

# Charge density waves in the heavy fermion systems $\text{LaPt}_2\text{Si}_2$ and $\text{UPt}_2\text{Si}_2$

– similar but different –

P. Hans <sup>1</sup>, R. Baumbach <sup>2</sup>, D. R. Tadisetti <sup>1</sup>, V. Petkov <sup>1</sup>

<sup>1</sup>Central Michigan University – Mt Pleasant, MI (United States)

<sup>2</sup>Florida State University – Tallahassee, FL (United States)



[phlpp.hns@gmail.com](mailto:phlpp.hns@gmail.com)  
[petko1vq@cmich.edu](mailto:petko1vq@cmich.edu)



# Starting point

- 2 compounds:  $\text{LaPt}_2\text{Si}_2$ ,  $\text{UPt}_2\text{Si}_2$
- Same structure type
- “strange”, non-linear behavior of magnetic and electric properties and phase transitions
- Properties of both compounds differ.
  
- Charge density waves (CDW) attributed to both compounds.
- Could be interesting materials for superconductors or magnets

# Used concepts

- Charge Density Waves: Charge Density Waves (CDWs) are periodic modulations of the crystal lattice by condensation of charges that lead to the emergence of superstructures, accompanied by the opening of a band gap at the Fermi level.

# This work: study using X-ray total scattering

Reduced and normalized radial distribution function ("PDF")

$$G(R) = 4\pi R[\rho(R) - \rho_0]$$

$$G(R) = \int_{Q_{min}}^{Q_{max}} Q[S(Q) - 1] \sin(QR) dQ$$



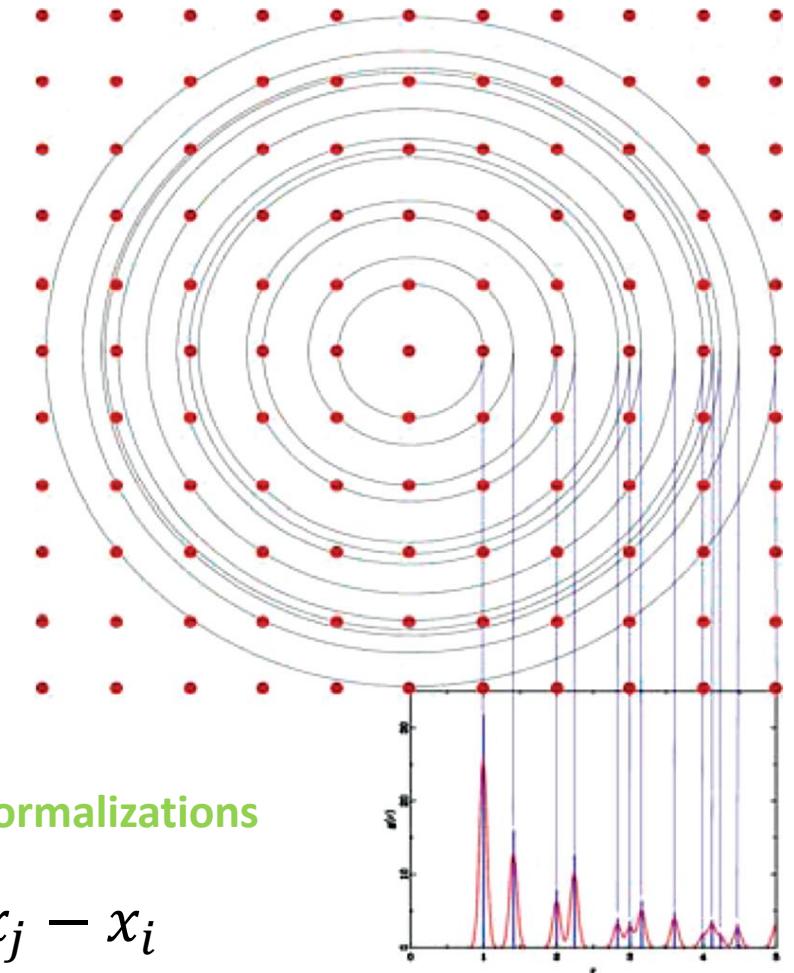
Corrections, approximations, normalizations

Debye equation  
(isolated atom approximation)

$$I(Q) = \sum_i^N \sum_j^N f_i^* f_j \frac{\sin(QR)}{R}$$

$$R = x_j - x_i$$

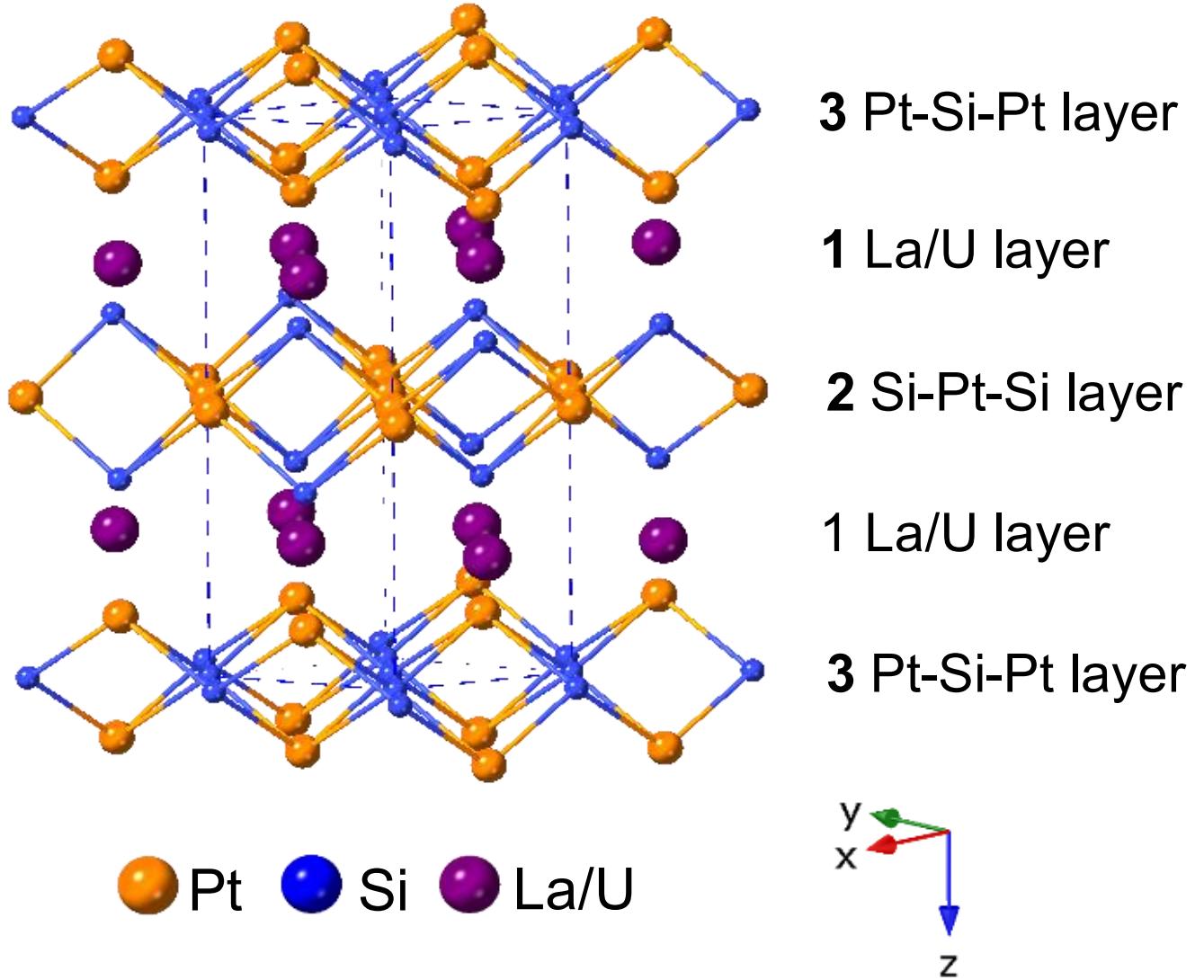
$$Q = \frac{4\pi \sin \theta}{\lambda}$$



