Charge density waves in the heavy fermion systems $LaPt_2Si_2$ and UPt_2Si_2 – similar but different

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$LaPt_2Si_2$ and UPt_2Si_2 – systems with "strange properties"

Using X-ray total scattering and reverse Monte Carlo simulations (RMC), we studied the lattice distortions accompanying the emergence of charge density waves (CDWs) in the heavy fermion systems $LaPt_2Si_2$ and UPt_2Si_2 over a temperature range extending from 10 K to 350 K, where their transport and magnetic properties exhibit a strong non-linear evolution. (**Fig. 1** and **Fig. 2**). Both systems are ternary compounds of the type AT_2X_2, where A is an electropositive metal, T is a transition metal, and M is a main group element, and crystalize in the layered $CaBe_2Ge_2$ type structure (space group $P\frac{4}{n}mm$; **Fig. 3**) thus differing from most AT_2X_2 compounds that are known to adopt a $ThCr_2Si_2$ type structure ($I\frac{4}{m}mm$).

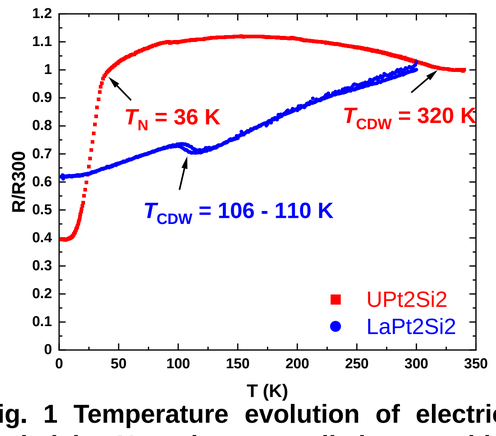


Fig. 1 Temperature evolution of electrical resistivity. Note the unusually large residual resistivity at T approaching zero K.

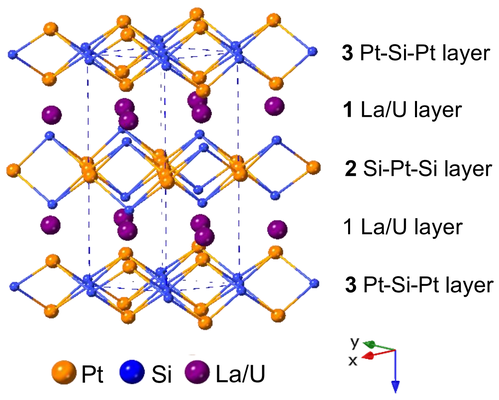


Fig. 3 $CaBe_2Ge_2$ structure type (space group $P\frac{4}{n}mm$) featuring a stack of 3 distinct layers.

