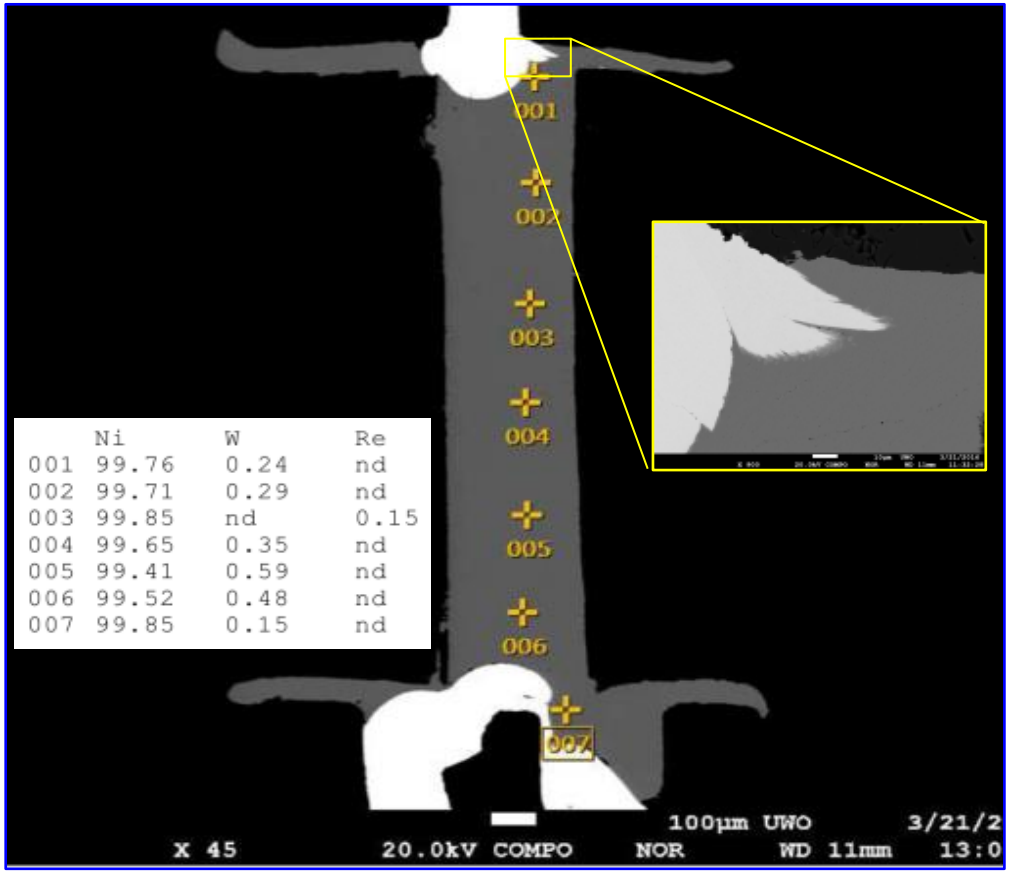
- *Everything should be made as simple as possible, but not simpler.* (Albert Einstein)



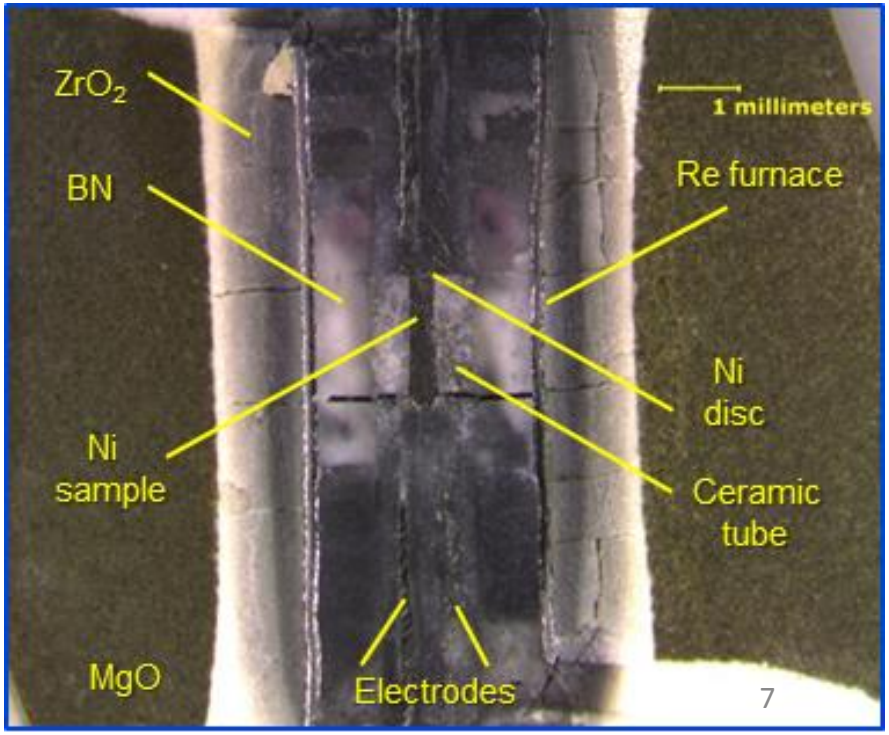


(a) Cross-section of experimental cell. (b) MgO cell. (c, f) 4-hole ceramic tube with TC emplaced wires. (d, e) Thick walled ceramic tube hosting the Ni or Fe sample.

Methods – Cont'd



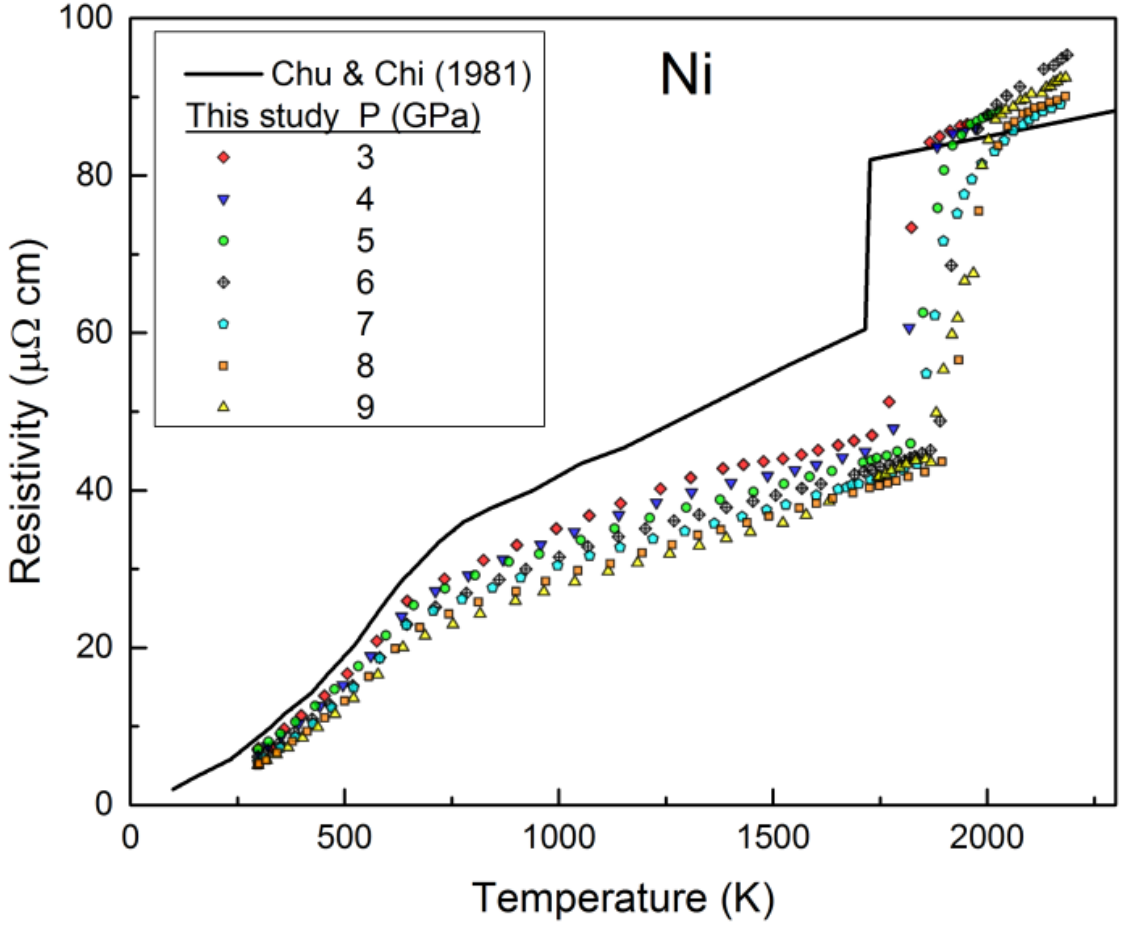
Electron microprobe image of a sample quenched at melting point. W/Re thermocouples appear white. The composition data are shown on the left. (4 GPa, 1840 K)

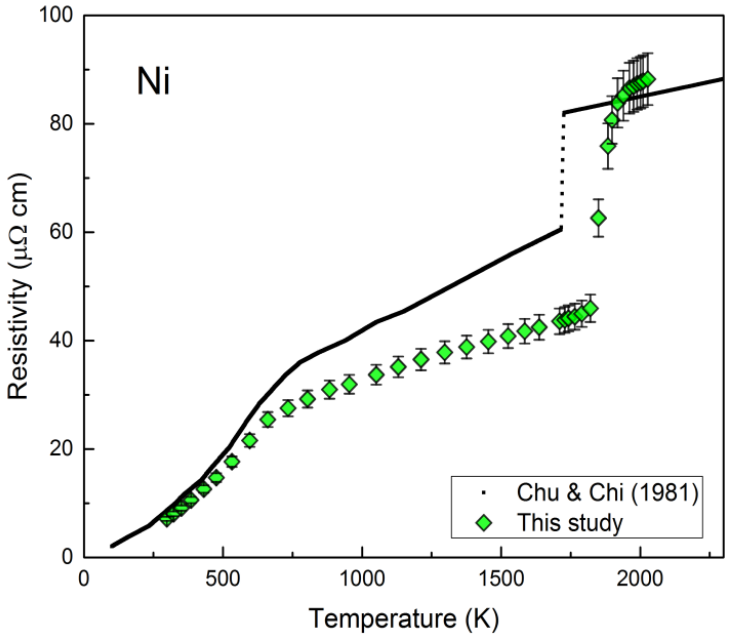


Recovered and sectioned sample. (Ni-32, 3 GPa, 1770 K)