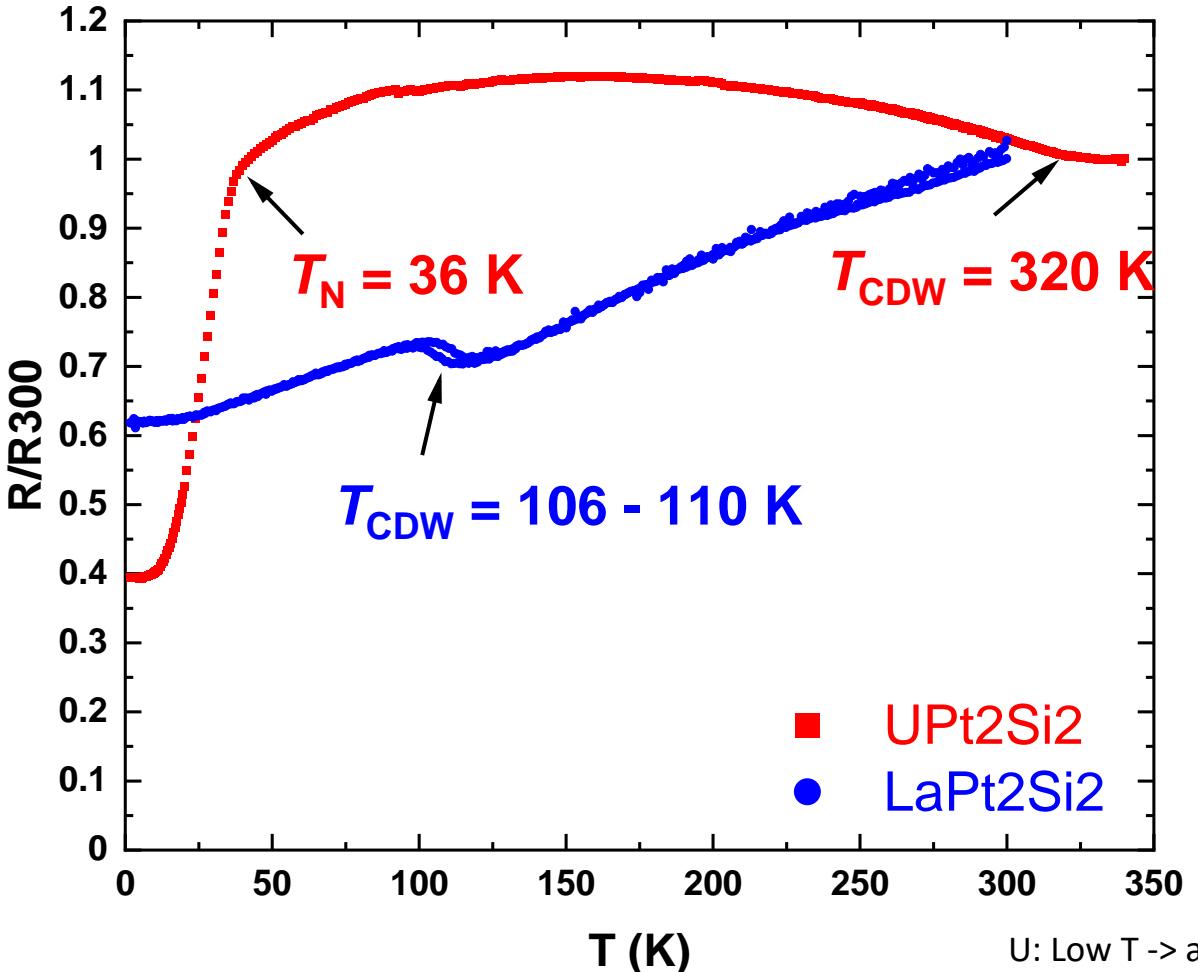
Petkov (2012) - PAIR DISTRIBUTION FUNCTIONS ANALYSIS

Crystal structure  
 $\text{CaBe}_2\text{Ge}_2$ -type  
 $P\frac{4}{n}mm$ ; tetragonal

$\text{AT}_2\text{X}_2$   
A ... electropositive metal  
T ... transition metal  
M... main group element.