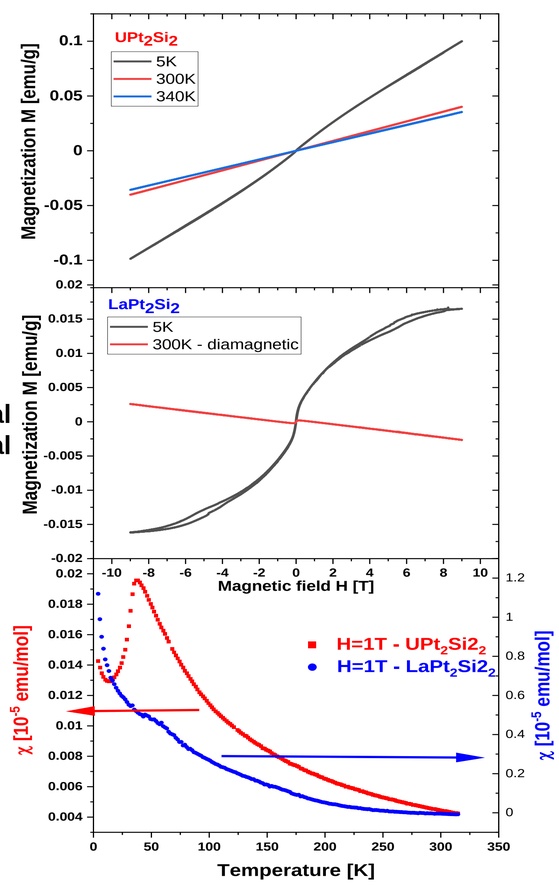
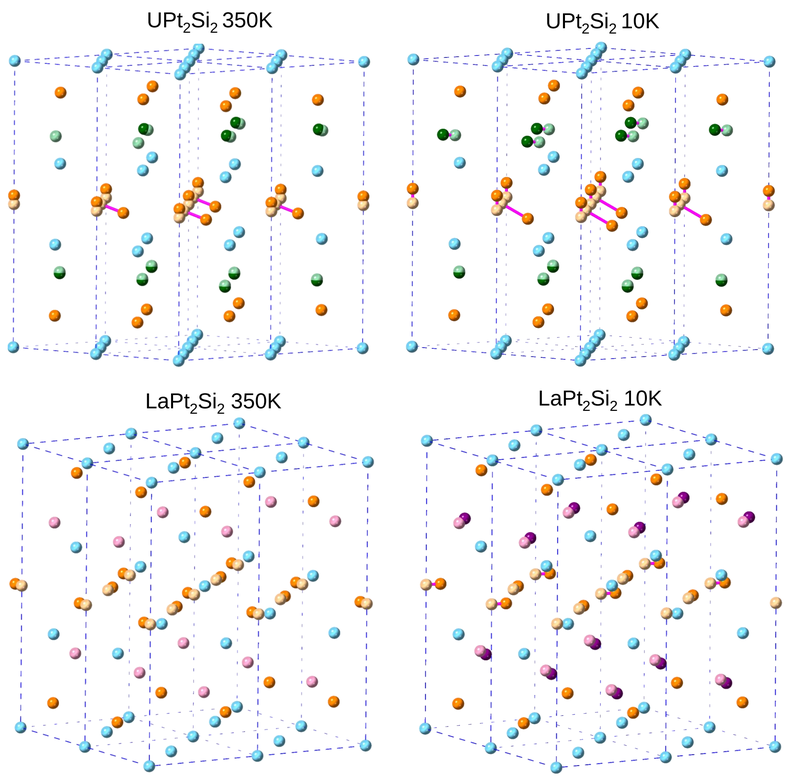


Fig. 2 Magnetization (top two panels) and susceptibility (bottom panel) for UPt_2Si_2 and $LaPt_2Si_2$.

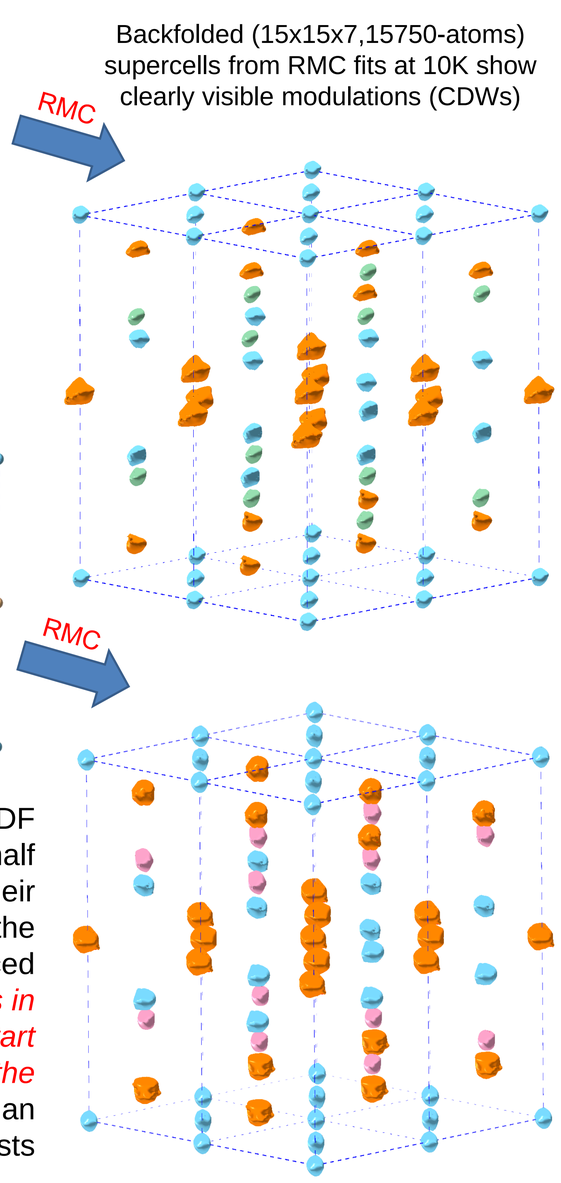
Small-box PDF-refinements, RMC and Interpretation