Results

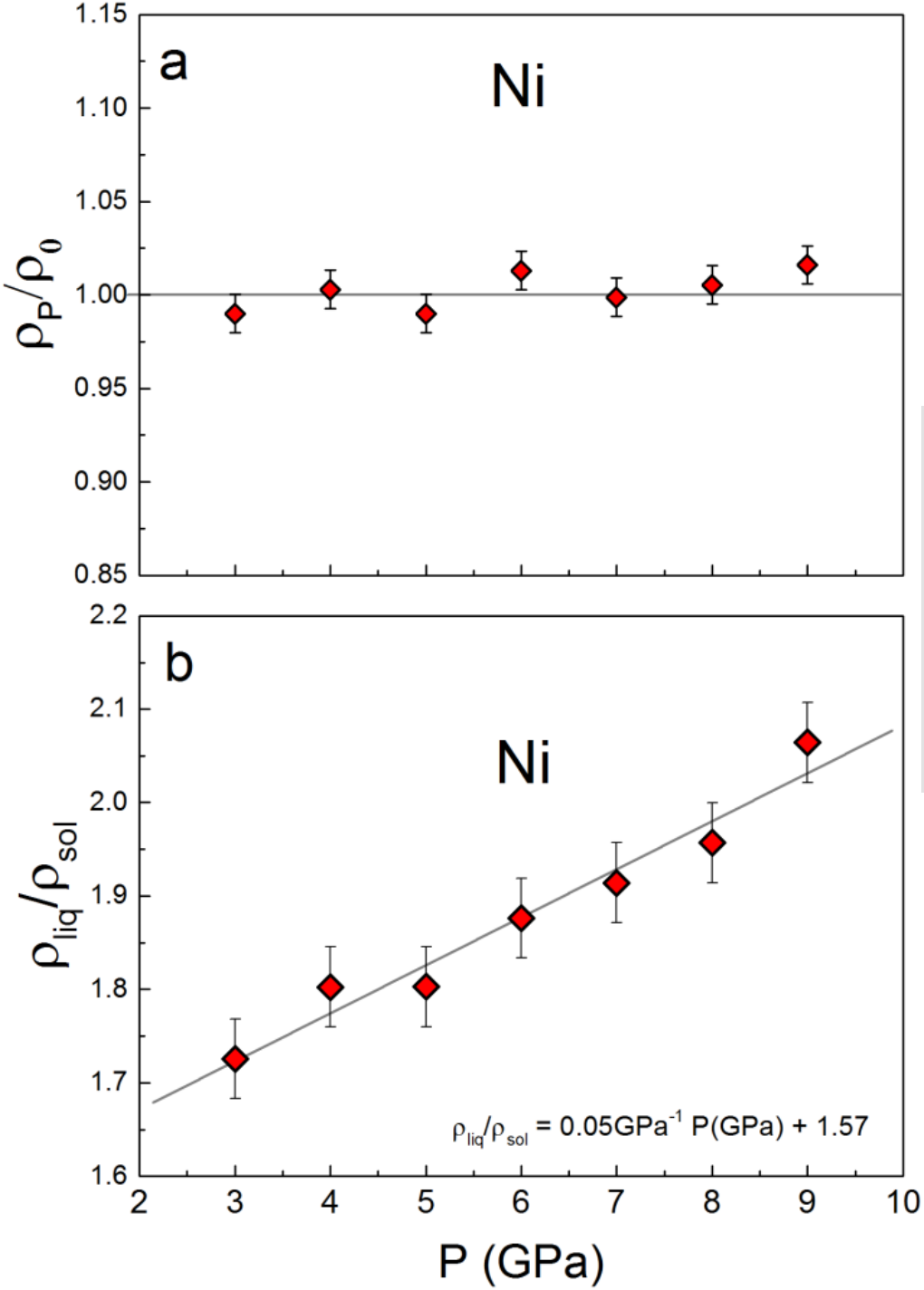
- **Electrical resistivity** of Ni at high P is **invariant along the melting boundary up to 9 GPa** within experimental uncertainty





Representative error bars for 5 GPa run in this study (error in T is within the symbol size).

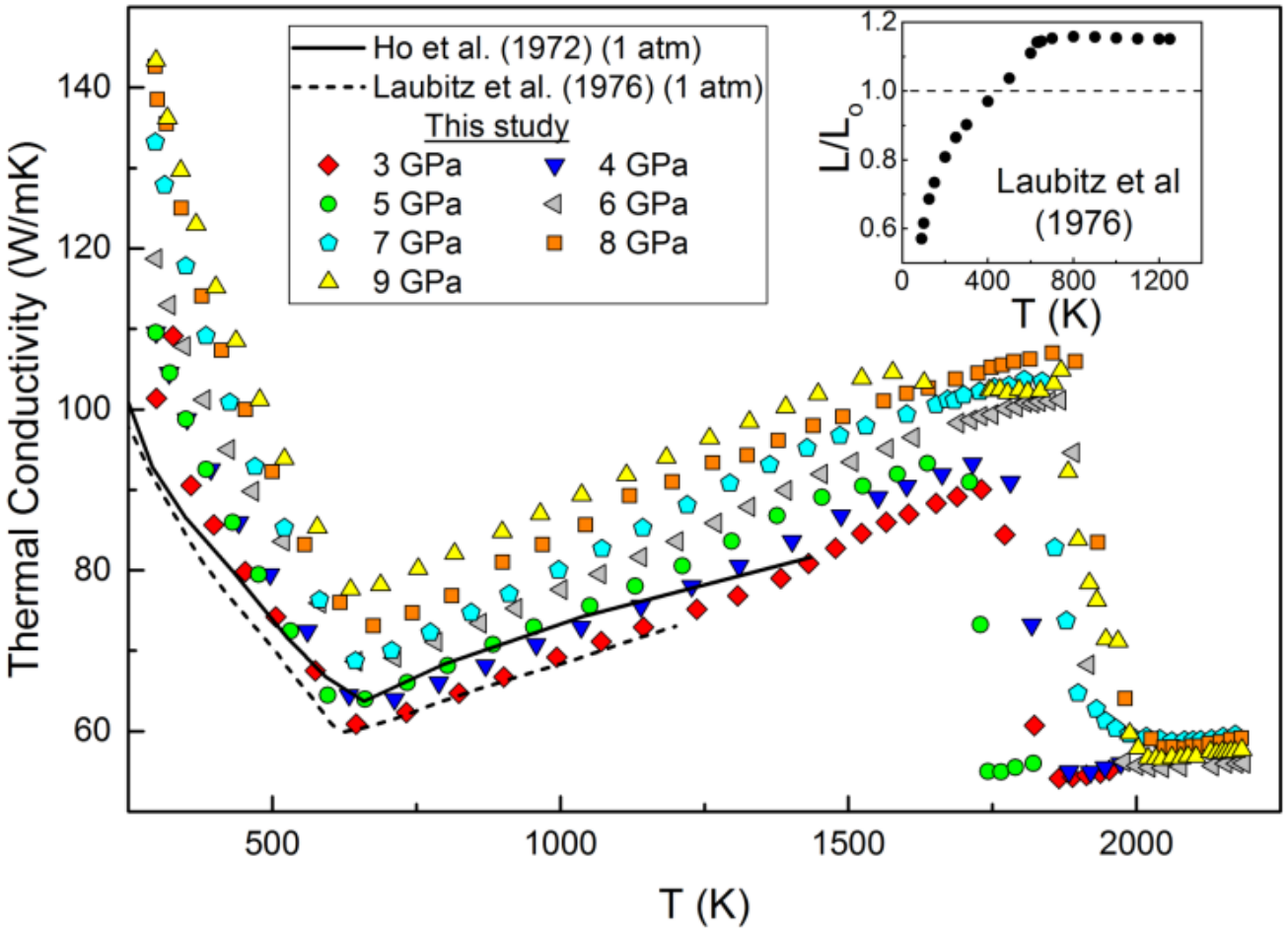
T-dependence of electrical resistivity of solid and liquid Ni at pressures in the range of 3 – 9 GPa compared with the resistivity at 1 atm.



(a) The ratio of electrical resistivity of molten Ni at pressures 3 – 9 GPa (ρ_P) and at ambient pressure (ρ_0).

(b) The ratio of electrical resistivity of liquid to solid Ni as a function of pressure.

Results – Cont'd



Wiedemann-Franz law

$$k = \frac{LT}{\rho}$$

k – thermal conductivity
L - Lorenz number
T – temperature
ρ - resistivity

Calculated electronic thermal conductivity for solid and liquid Ni at pressures 3 - 9 GPa using the Wiedemann-Franz law and the Sommerfeld value of the Lorenz number ($2.44 \cdot 10^{-8}$ WΩK^{-2}).
The inset is the ratio of the experimentally derived temperature dependent Lorenz number for Ni and the Sommerfeld value given above.

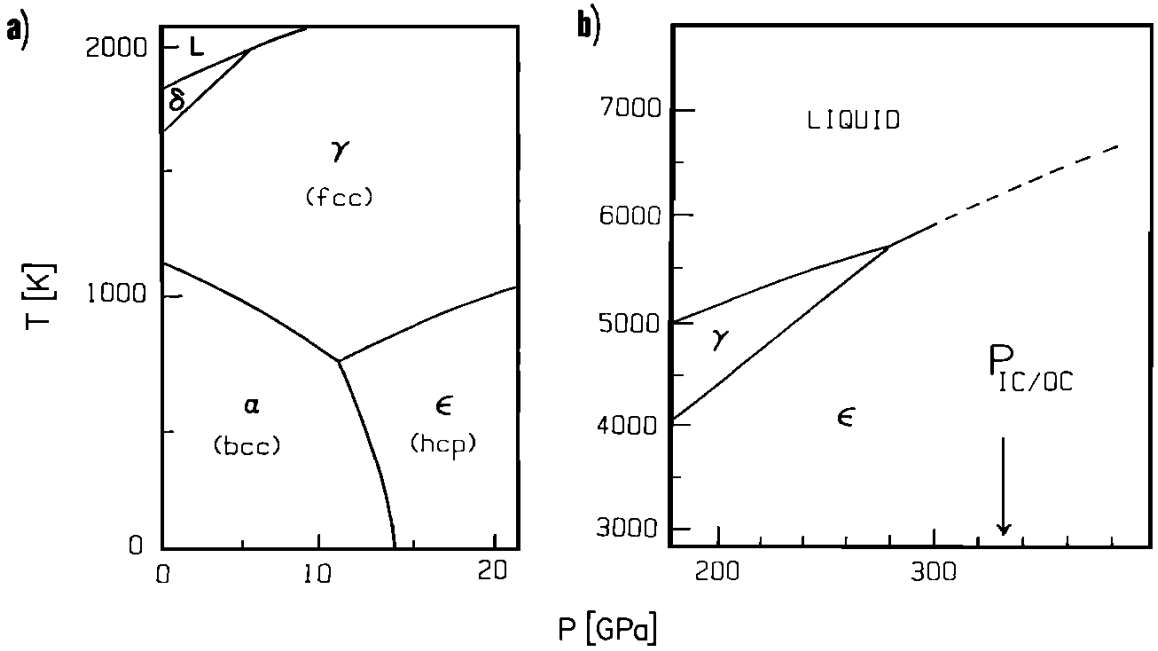
Discussion

- Possible explanation(s) of invariant electrical resistivity of Ni along the melting curve:
  - **constant Fermi surface** and an **invariant electron mean free path** (offset of P and T effects)?
  - role of **local symmetry structures** observed in molten transition metals with unfilled d-band?

- Can Fe be expected to mimic Ni behaviour?

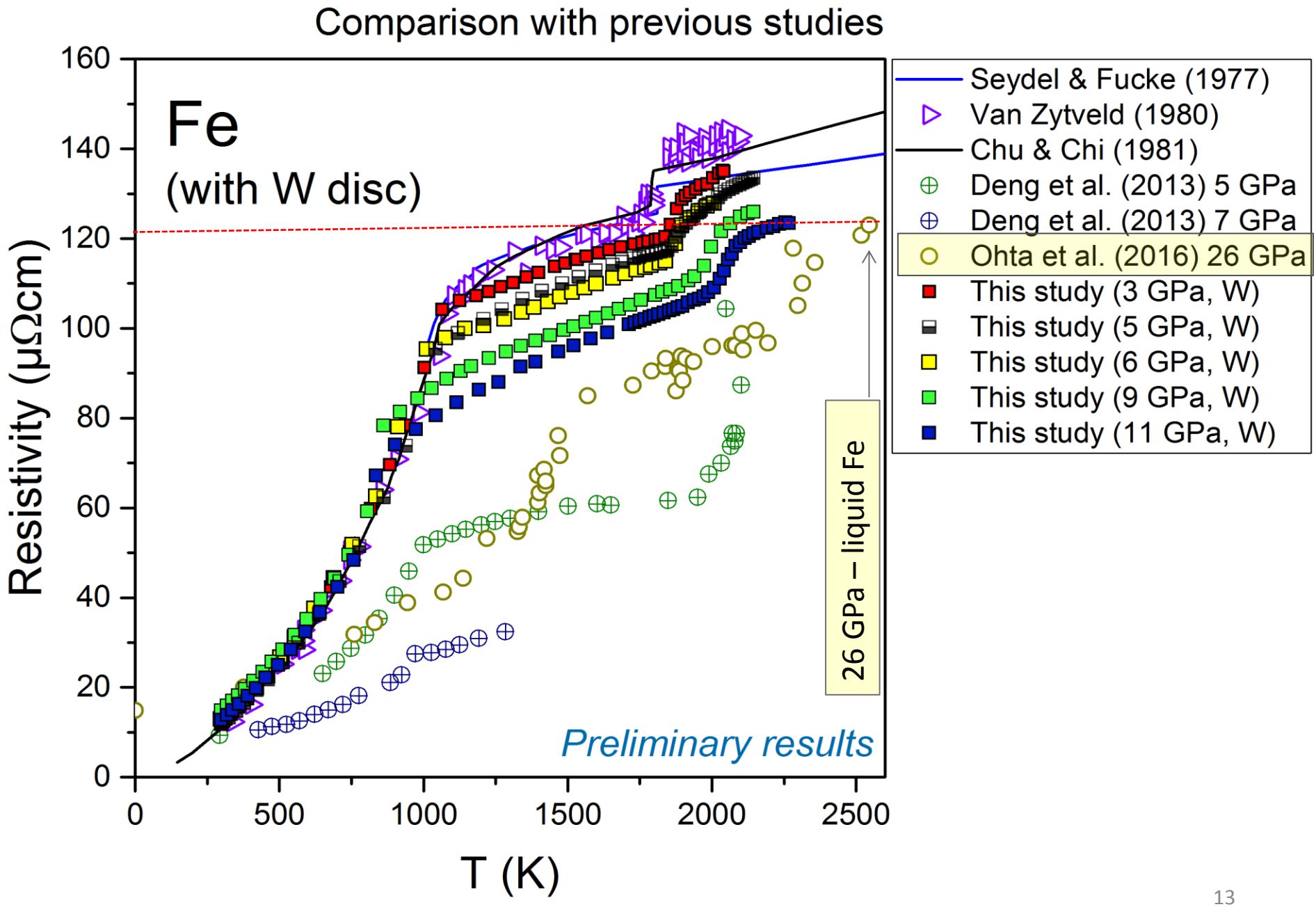
Electrical Resistivity of Solid and Liquid Fe at High Pressure