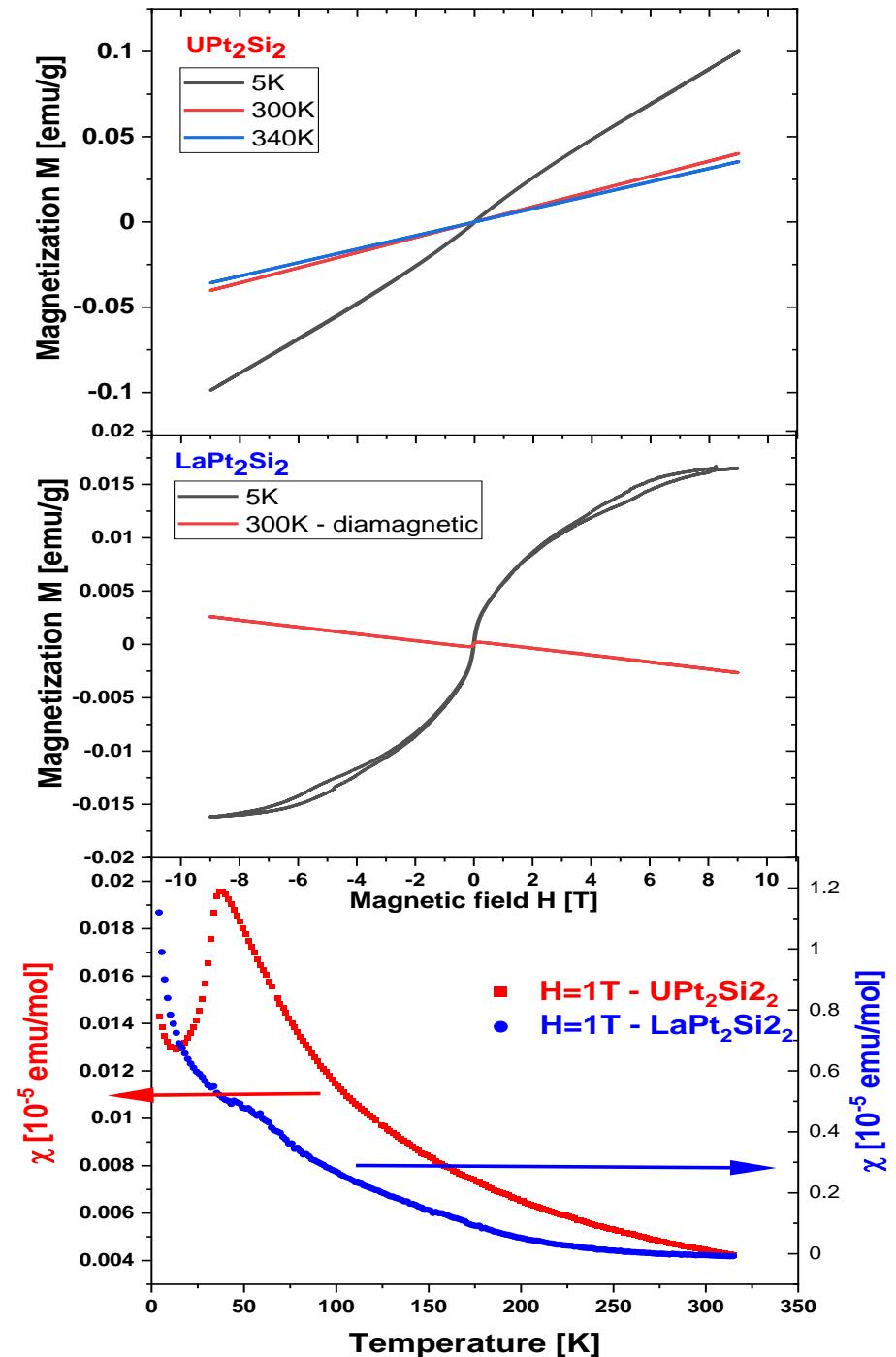


# Magnetic + transport properties



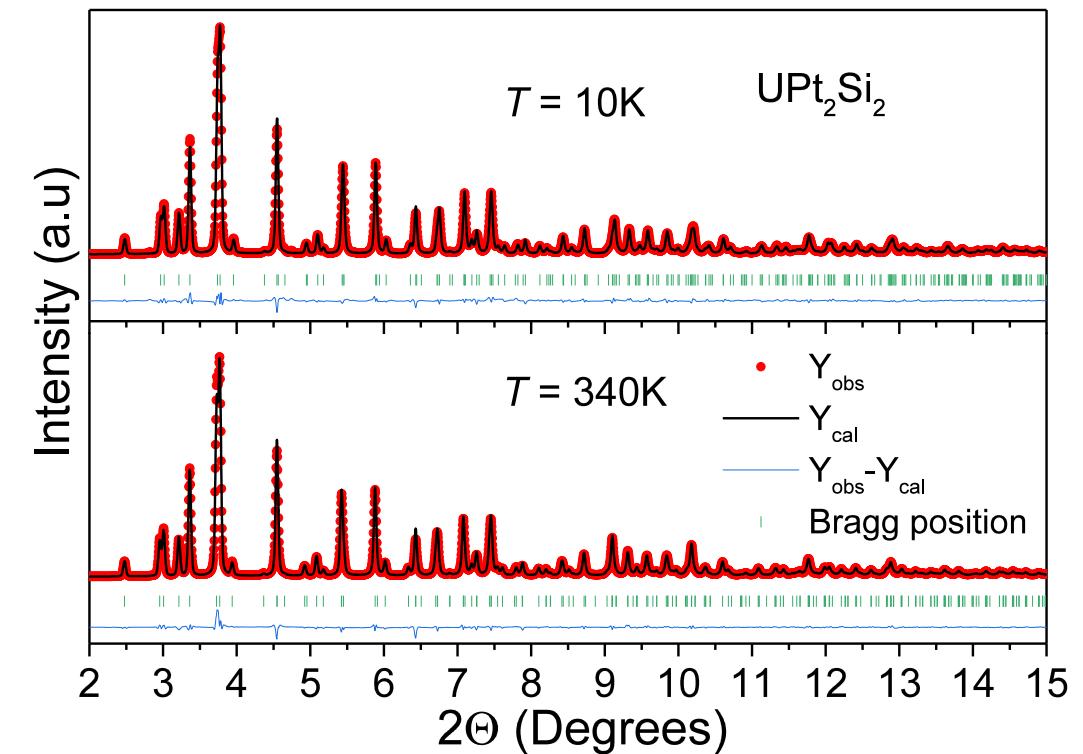
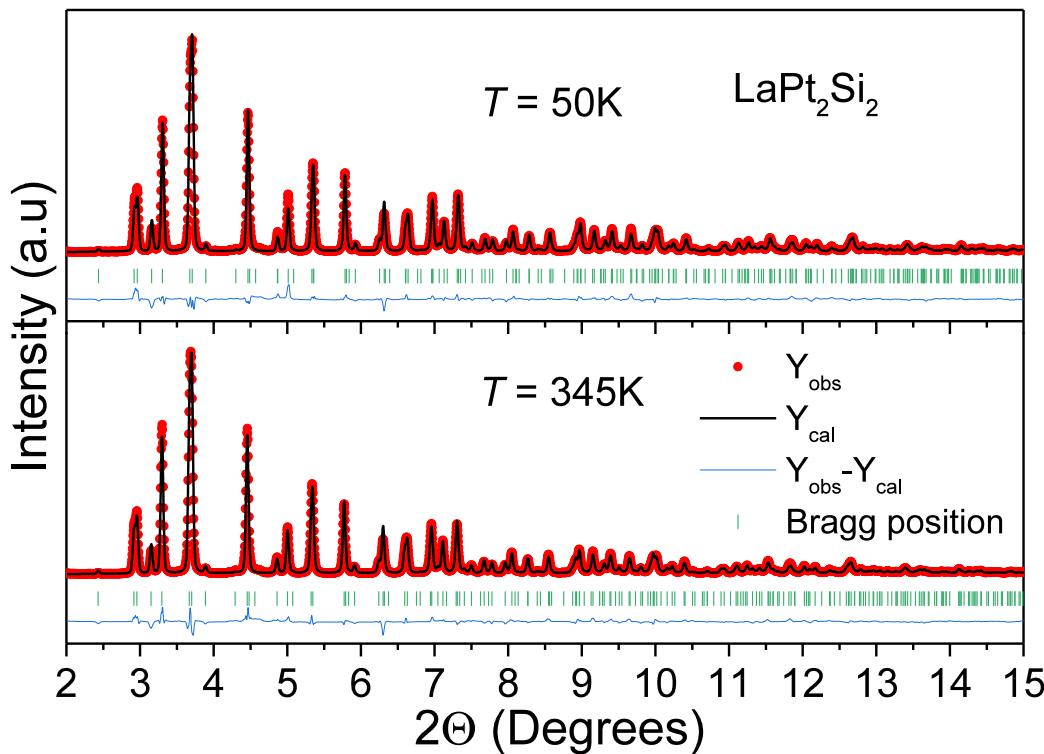
Residual electric resistivity  
at low T -> why?

U: Low T -> antiferromagnetic  
Then -> positive slope of M vs H ->  
paramagnetic  
La: low T -> Antiferromagnetic - H to  
5T at -> ferromagnetic behavior.  
RT Negative slope of M vs H ->  
diamagnetic

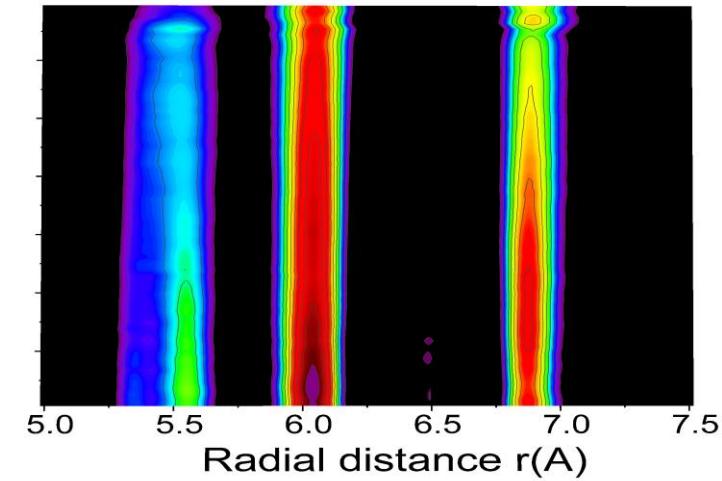
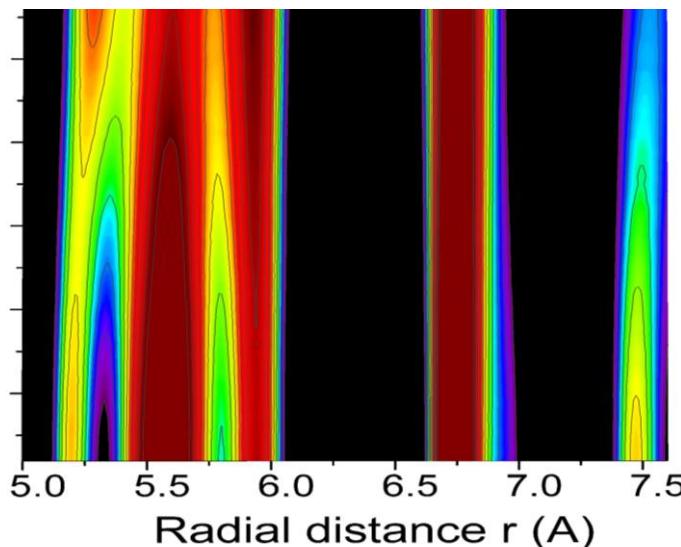
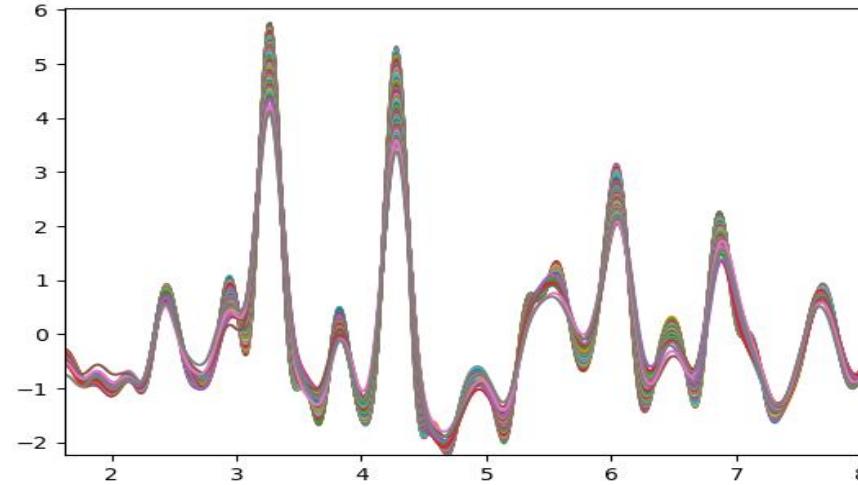
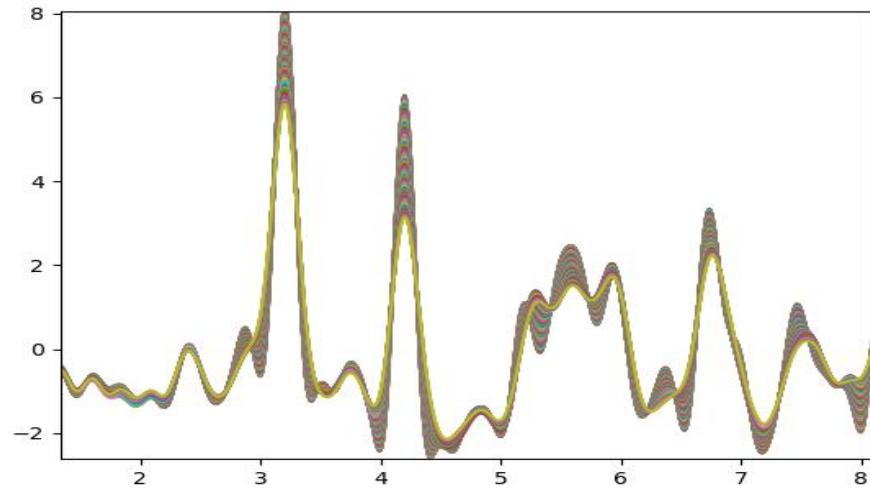