Small box PDF-refinements: what did we learn so far?



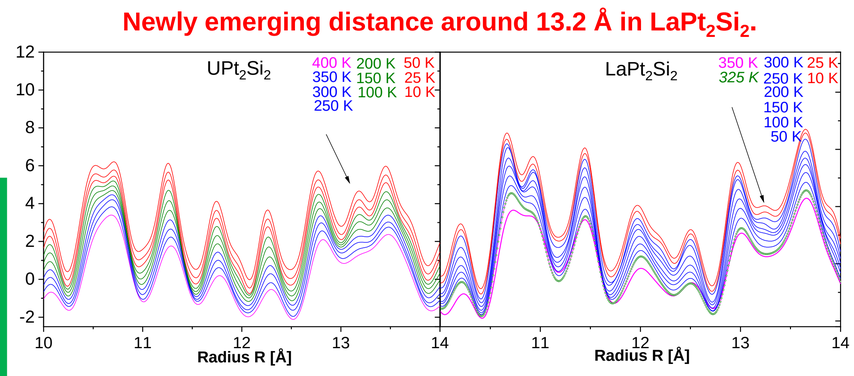
A detailed view from RMC.

Backfolded (15x15x7,15750-atoms) supercells from RMC fits at 10K show clearly visible modulations (CDWs)