- Even more challenging
- Different approach from Ni (use of W and Pt discs to contain the liquid)
- Low P-T phase transitions



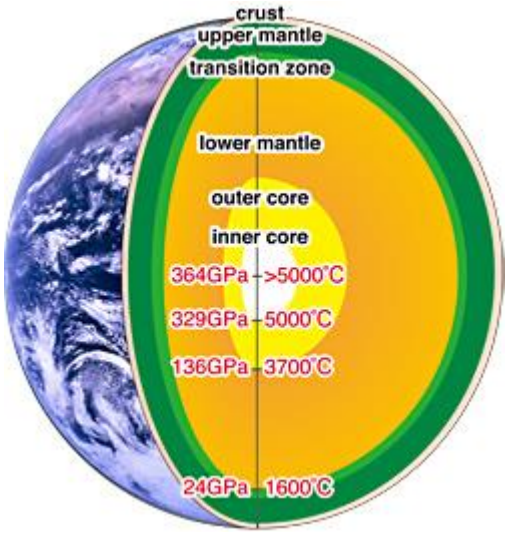
P, T phase equilibrium lines of the solid polymorphs of Fe and liquid Fe (a) remote from, and (b) close to, the P, T conditions of the Earth's core.

(Secco and Schloessin, 1989; after Anderson 1986)

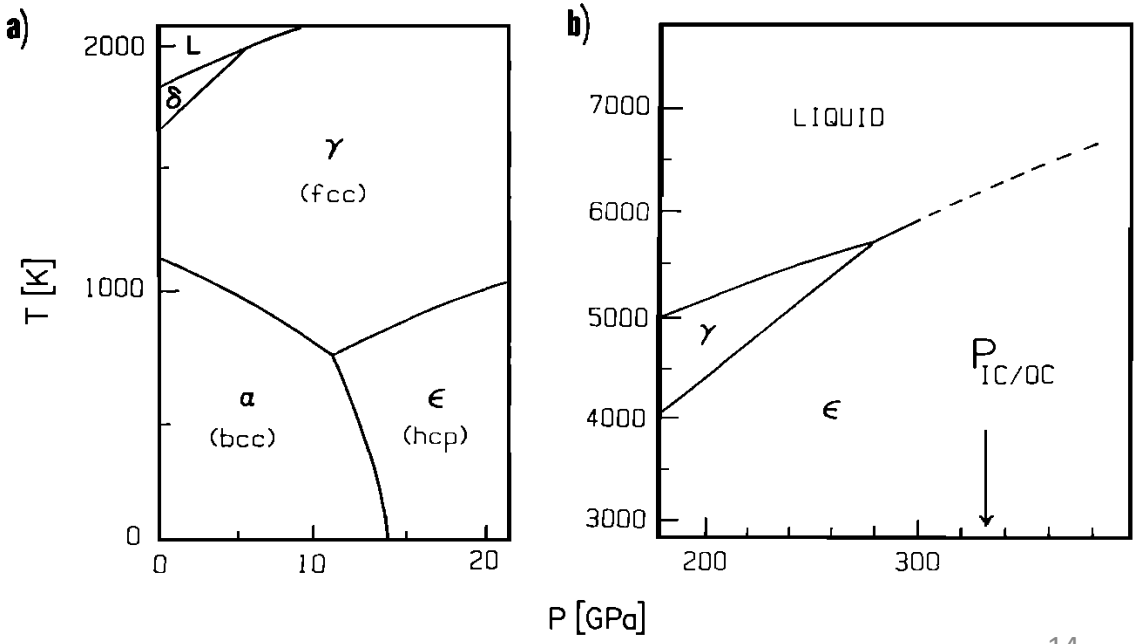

Comparison with previous studies
Fe
(with W disc)
Resistivity (μΩcm)
T (K)
0
20
40
60
80
100
120
140
160
0
500
1000
1500
2000
2500
Seydel & Fucke (1977)
Van Zytveld (1980)
Chu & Chi (1981)
Deng et al. (2013) 5 GPa
Deng et al. (2013) 7 GPa
Ohta et al. (2016) 26 GPa
This study (3 GPa, W)
This study (5 GPa, W)
This study (6 GPa, W)
This study (9 GPa, W)
This study (11 GPa, W)
26 GPa – liquid Fe
Preliminary results

Discussion and Conclusion

- Our results suggest the importance of pre-melt crystalline structure on behaviour of initial melt.
- Geophysical implications (e.g. dynamo, core heat flow…).
- However, the role of local structures in the initial Fe and Ni melt, along with the electron scattering mechanisms, and competing offsetting effects of P and T need further theoretical investigation.



Credit: Japan Agency for Marine-Earth Science and Technology



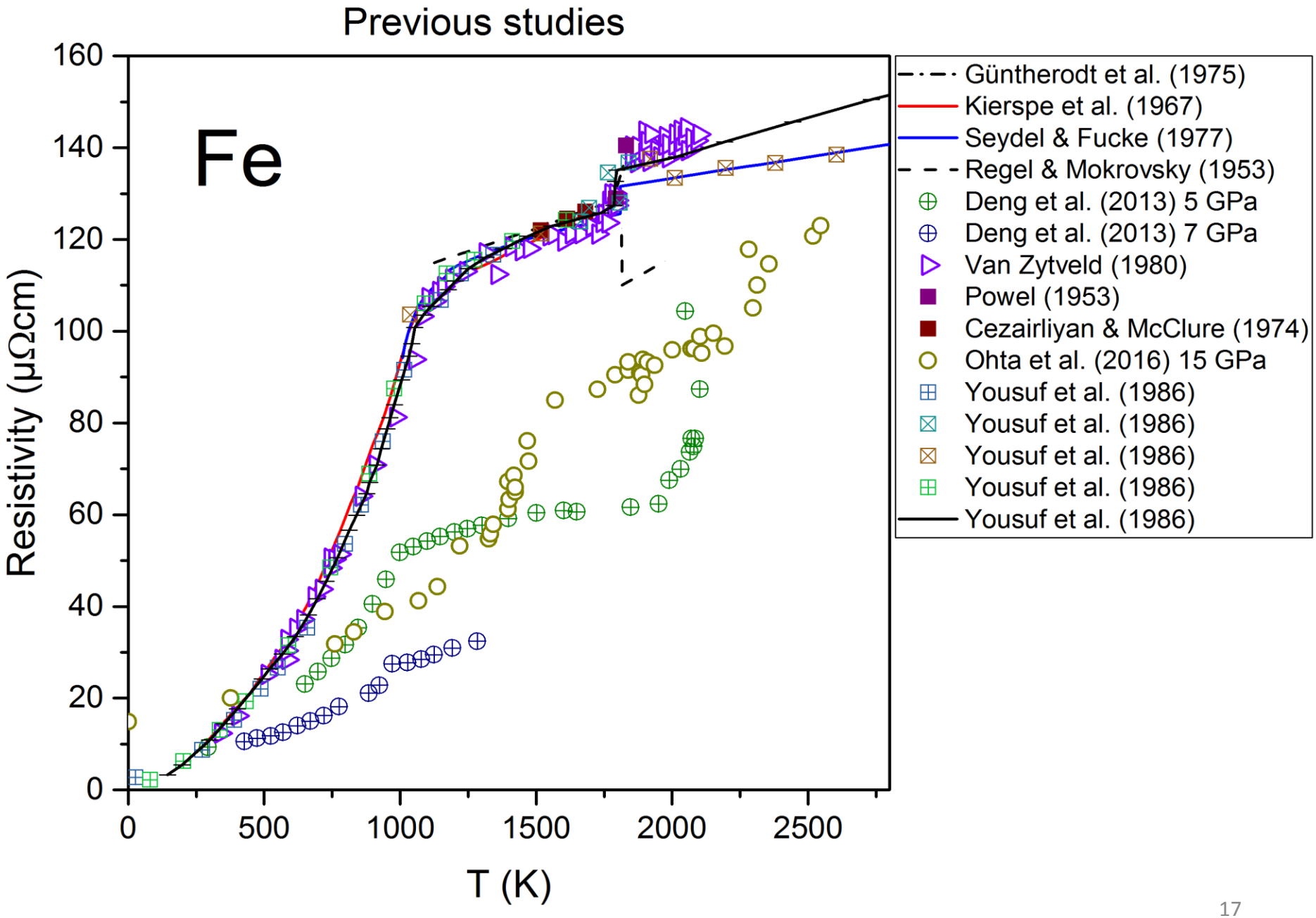
(Secco and Schloessin, 1989; after Anderson 1986)

Thank You!
Questions?

Acknowledgments:

We thank J. Jacobs for his assistance in fabrication of parts, S. Woods for preparing the recovered samples for electron microprobe analysis, and M. Beauchamp for running EMP analysis. RES also acknowledges useful discussions with T. Officer and I. Ezenwa. RAS acknowledges Canada Foundation for Innovation and NSERC for equipment and research funding, respectively.