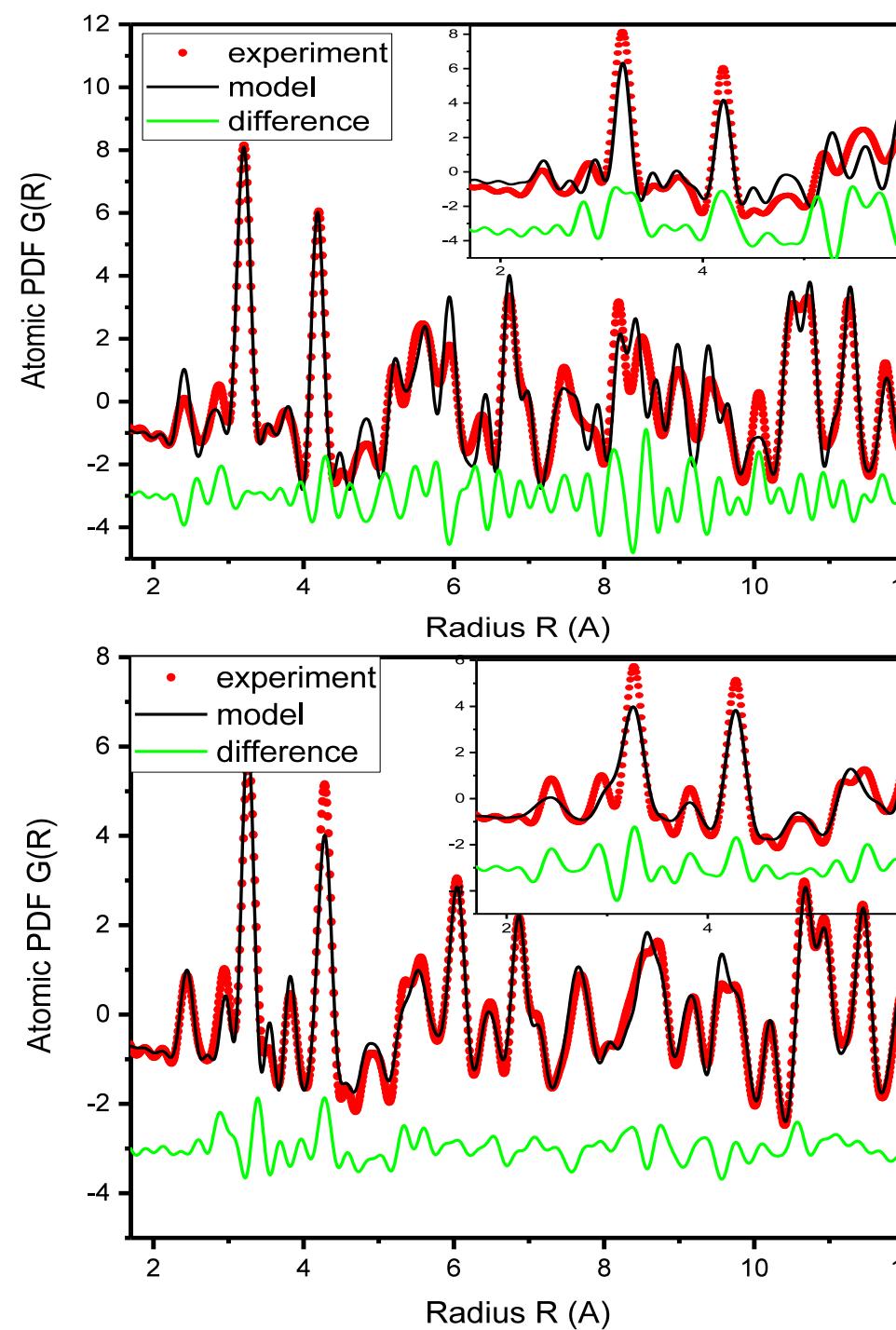
# Rietveld refinements

Average structure in space group  $P\frac{4}{n}mm$  explains data well



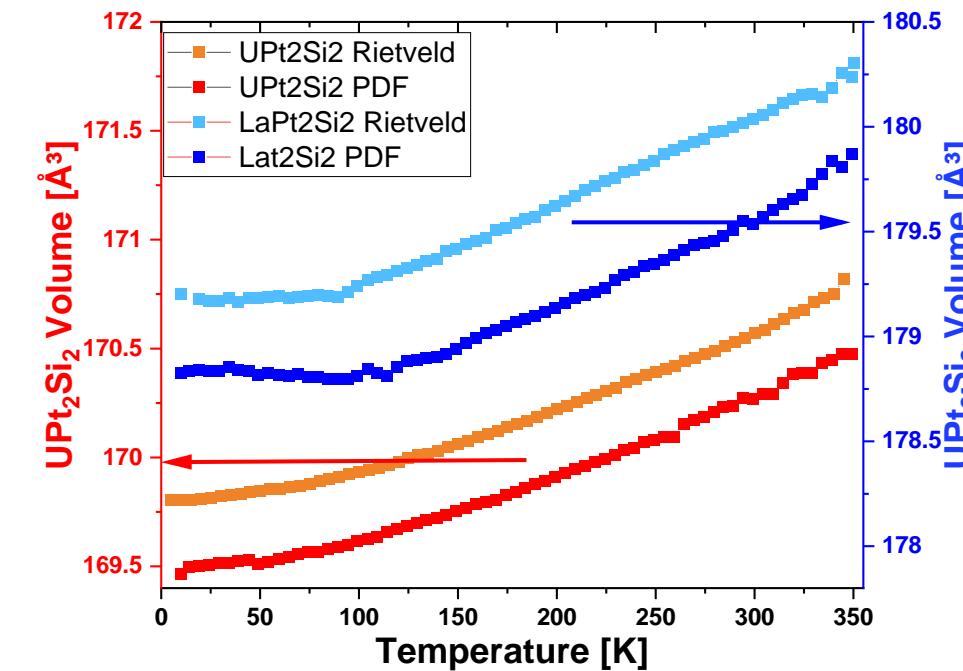
But PDF from powder XRD data show evolution with temperature, so the structures changes.