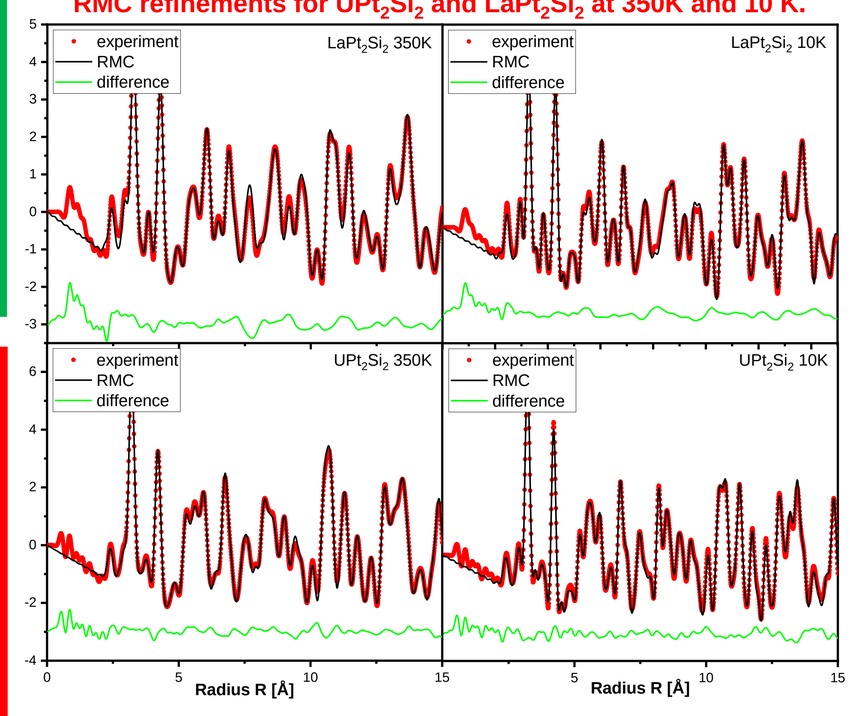


Structure models resulting from small-box PDF refinements: Particularly notable is that in both systems half of Pt (T-type; orange) atoms are displaced from their expected positions in the undistorted lattice. In addition, the A-type (U, green) atoms in UPt_2Si_2 appear displaced already at 350 K. *By contrast, the A-type (La, pink) atoms in $LaPt_2Si_2$ do not appear displaced at high T. They only start to move away from their average positions when the temperature drops below 150 K.* The emergence of an interatomic distance at ~13.2 Å in $LaPt_2Si_2$ that exists already at high T in UPt_2Si_2 is shown below.

Newly emerging distance around 13.2 Å in $LaPt_2Si_2$.



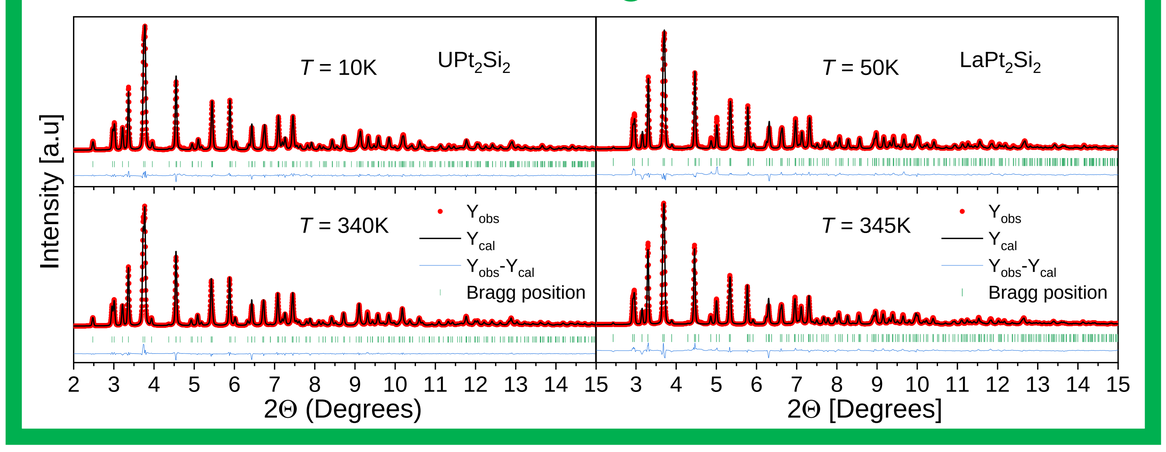
RMC refinements for UPt_2Si_2 and $LaPt_2Si_2$ at 350K and 10 K.