Supplemental Material


Previous studies
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Resistivity (μΩcm)
T (K)
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Güntherodt et al. (1975)
Kierspe et al. (1967)
Seydel & Fucke (1977)
Regel & Mokrovsky (1953)
Deng et al. (2013) 5 GPa
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Van Zytveld (1980)
Powel (1953)
Cezairliyan & McClure (1974)
Ohta et al. (2016) 15 GPa
Yousuf et al. (1986)
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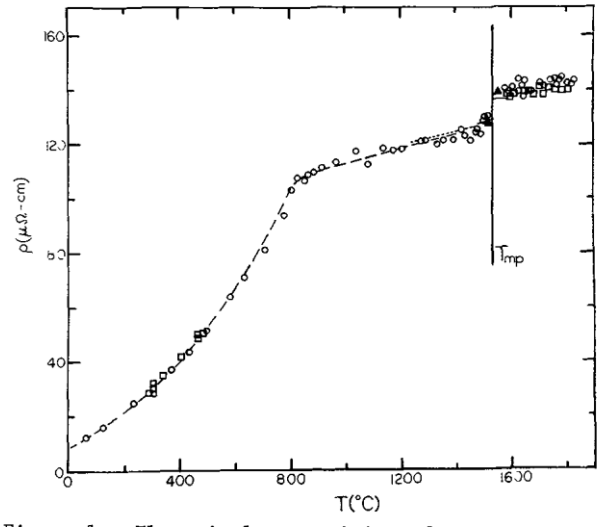
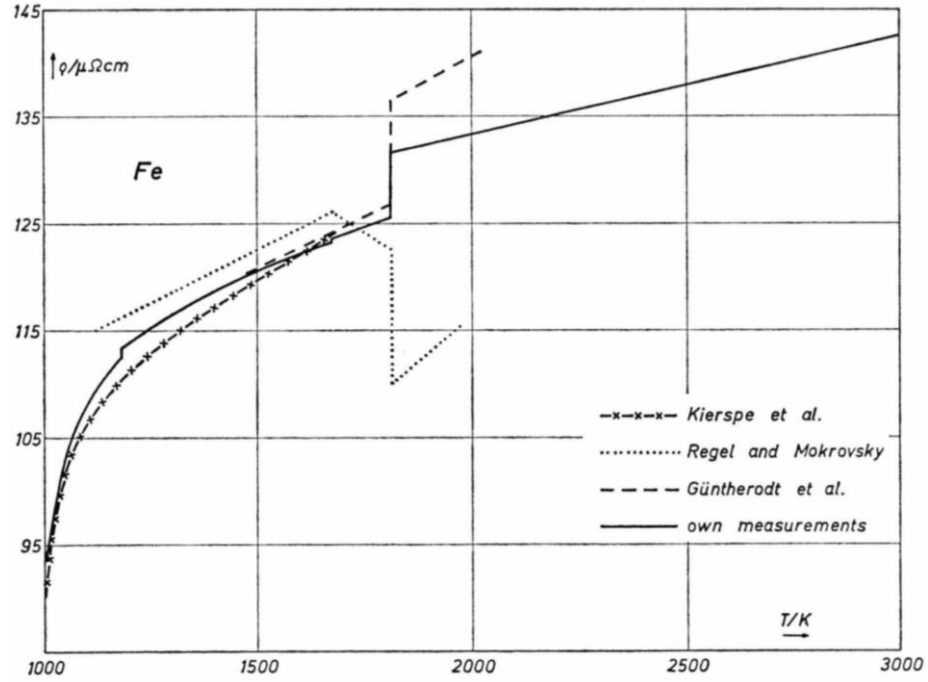
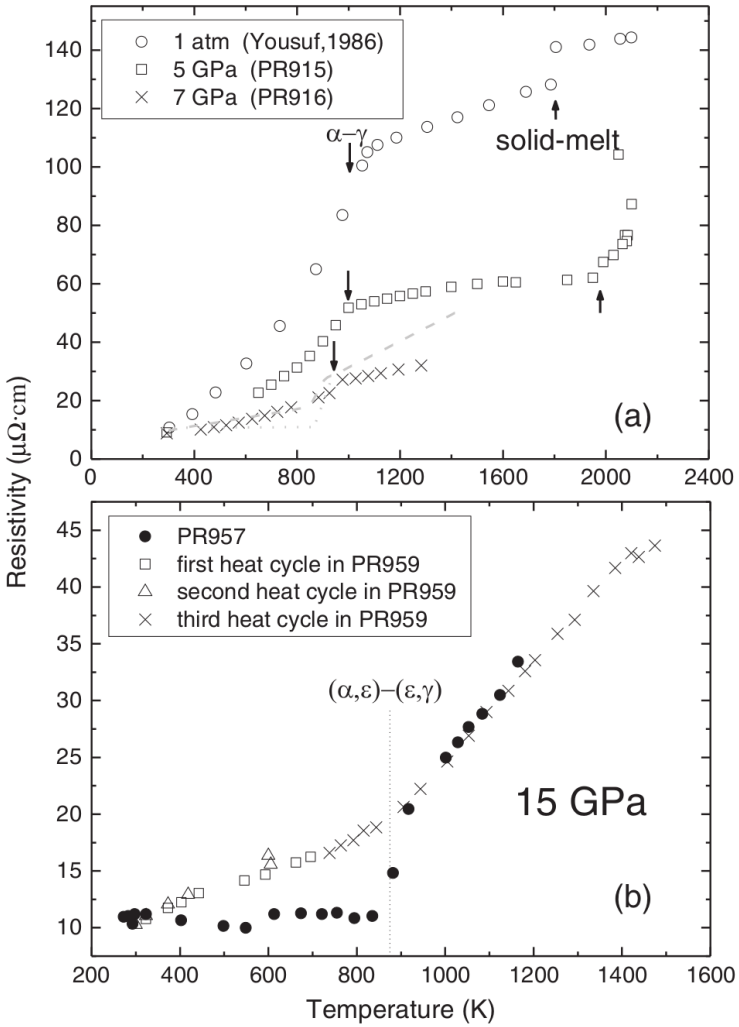


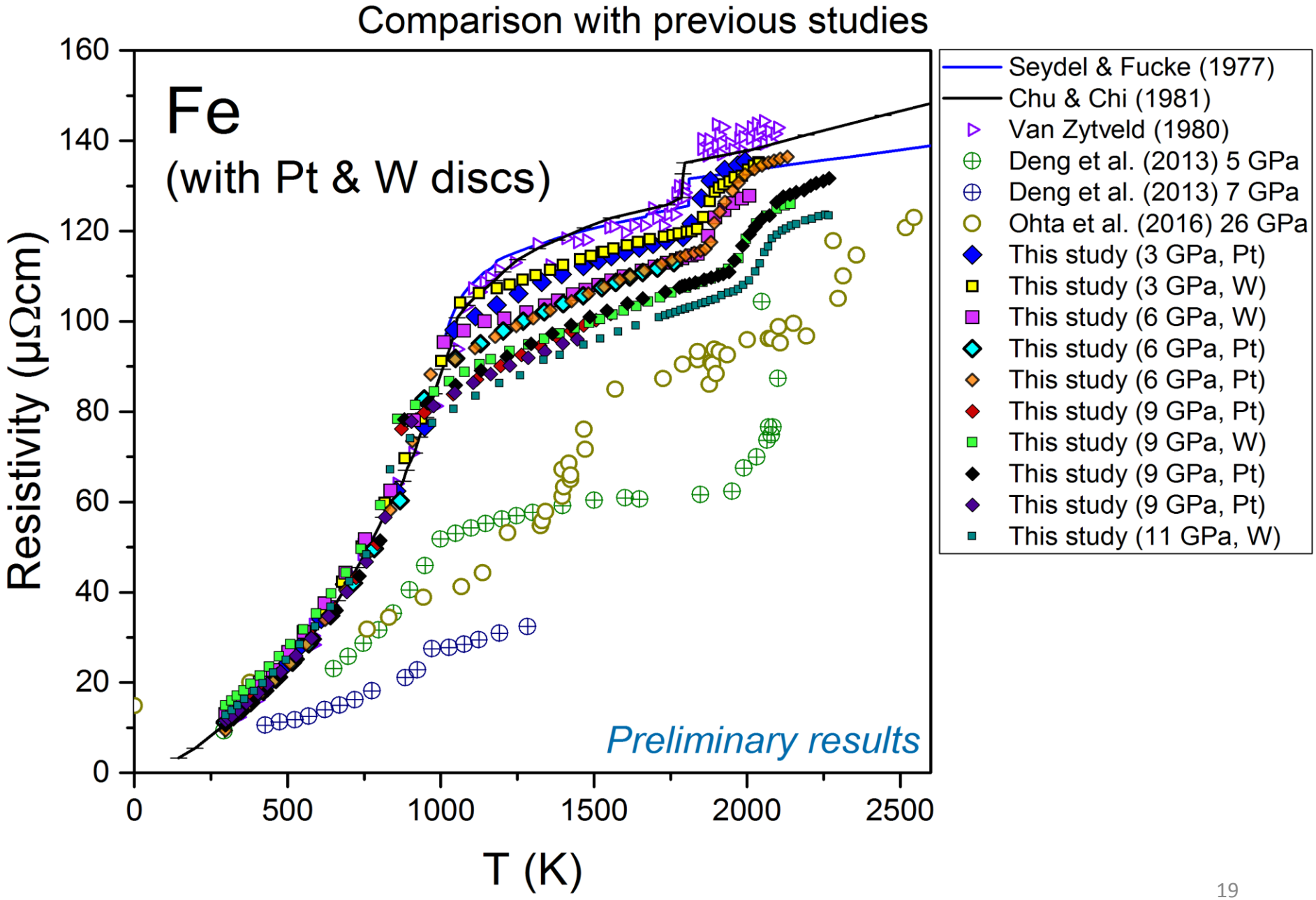
Van Zytveld, (1980)



Seydel and Fucke, (1977)