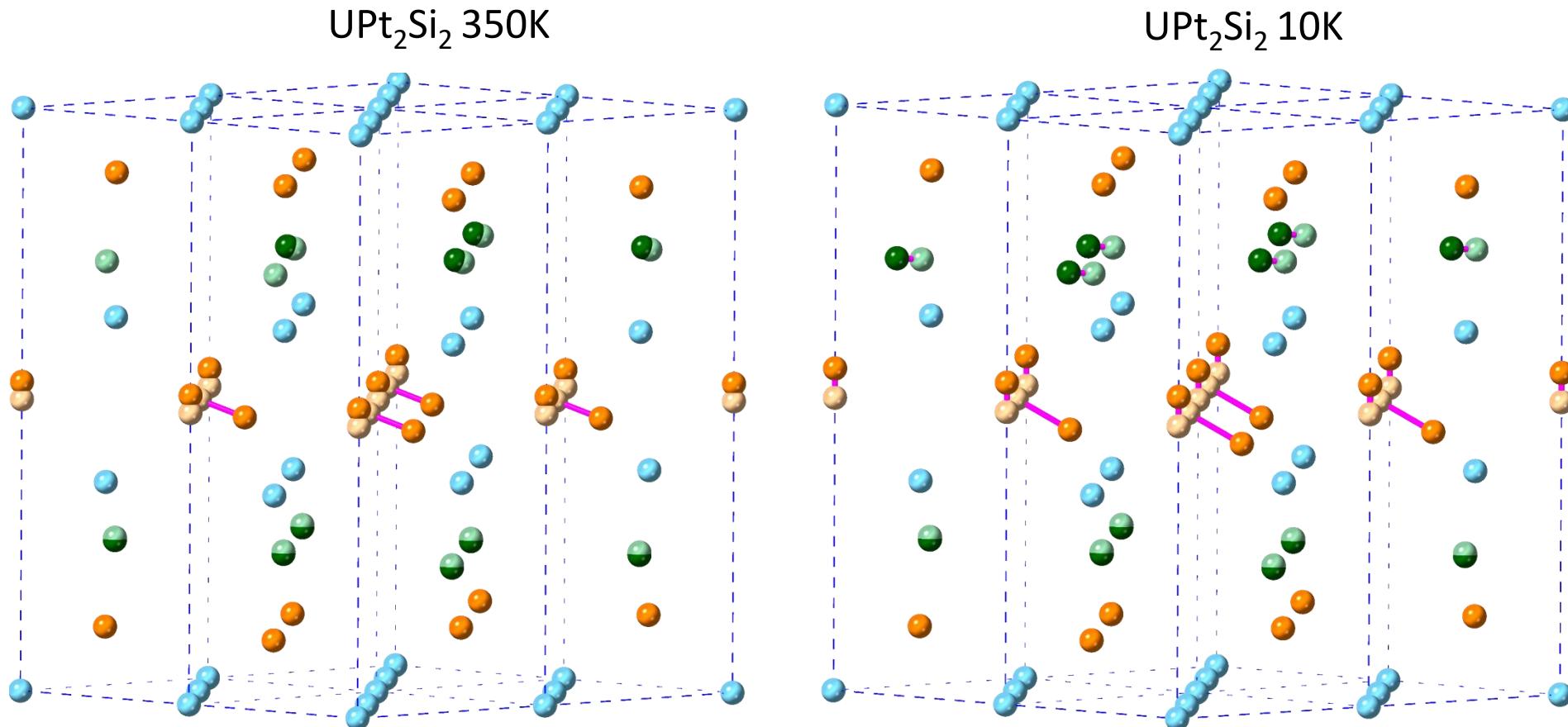
# Rietveld refinements and PDF (examples taken at 10K)

Models from Rietveld do not reproduce the local structure  
( $\text{UPt}_2\text{Si}_2$  needed to be refined in fully triclinic cell)



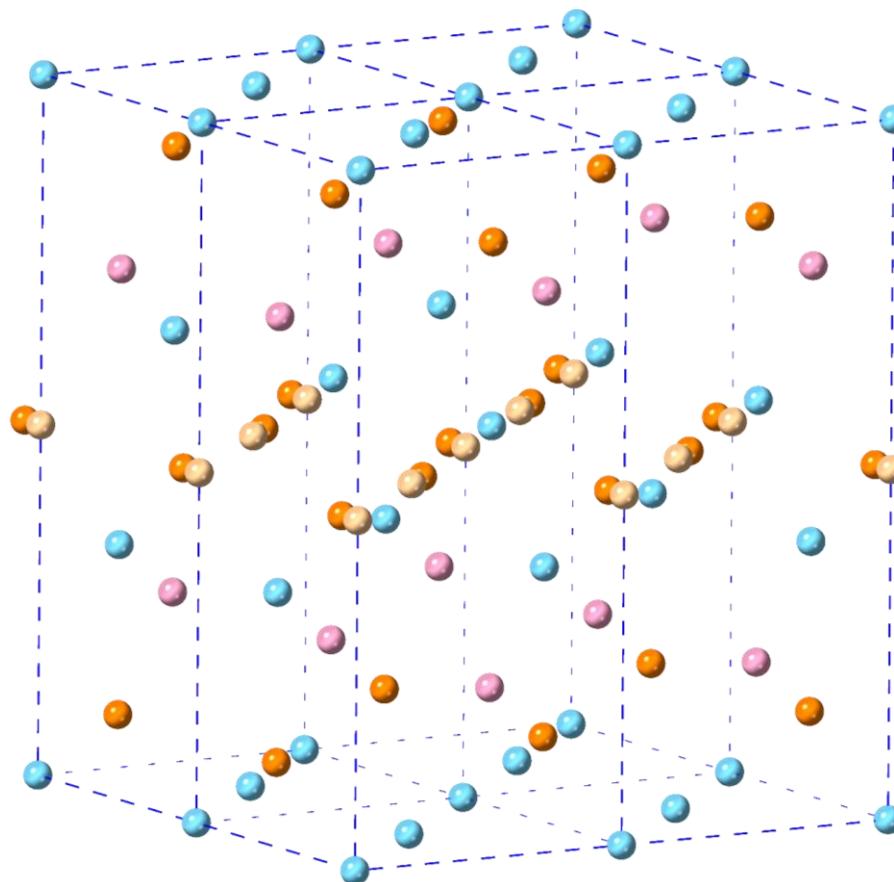
Resulting unit cell volumina from models from  
PDF refinements are smaller than from Rietveld  
-> locally denser packing than in average structure?