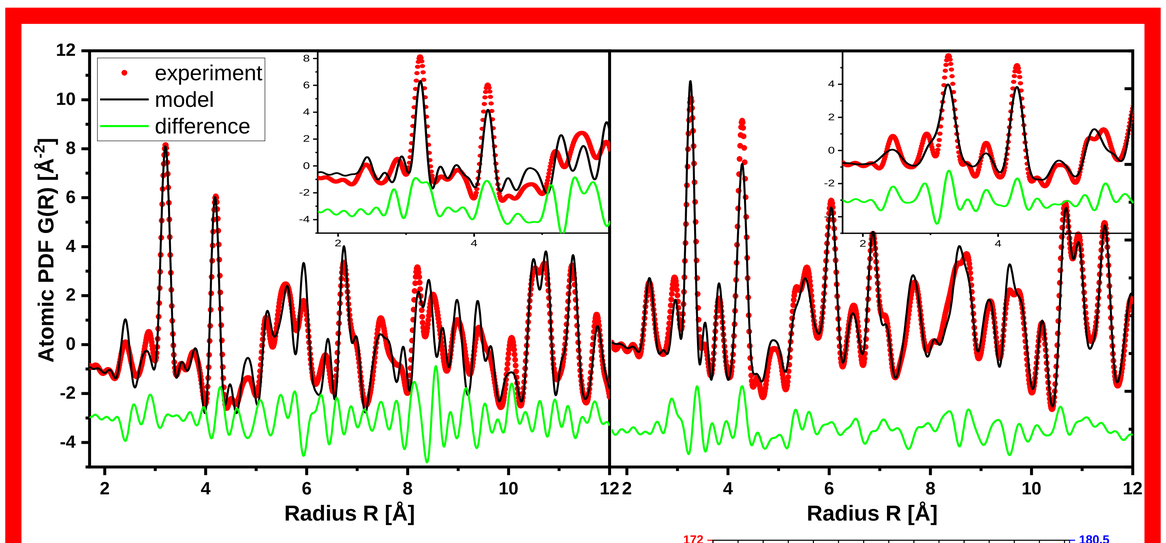


To get a detailed picture of the average atomic displacements, Reverse Monte Carlo (RMC) calculations were performed, where atoms in a simulation box are moved randomly as to make a plausible structure model reproduce experimental PDF data as close as possible.

RMC fits are shown to the left. Back folded unit cells for the 10K datasets are shown above, next to the models from small box PDF refinements. In line with their findings, the distribution of atoms about their average positions in the undistorted lattice was found to be markedly anisotropic, consistent with lattice distortions observed with CDWs. The distortions are present already at room temperature and increase in magnitude when it is decreased. They also appear dependent on the nature of A-type atoms (U, La) or chemical bonding.

Rietveld refinements look good …

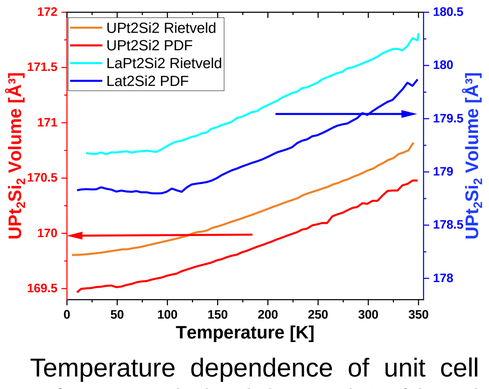




Rietveld models, however, do not reproduce the local structure accurately

To improve the PDF fits, the local crystal symmetry needs to be reduced such that atoms are allowed to displace from their positions in the undistorted tetragonal lattice

Values for lattice parameters and unit cell volume resulting from PDF fits are smaller than Rietveld results. The result implies that atoms in both systems are more closely packed locally in comparison to the average tetragonal crystal.



Temperature dependence of unit cell volume as derived from Rietveld and PDF fits

Conclusions

We showed by PDF analysis that periodic atomic displacements breaking the average crystal symmetry appear in both $LaPt_2Si_2$ and UPt_2Si_2, In particular, atoms are packed closer locally than in the average structure, suggesting that this "repacking" is behind the emergence of CDWs and non-linearity of physical properties. There also exists substantial local disorder that is different in both compounds, while their average structure type is the same. The modulation of Pt (T-type atoms) persists to high temperatures and is masked by thermal motion, while the displacement of the A-type atoms (U, La) is different.

Our study is an example for methods beyond "traditional crystallography" being capable of accounting for fine structural details reflected by the diffuse component of XRD patterns. Because the structure we study is complex and small box modelling was unable to capture well the "fine" details, large-scale RMC modelling was performed.

Information about used software and parameters will be given in subsequent publications. Thanks are due to DOE grant DE-SC0021973. Support for travelling was received by TU WIEN x-ray center

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