Deng et al., (2013)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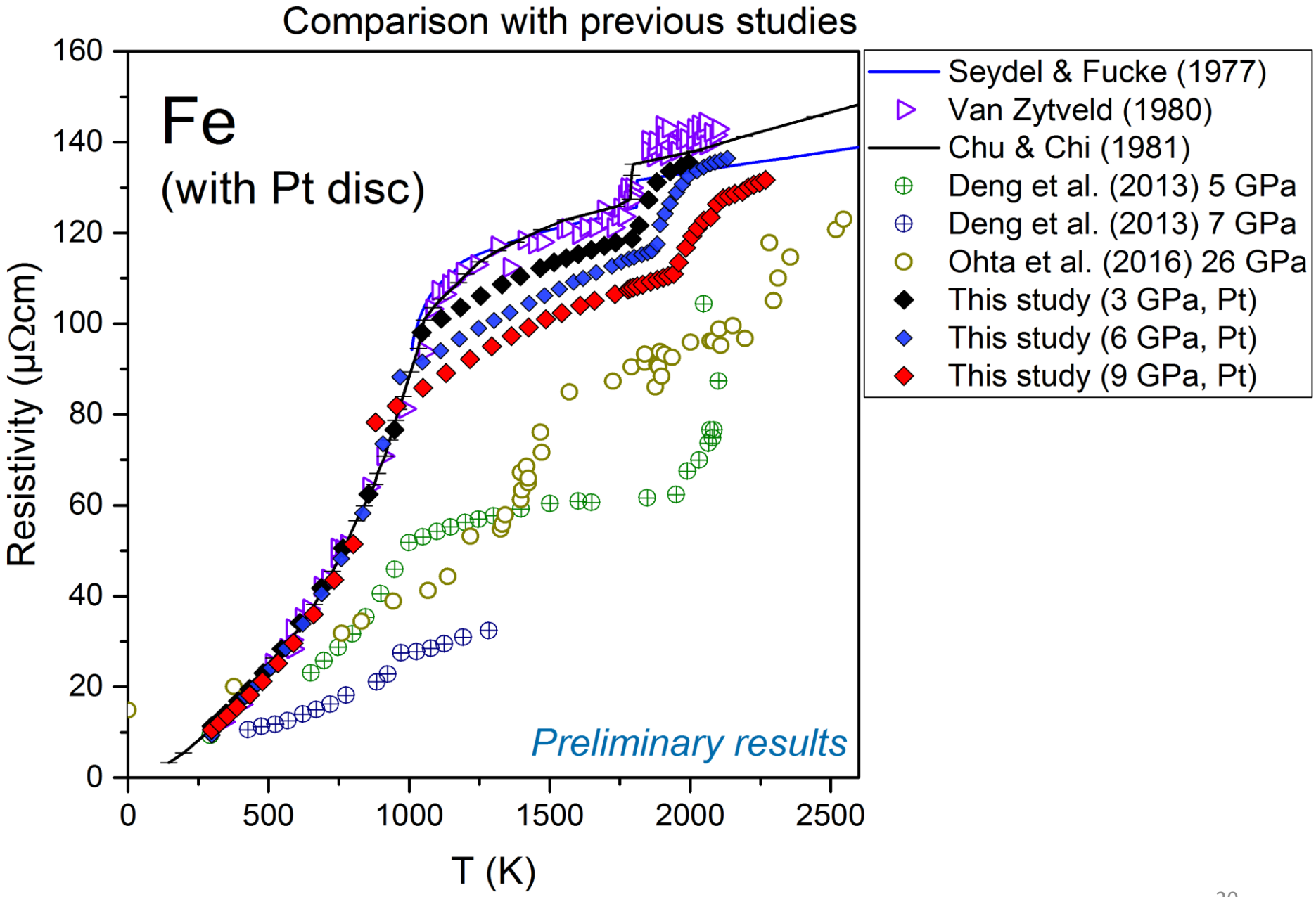
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Deng et al. (2013) 5 GPa
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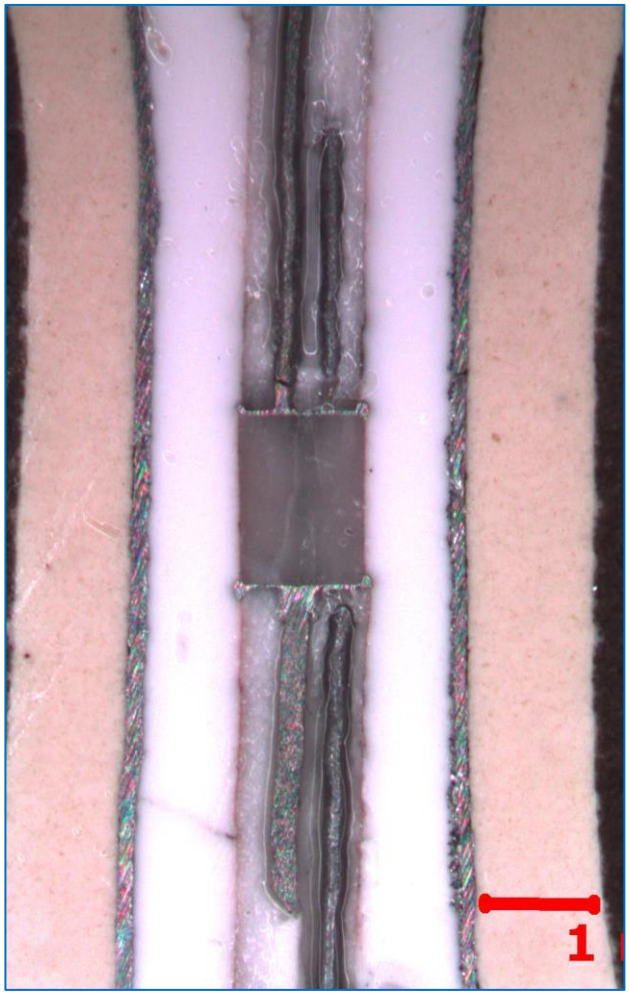
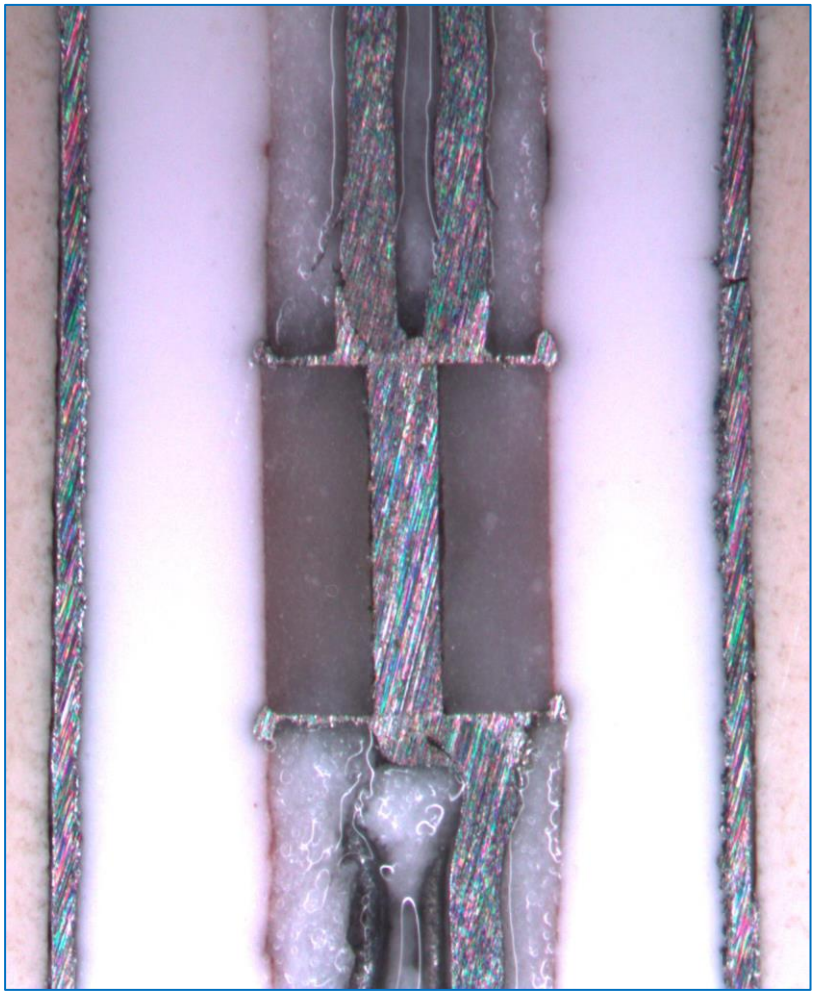
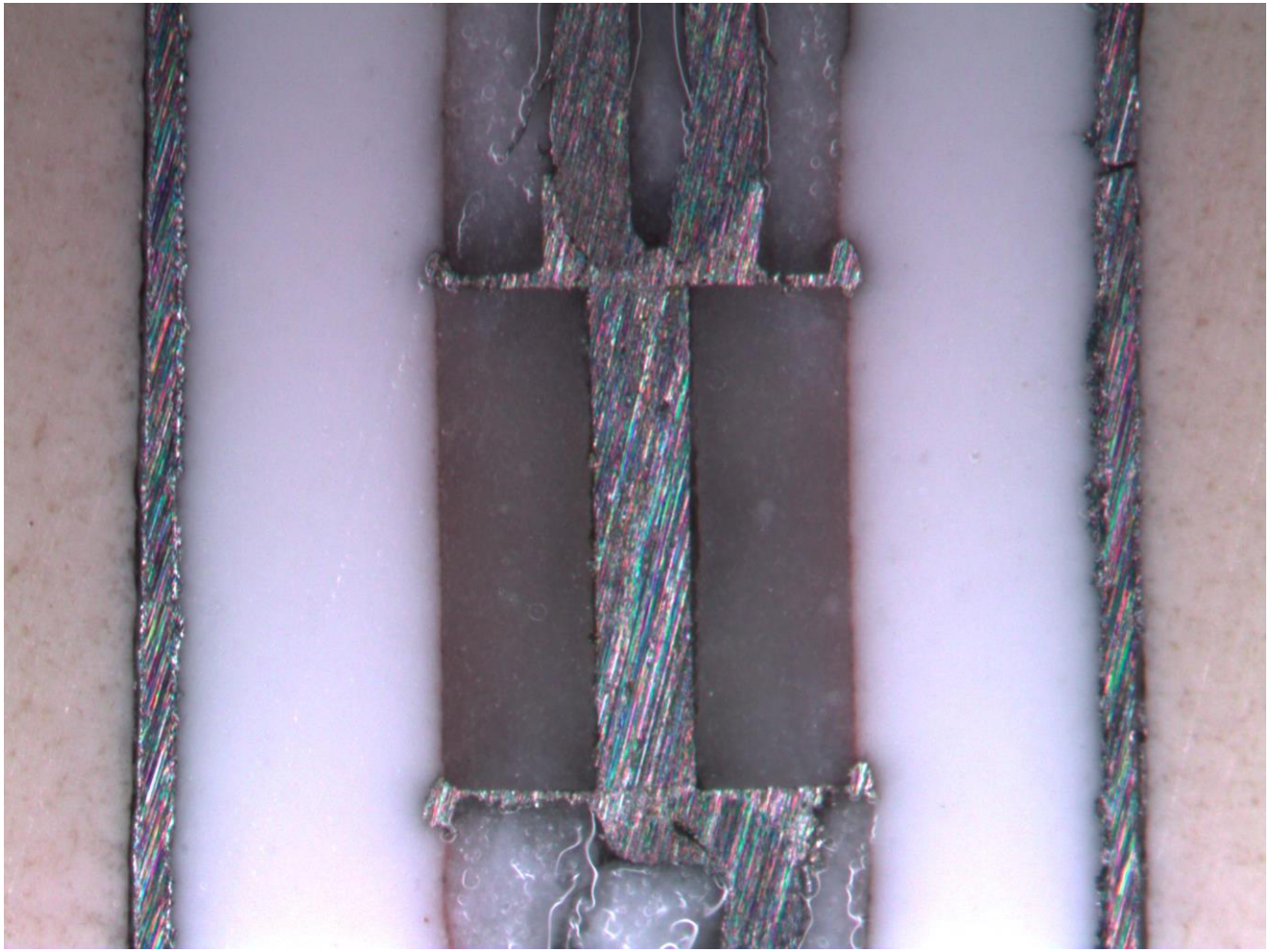
Ohta et al. (2016) 26 GPa
This study (3 GPa, Pt)
This study (6 GPa, Pt)
This study (9 GPa, Pt)

Pt-disc (6 Gpa, heated above Curie T)