## Small box PDF-refinements: $\text{UPt}_2\text{Si}_2$

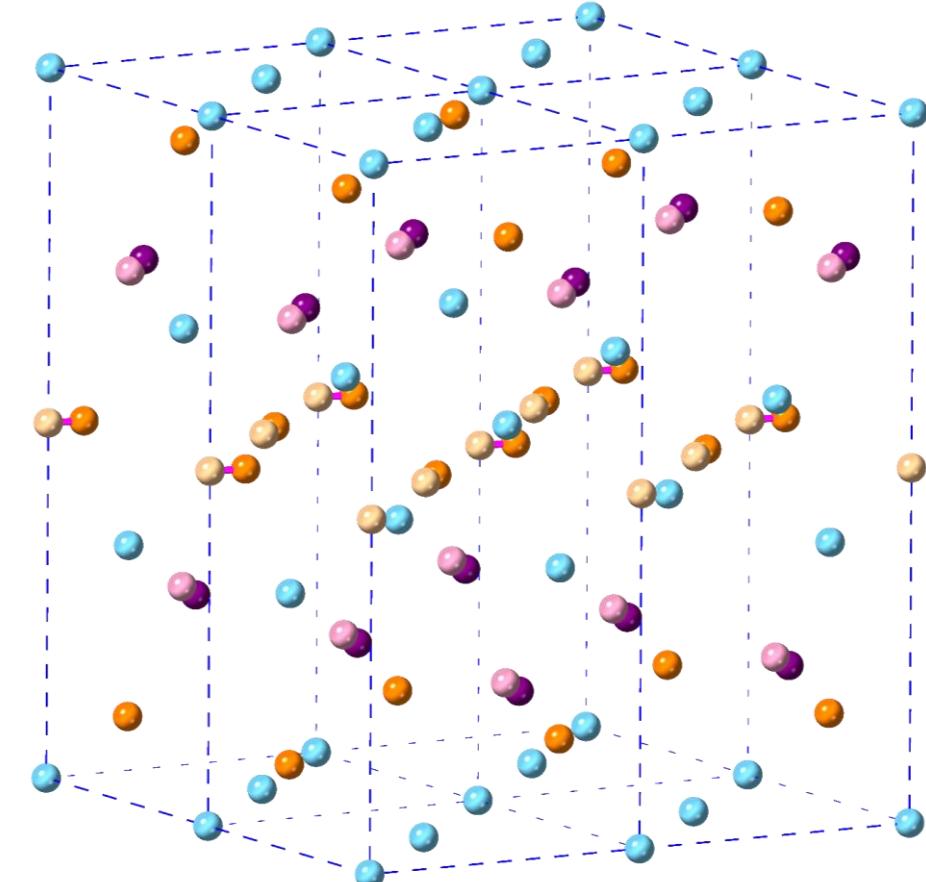


## Small box PDF-refinements: $\text{LaPt}_2\text{Si}_2$

$\text{LaPt}_2\text{Si}_2$  350K



$\text{LaPt}_2\text{Si}_2$  10K



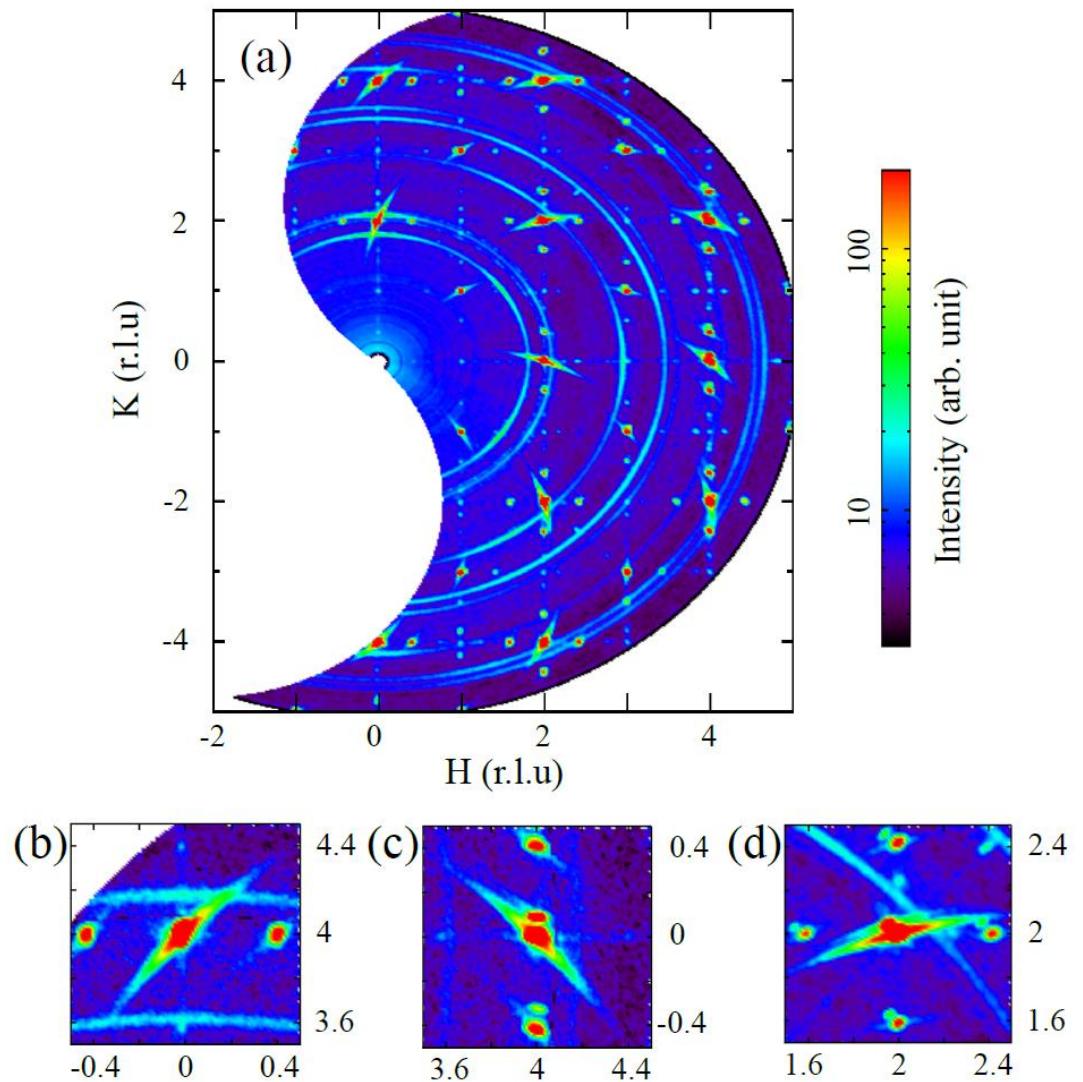
-> La atoms only get displaced at low temperatures – in contrast to  $\text{UPt}_2\text{Si}_2$

Literature states that there is no disorder in  $\text{UPt}_2\text{Si}_2$  (valid for  $\text{LaPt}_2\text{Si}_2$ ?).

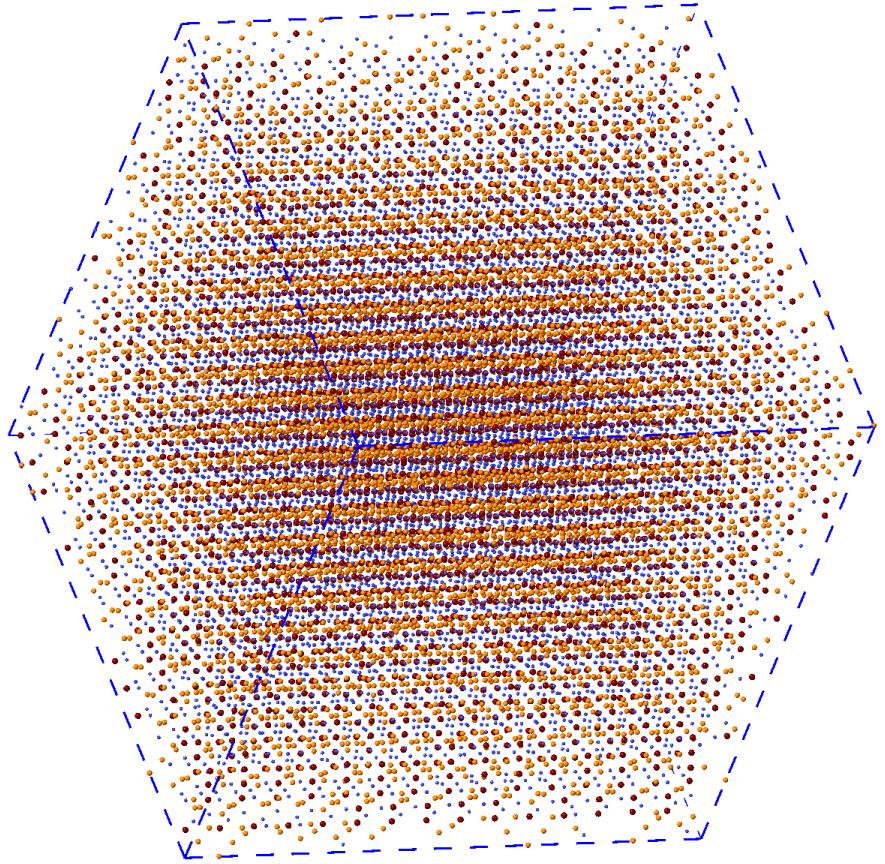
The appearance of the CDW is responsible for the heat capacity and electrical resistivity anomalies and resolves the controversy surrounding the putative “disorder”. (Lee, Prokeš et al (2020))

But there is strong diffuse scattering in their single crystal neutron scattering recordings (they tried to explain everything from the satellite reflections).

-> reverse Monte Carlo (RMC)  
refinements were carried out by us.



# RMC refinements for UPt<sub>2</sub>Si<sub>2</sub> and LaPt<sub>2</sub>Si<sub>2</sub> at 350/10 K.



15x15x7  
15750-atoms supercell

- Parameter space: the positions of atoms of the material under study put in a 'box', with the required density