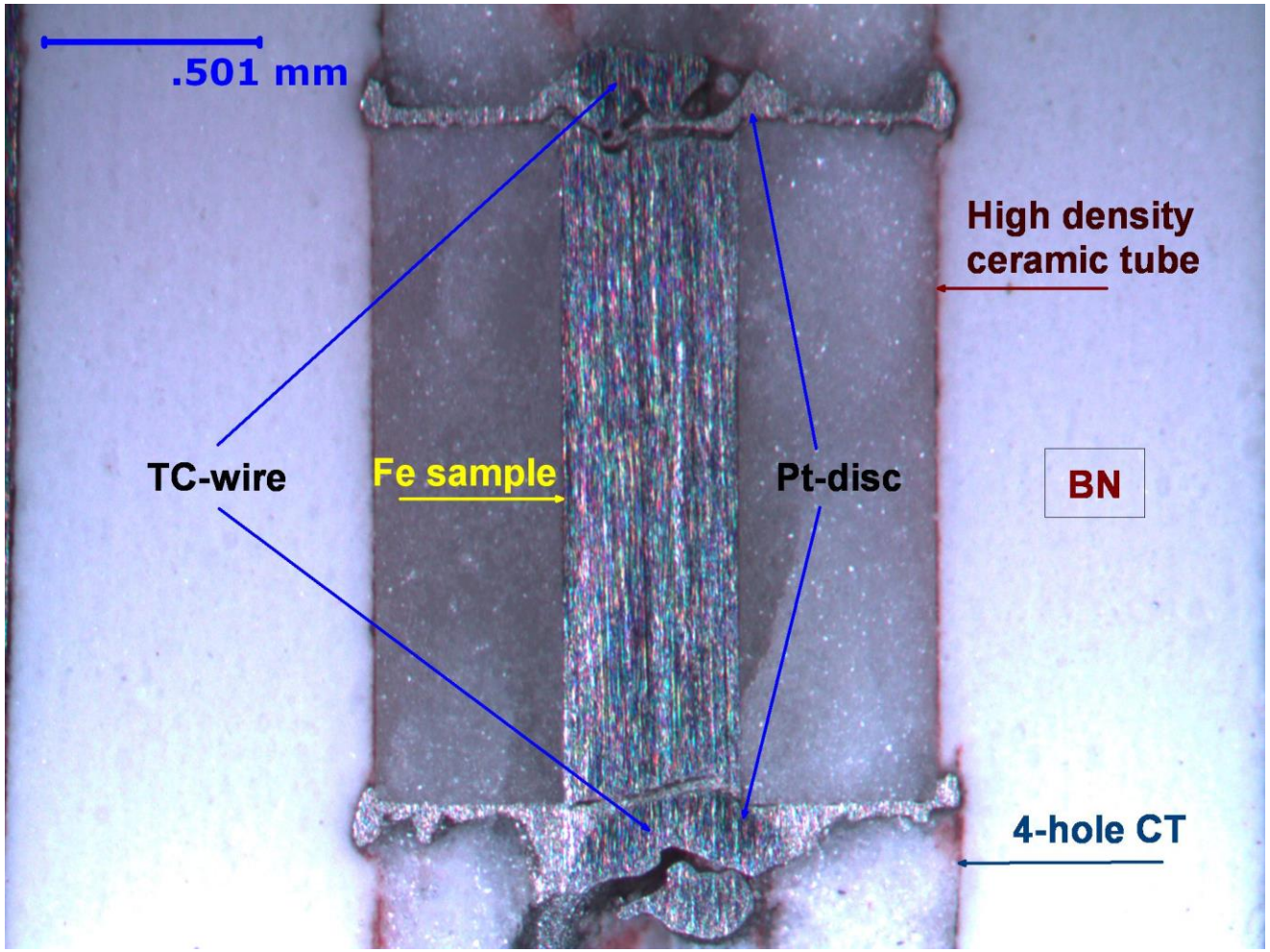


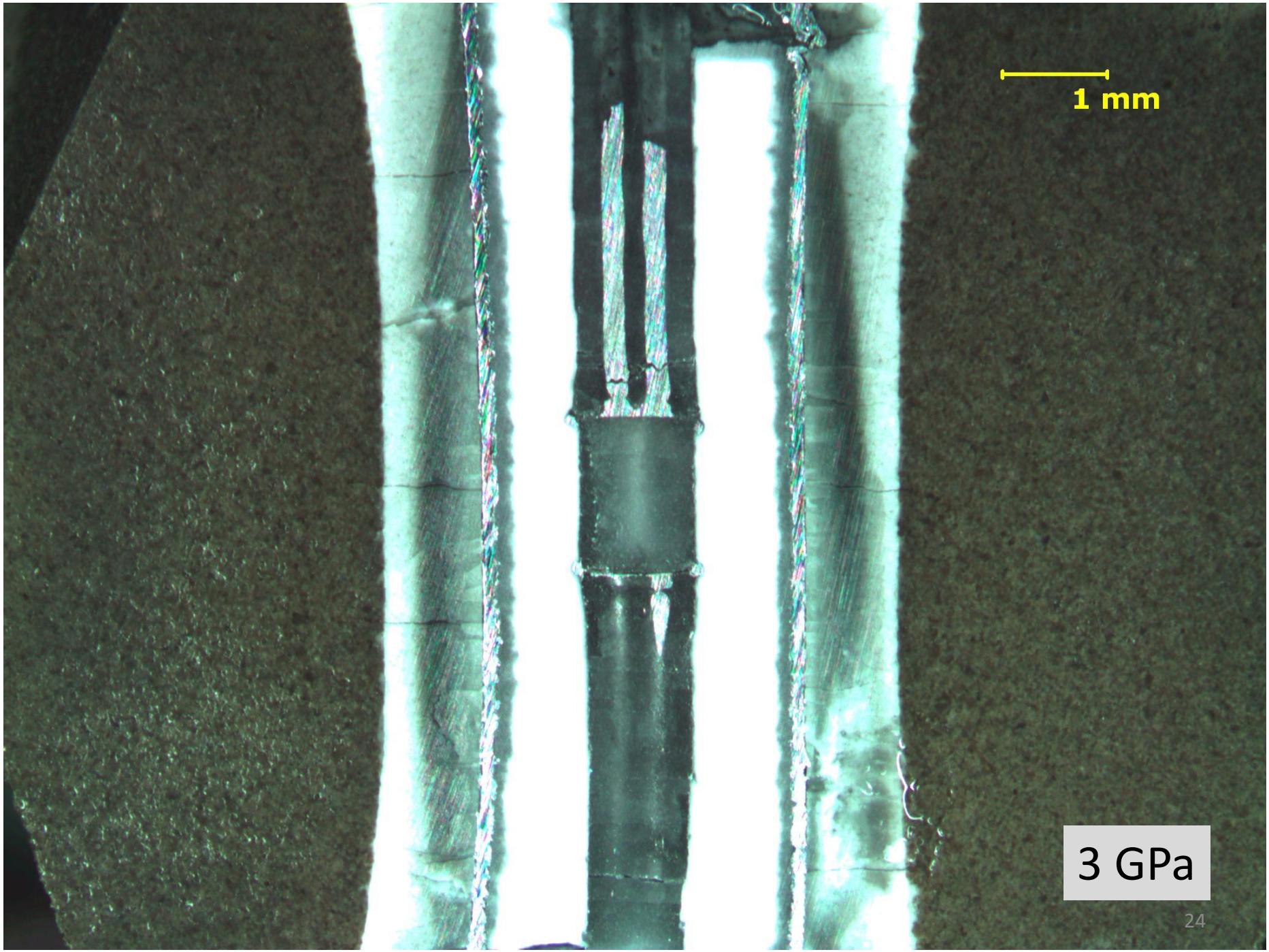


Pt-disc (6 GPa and Heated Above Curie T)



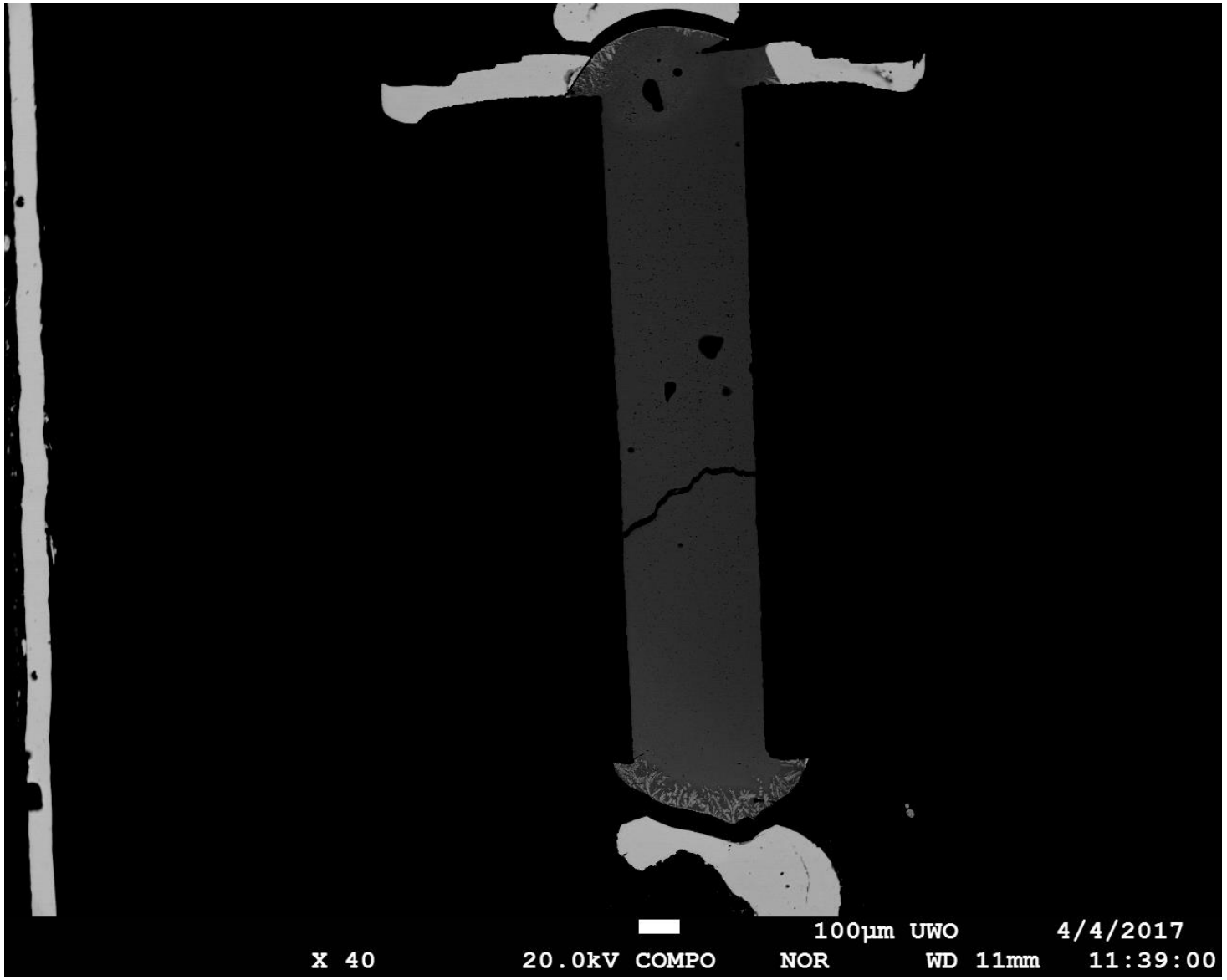
Pt-disc (9 GPa and Heated Above Curie T)




1 mm
3 GPa

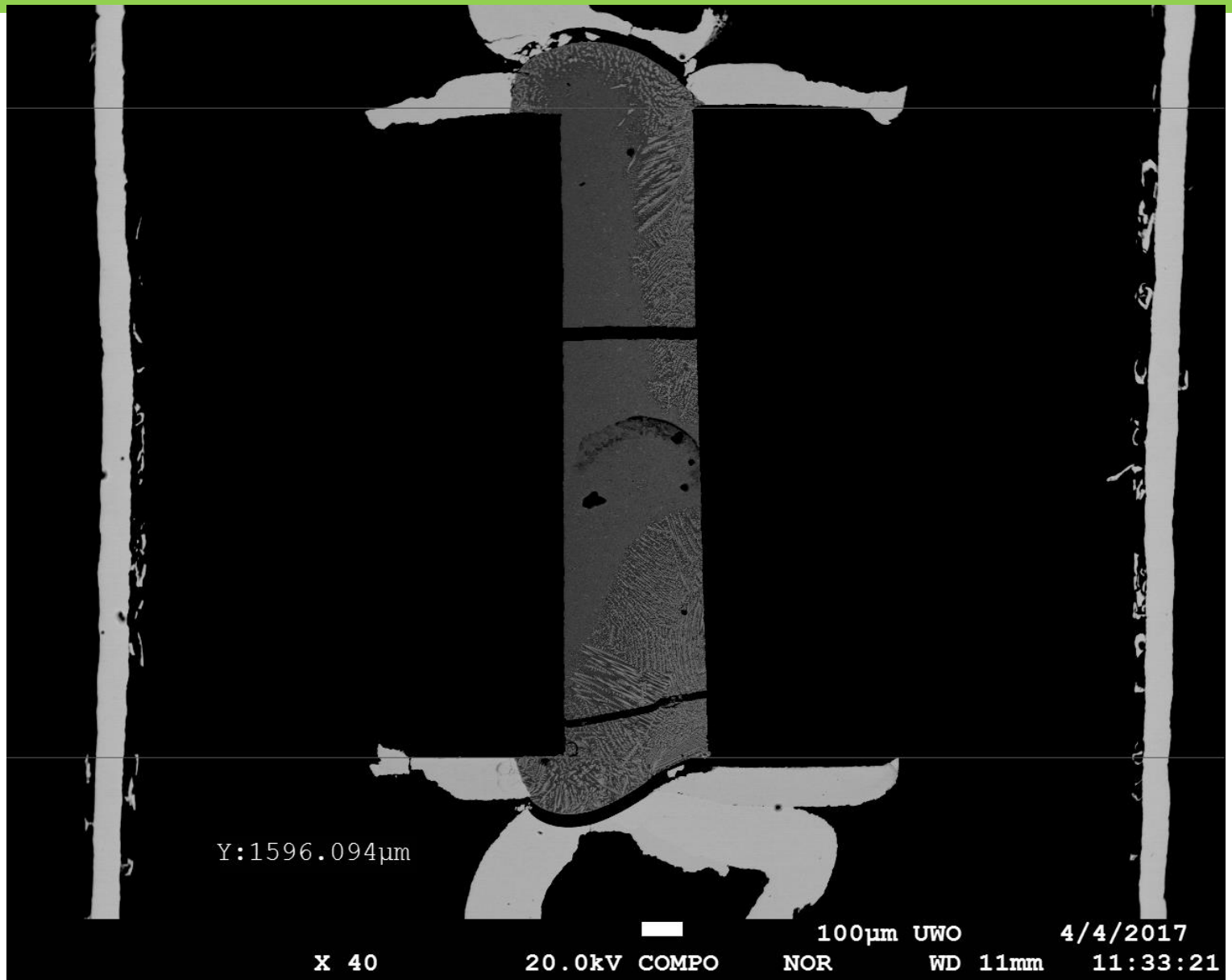
Fe-14 (Pt-disc, 6GPa, 1289 ± 10 °C (data exists)

Fe-6 (W-disc, 3GPa, 1768 ± 10 °C) –data exists

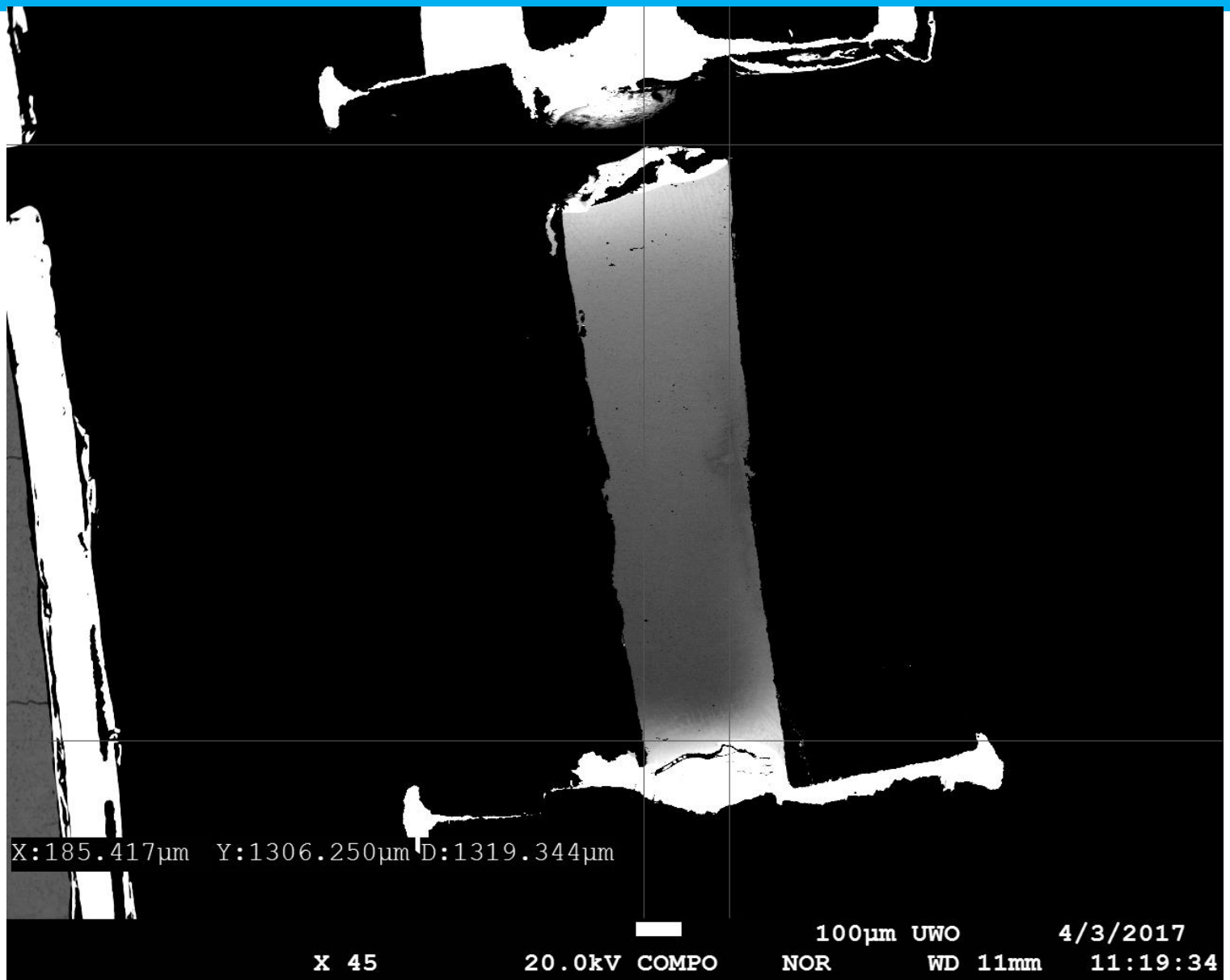


Fe-9 (Pt-disc, 6GPa, 1670 ± 20 °C (controlled heating)

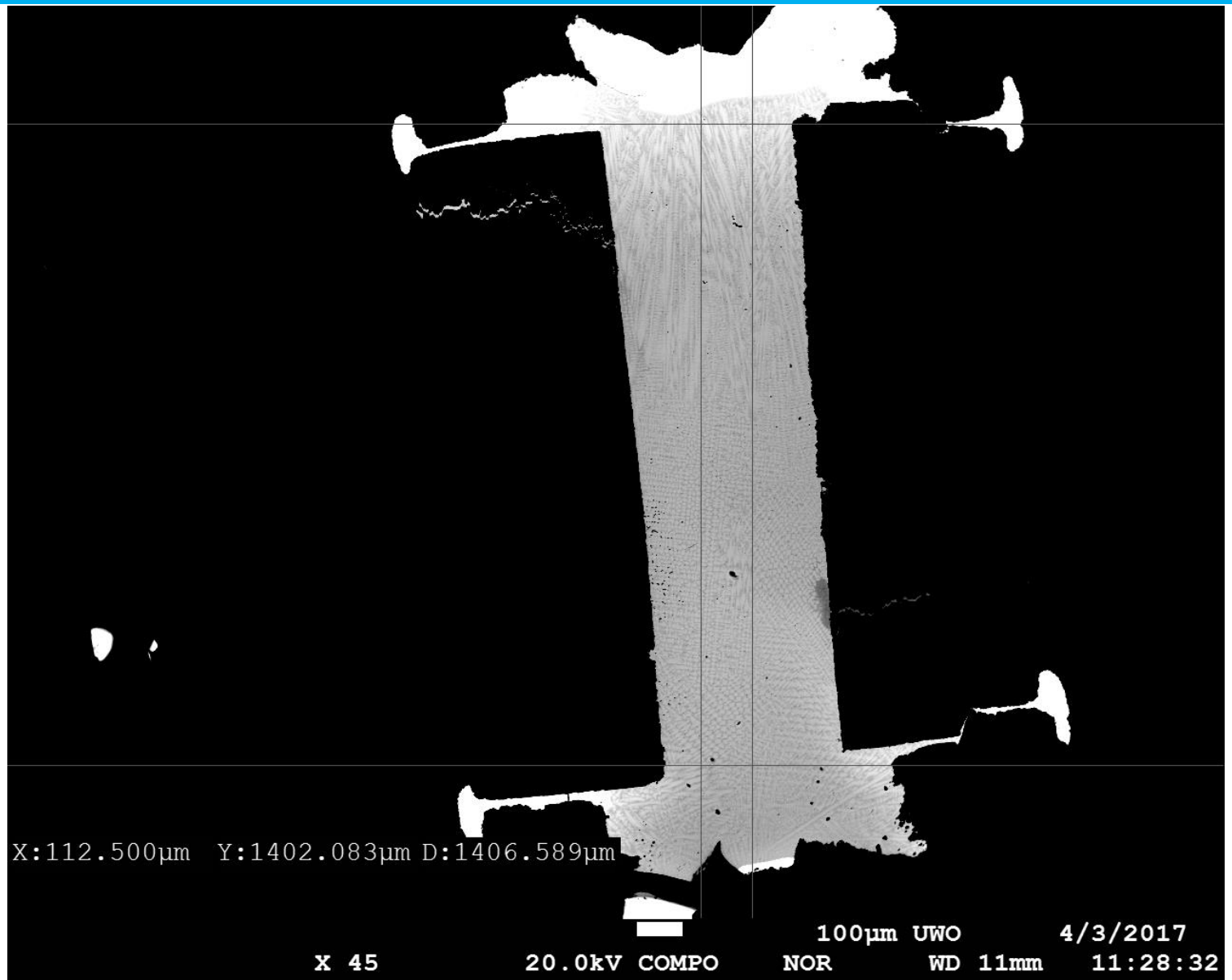
Fe-7 (W-disc, 3GPa, 1829 ± 10 °C (data exists)