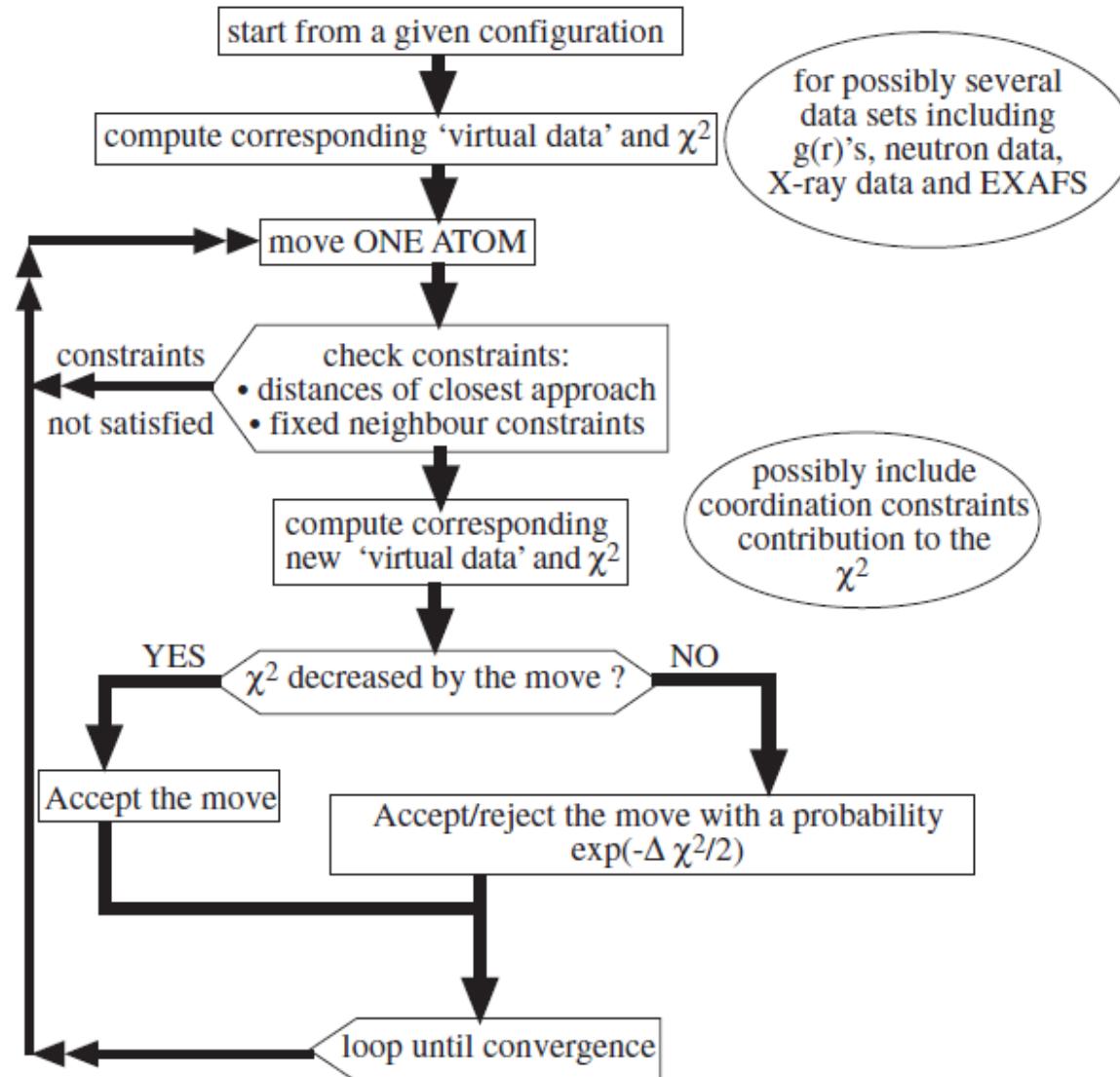
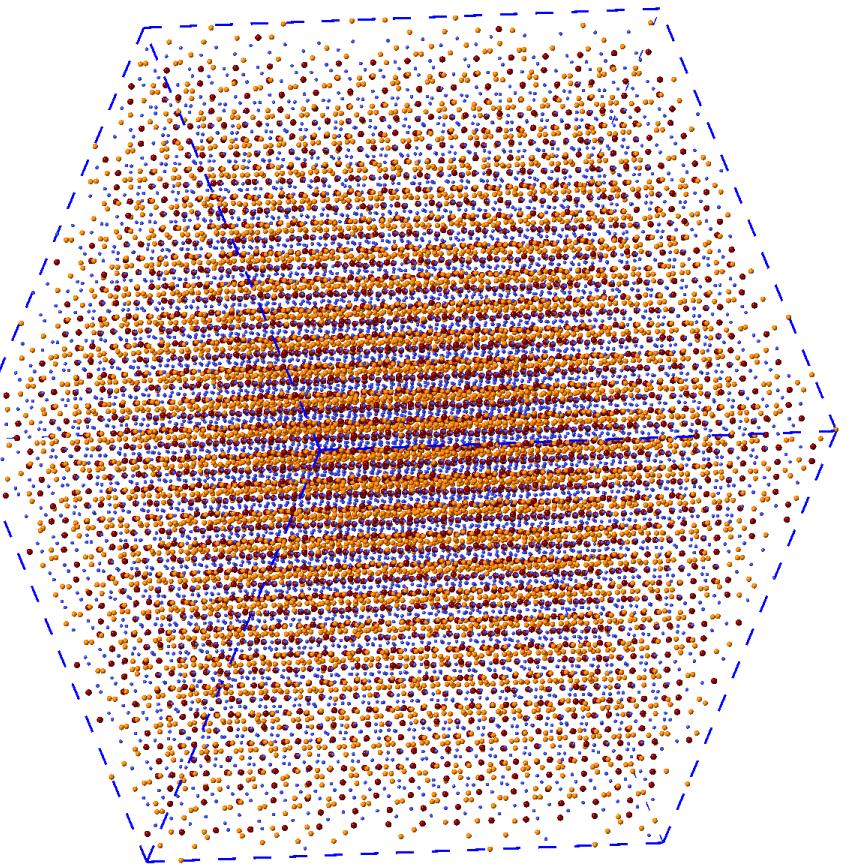


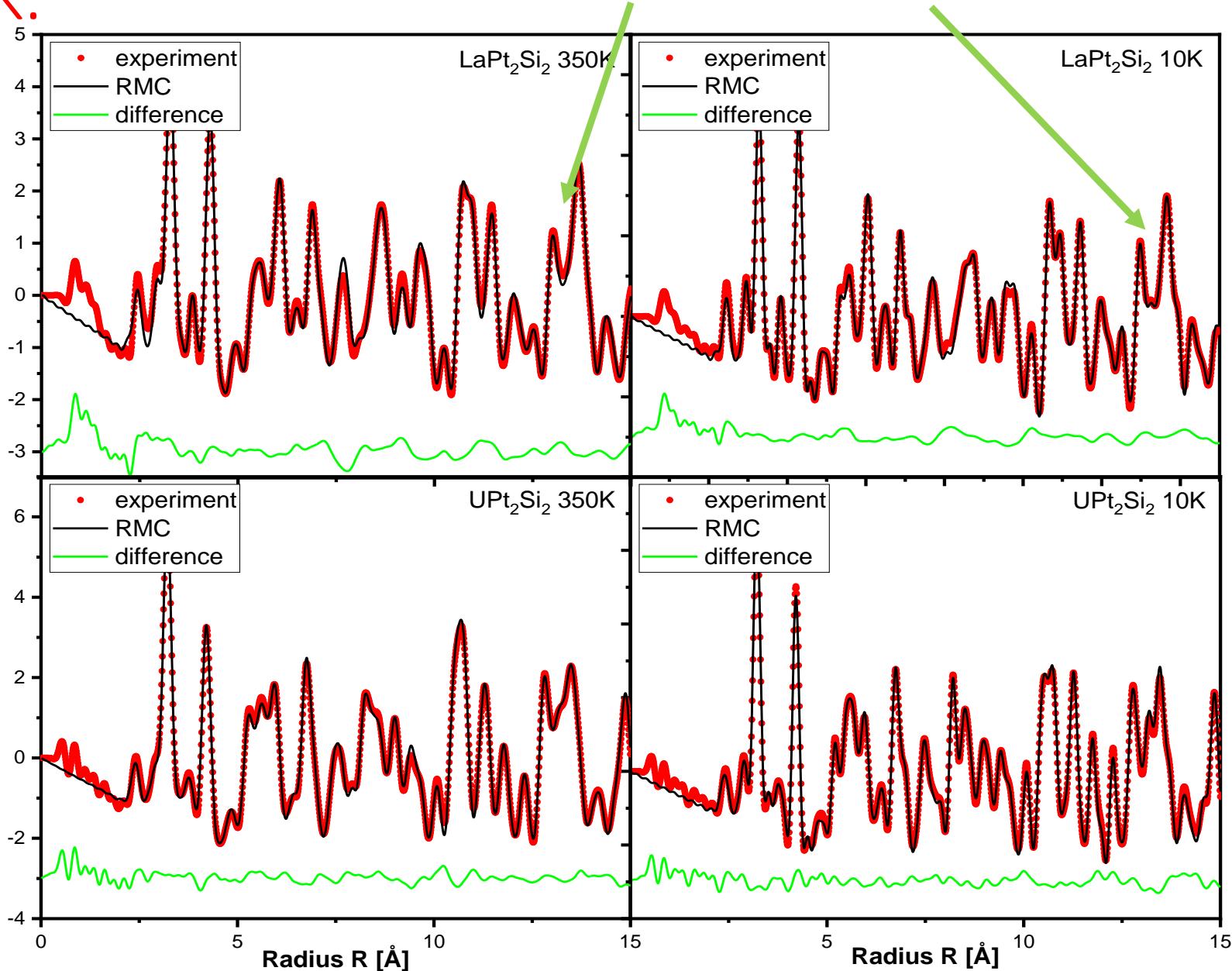
Figure 2. Flowchart of the 'standard' RMC algorithm.