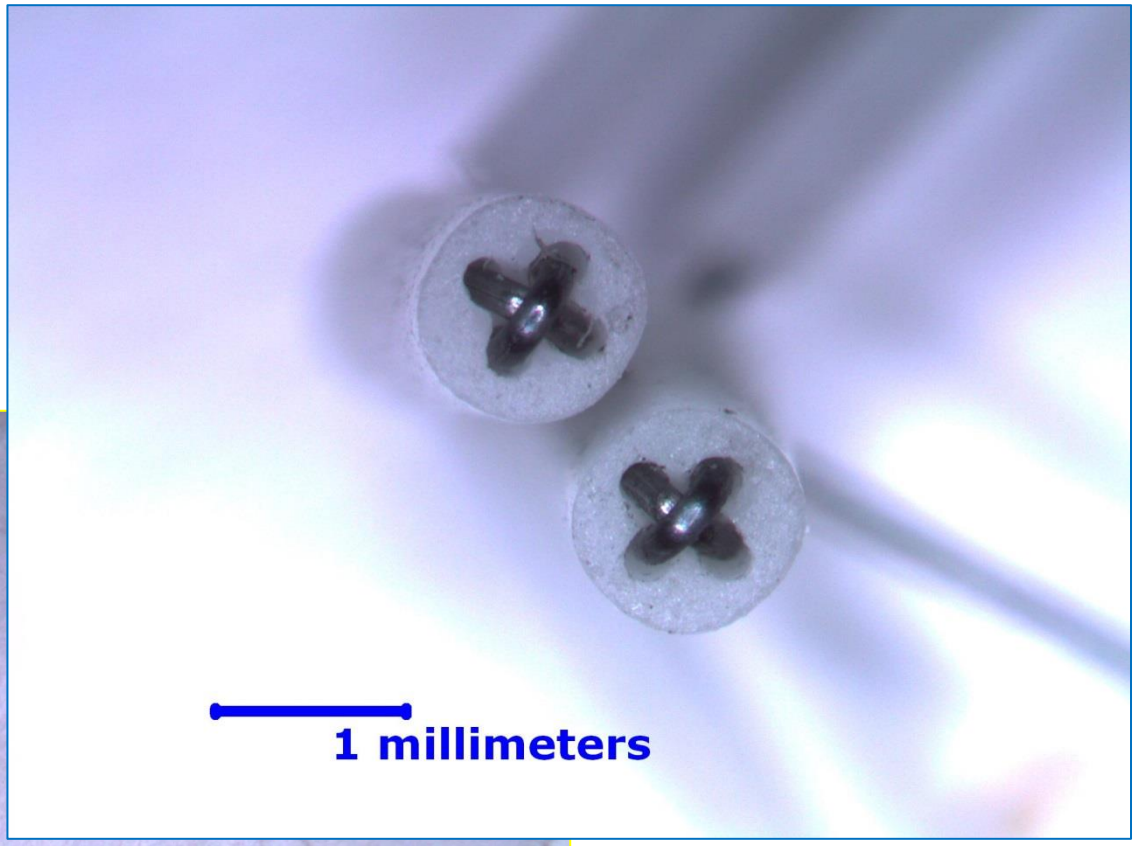


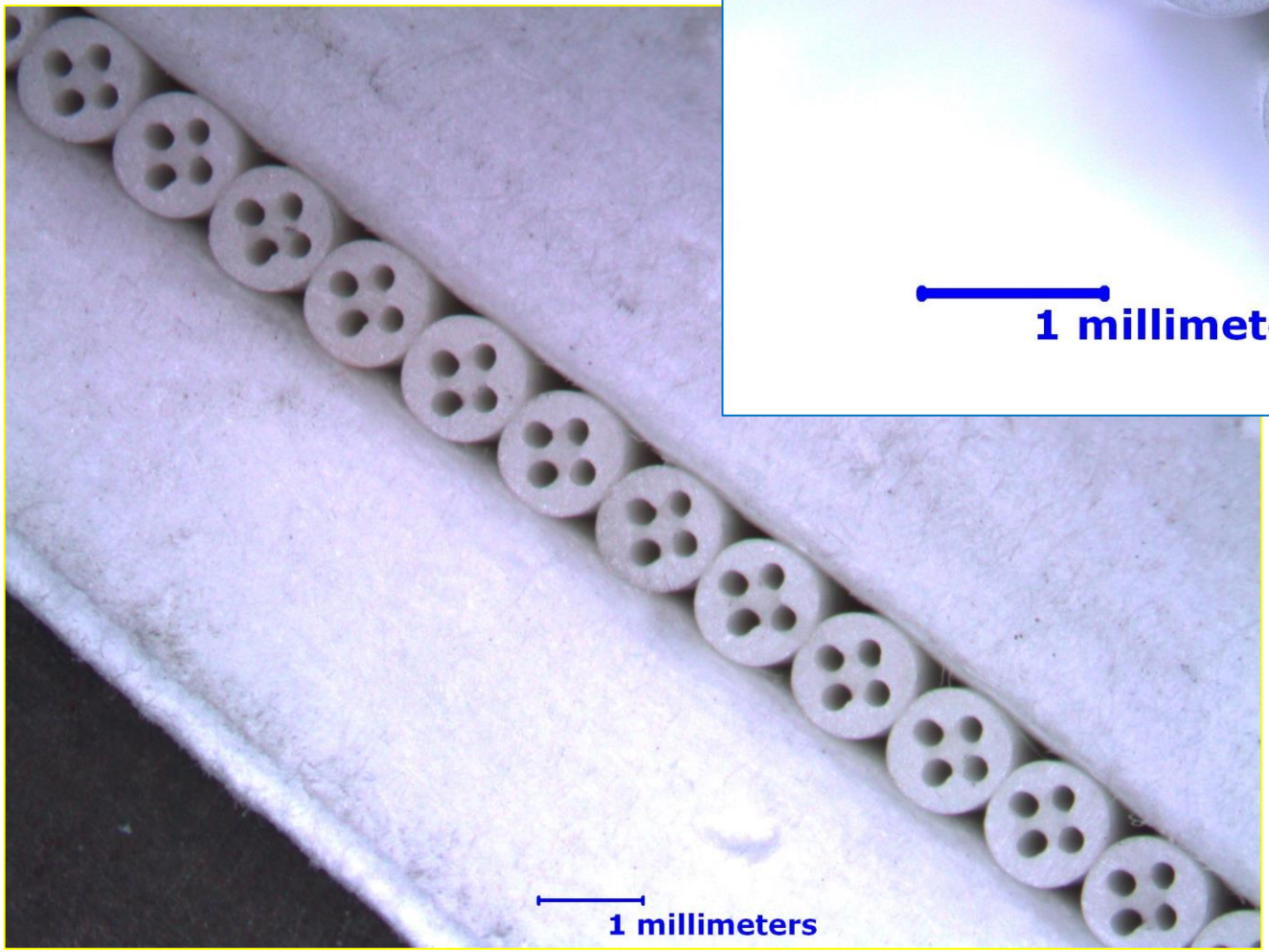
Fe-10 (Pt-disc, 6GPa, 1750 ± 40 °C (increased error because of experimental uncertainty)



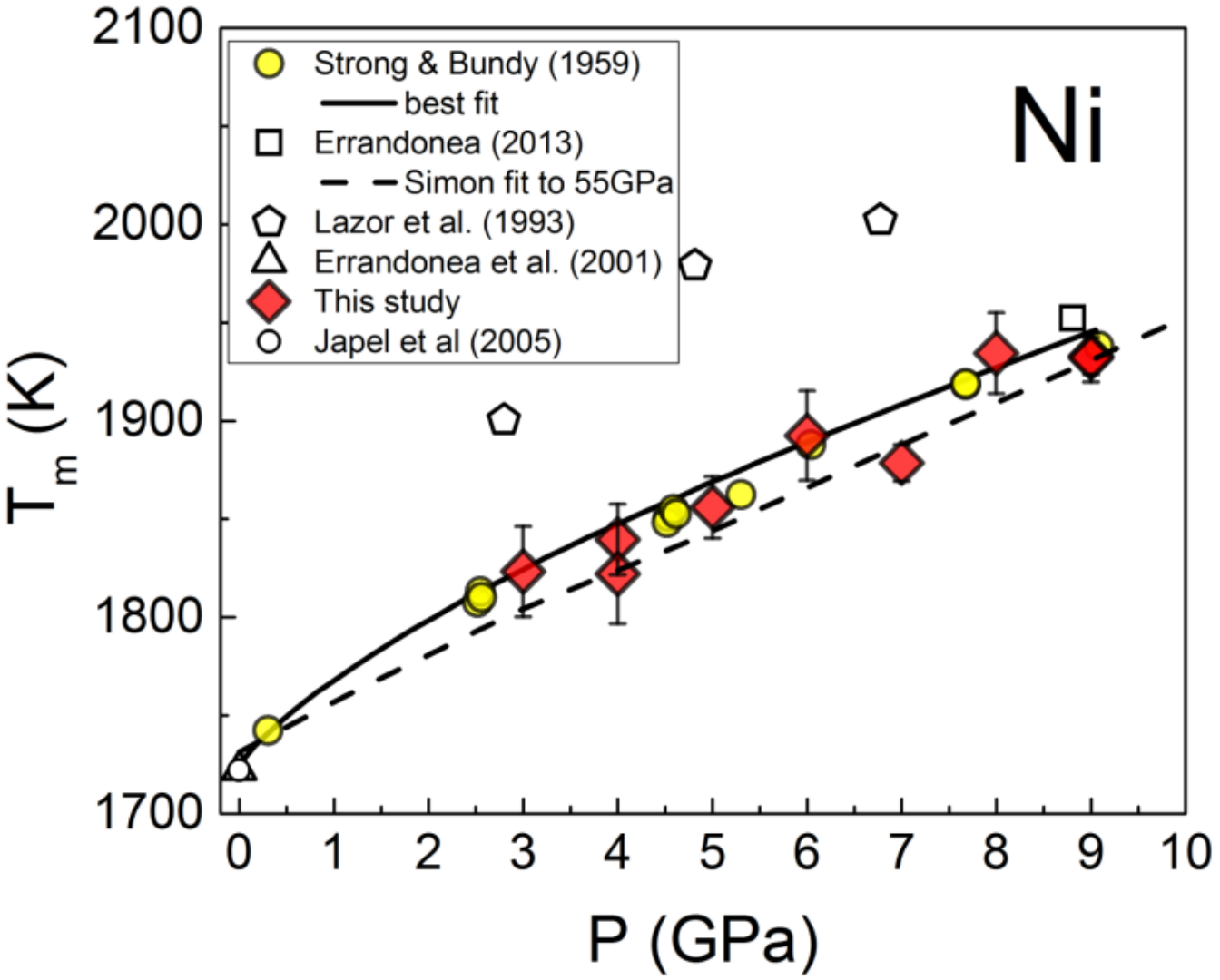
Fe-11 (Pt-disc, 6GPa, 1850 ± 40 °C (increased error because of experimental uncertainty)




1 millimeters


1 millimeters

Results



Melting temperatures of Ni obtained in this study compared with earlier studies in large volume press (yellow circles) and diamond anvil cells (other symbols)

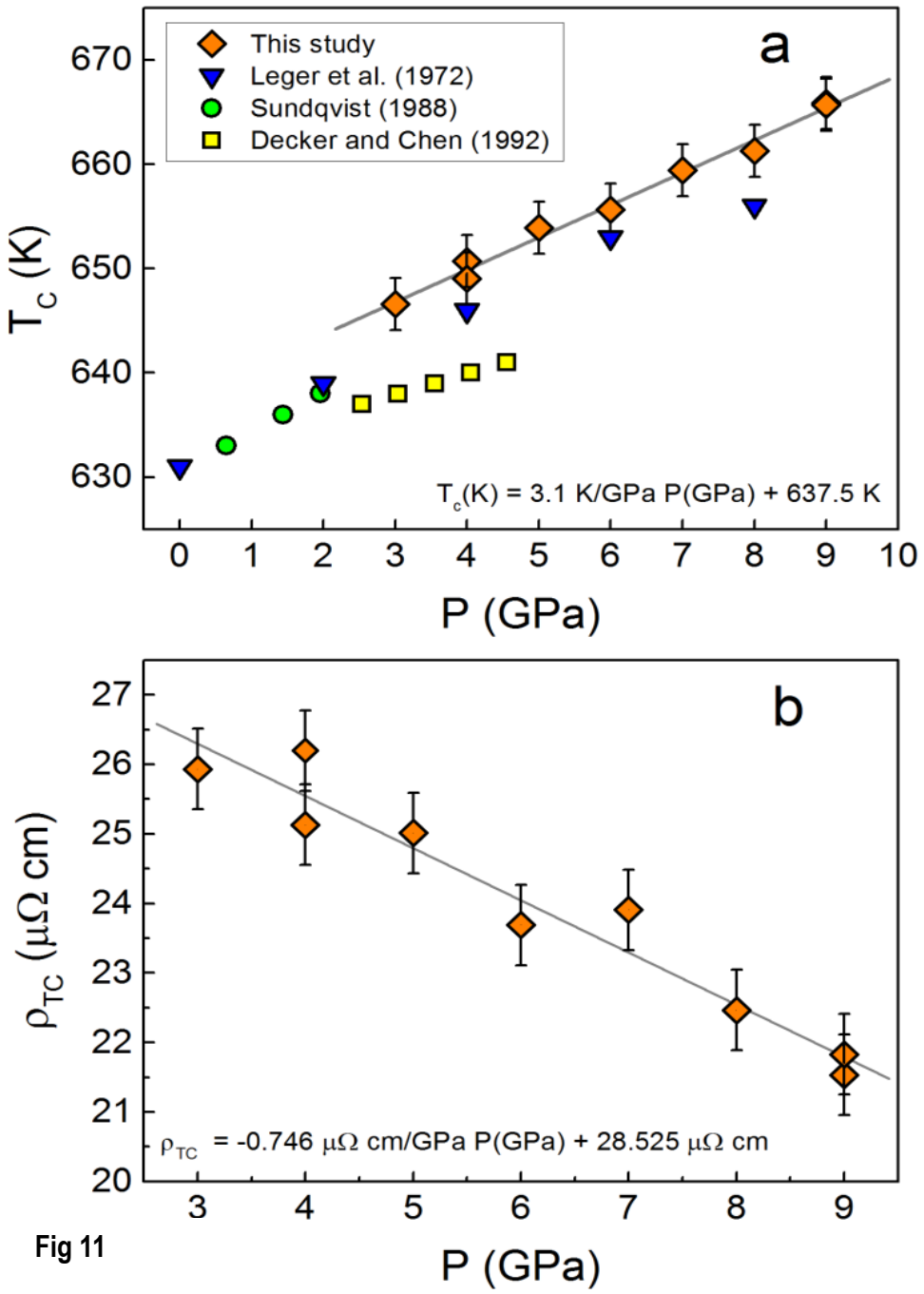

a
This study
Leger et al. (1972)
Sundqvist (1988)
Decker and Chen (1992)
T_c (K)
T_c(K) = 3.1 K/GPa P(GPa) + 637.5 K
P (GPa)
b
ρ_{TC} ($\mu\Omega$ cm)
ρ_{TC} = -0.746 $\mu\Omega$ cm/GPa P(GPa) + 28.525 $\mu\Omega$ cm
P (GPa)

Fig 11

From Mott's formula for electrical conductivity [e.g. Mott, 1972; Shimoji, 1977; Mott, 1980] which is the basis forthe Kubo-Greenwood expression used to calculate resistivity of transition metals[e.g. deKoker et al, 2012; Pozzo et al, 2012;13;14], the electrical resistivity of liquid metals is inversely proportional to the Fermi surface size and the mean free path of conduction electrons. Notably, in its unmodified form, the expression below is best applicable to simple liquid metals; however it can also be applied to complex liquids (i.e. molten transition metals) [Mott, 1980]:

$$\sigma_e = S_F \mid e \mid^2 l/12\pi^3\hbar \qquad (1)$$

Here, σ_e is electrical conductivity, S_F is the area of the Fermi surface, and l is the electron mean free path which is defined as the product of the Fermi velocity, v_F, and the conduction electron relaxation time, τ, where $l = v_F\tau$ and $\hbar$ is the modified Planck constant. The relaxation time varies with the density of states (DOS) at the Fermi level [Shimoji, 1977] and the combined scattering mechanisms operating at a specific P and T.

- Ni ([Ar] 4s2 3d8) is a close electronic analogue of Fe ([Ar] 4s2 3d6)