# RMC refinements for $\text{UPt}_2\text{Si}_2$ and $\text{LaPt}_2\text{Si}_2$ at 350/10 K.

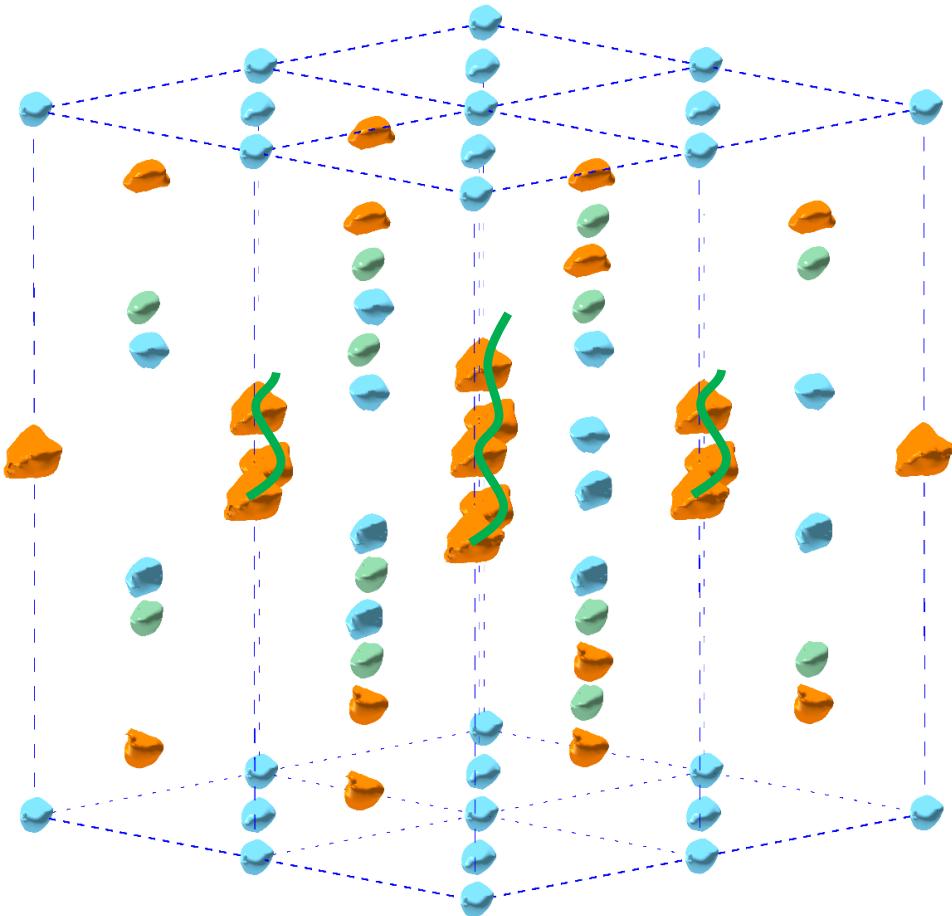


15x15x7  
15750-atoms supercell

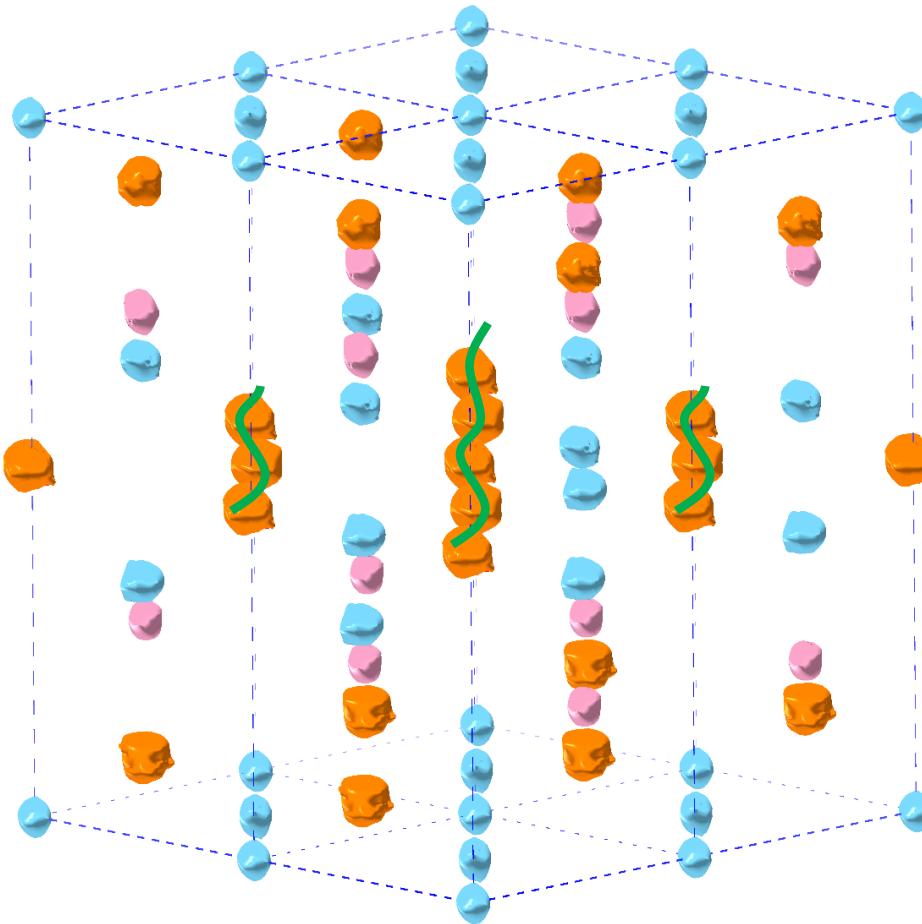
Subtle changes captured by simulation



# RMC – 15x15x7 collapsed to 1x1x1 unit cell

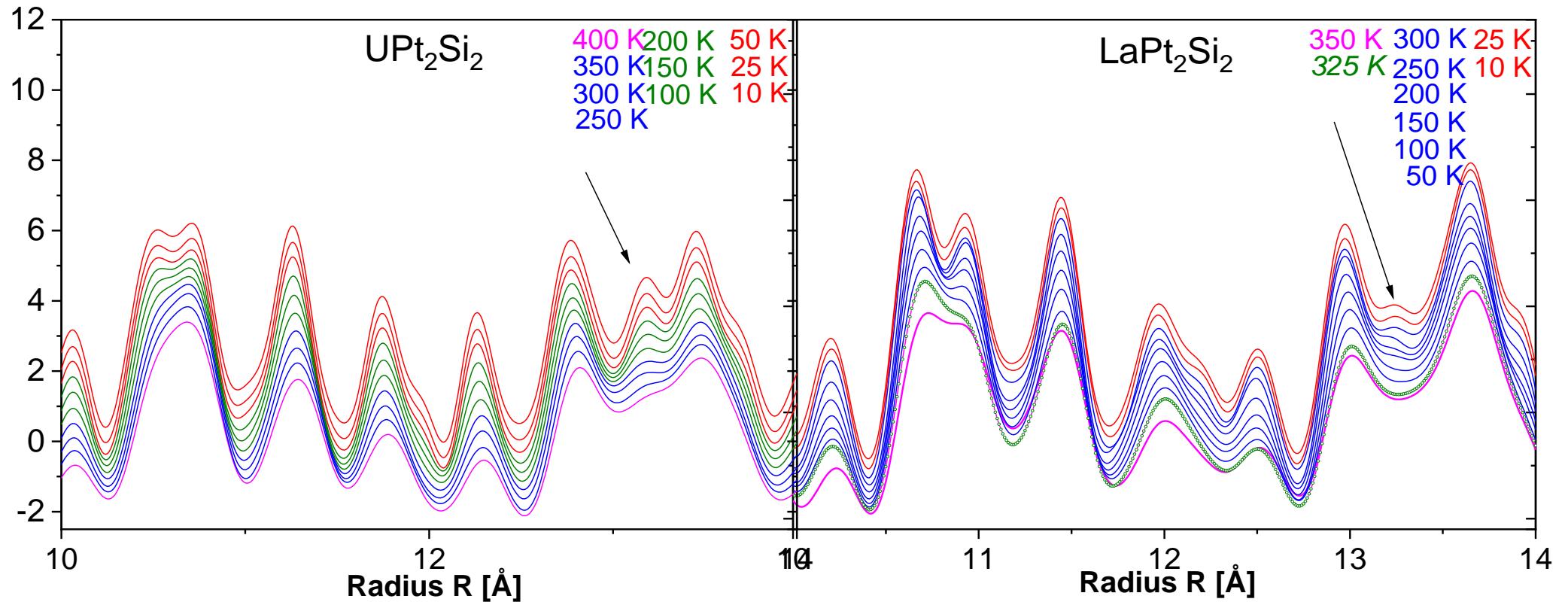


UPt<sub>2</sub>Si<sub>2</sub>



LaPt<sub>2</sub>Si<sub>2</sub>

Moreover:  
New distances are already visible without modelling.



# Summary

- Transport properties – residual electrical resistance arises from structural distortions.
- Properties are non-linear because structure is evolving continuously.
- Rietveld does not see atomic distortions.
- PDF shows interatomic correlations without any assumptions of crystallographic symmetry.
- We need modelling only to explain correlations revealed by PDF but not by Rietveld.
- Modelling shows that, locally, half of Pt-atoms are displaced in a correlated manner resembling a Charge Density Wave CDW.
- The displacements result in local compressive stress.
- In  $\text{UPt}_2\text{Si}_2$ , the CDW involves both U and Pt atoms and is already visible at RT.
- In  $\text{LaPt}_2\text{Si}_2$ , the CDW only involves Pt at RT and La at low T.

Thanks are due to DOE grant DE-SC0021973.