



	Experiment title: Using phonons to probe the hidden order in URu ₂ Si ₂	Experiment number: HC-1935
Beamline: ID28	Date of experiment: from: 29/4/15 to: 5/5/15	Date of report:
Shifts: 18	Local contact(s): Thomas Forrest	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): Helen Walker * - ISIS Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Chilton, Oxon, OX11 0QX, GB Russell Ewings * - ISIS Facility, STFC Rutherford Appleton Laboratory, Harwell Science and Innovation Campus, Chilton, Oxon, OX11 0QX, GB Dai Aoki – CEA Grenoble – INAC SPSMS, 17 rue des Martyrs, FR – 38054 Grenoble Cedex 9		

Report:

URu₂Si₂ is one of the most actively studied materials exhibiting unconventional behaviour due to electronic correlations [1]. The central mystery of this compound is the nature of the so-called 'hidden order' (HO) state, which sets in below 17.5 K. This transition is characterised by an enormous change in entropy ($\sim 0.2R \ln 2$). A small 'parasitic' antiferromagnetic order sets in in the HO phase, but its ordered moment is far too small ($\sim 0.02\mu_B / U$) to explain the change in entropy. The nature of the order parameter is still uncertain, despite intensive theoretical [1,2] and experimental [3-8] efforts. In addition, URu₂Si₂ superconducts below 1.2 K, and can also be driven into an antiferromagnetic state with a comparatively large moment of $0.4\mu_B / U$ with the application of a modest pressure of ~ 0.5 GPa. This would seem to indicate that URu₂Si₂ exists in a delicately balanced state due to strong electronic correlations.

Very recently a small orthorhombic structural distortion has been observed on cooling into the HO phase using synchrotron x-ray diffraction [9]. However, this effect has only been seen in ultra-clean samples (as defined by high residual resistivity ratio, RRR), and not in "dirty" samples. One curious aspect of this is that the large characteristic entropy change described above is *not* limited to such ultra-clean samples. In recent years (again, with ultra-clean samples) much effort has been devoted to understanding the electronic structure of URu₂Si₂, in particular the precise details of the Fermi surface (FS). Recent advances in ARPES techniques [3] have, to an extent, resolved contradictory interpretations of band structure measurements. FS hotspots of enhanced quasi-particle density are revealed together with a reconstruction of the FS on entering the HO state, characterised by FS nesting.

Phonons are highly sensitive to the band structure, so we would expect to observe subtle modifications in the phonon spectra accompanying changes in the FS and/or structural distortions. A generic example of the former would be a Kohn anomaly in the phonon dispersion at a nesting wavevector. Surprisingly, there had at the time of our proposal never been a systematic study of the phonons in URu₂Si₂, with only a single inelastic neutron scattering (INS) study [6] over a limited energy and **Q** range, and some Raman spectroscopy measurements [7]. The main finding in ref. 6 is that in the HO phase there is a longitudinal optic mode that

softens dramatically along the $(\zeta, \zeta, 0)$ trajectory, and that this mode appears to be absent well above the HO transition temperature. This is ascribed to an instability of the FS towards breaking of the body-centred tetragonal symmetry in the HO phase.

The aim of our proposal was to perform measurements of the phonons in URu_2Si_2 using a high purity (small, hence not accessible to neutrons) sample. We compared our measurements with neutron data we took on a lower purity (large) sample, e.g. fig. 1, which had the additional problem of a significant magnetic

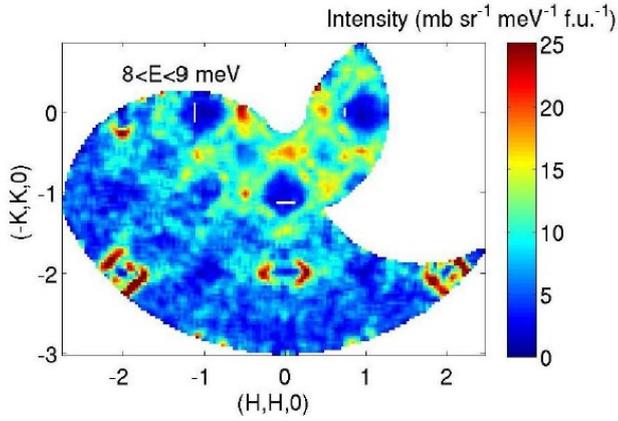


Figure 1: sample time-of-flight neutron data showing the magnetic and phonon contributions to $S(Q, E)$

contribution to the scattering which made analysis of the phonons more difficult. Shortly before the experiment we became aware of references [10] and [11], both of which reported much more comprehensive measurements of the phonons in URu_2Si_2 , including measurements using IXS, albeit on an offcut of a lower purity large sample. Using ID28 we were able to map out the phonon dispersion in several Brillouin zones along all of the major symmetry directions. We found that our measurements were fully consistent with the published data, indicating that sample purity has little bearing on the lattice dynamics in URu_2Si_2 . As was found in refs. [10] and [11], we observed no change in the phonon dispersion on cooling from the paramagnetic to the hidden order phase. Sample data taken in the HO and PM are shown in fig. 2.

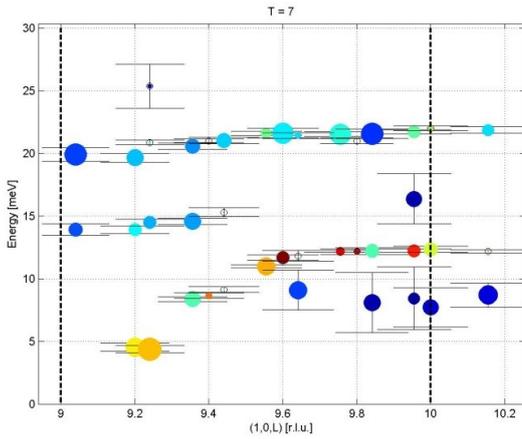


Figure 3a: sample dispersion along the $(0,0,1)$ direction measured on ID28 at 7 K. Marker colour indicates peak intensity and marker size peak width

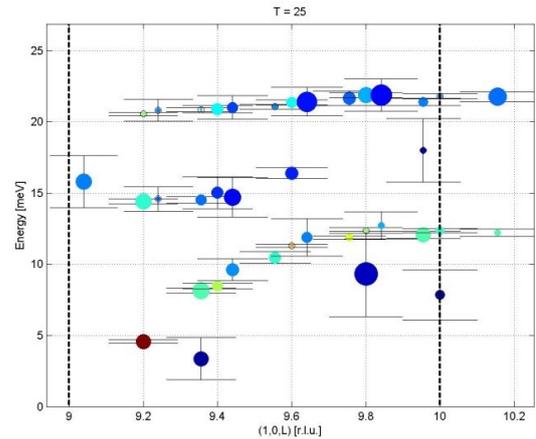


Figure 2b: as figure 2a, measured at 25 K

References

- [1] J. A. Mydosh and P. M. Oppeneer, Rev. Mod. Phys **83**, 1301 (2011).
- [2] P. Chandra *et al*, Nature **493**, 621 (2013); P. M. Oppeneer *et al*, Phys. Rev. B **82**, 205103 (2010); K. Haule *et al*, Nature Phys. **5**, 796 (2009).
- [3] J. -Q. Meng *et al*, Phys. Rev. Lett. **111**, 127002 (2013).
- [4] C. R. Wiebe *et al*, Nat. Phys. **3**, 96 (2007).
- [5] E. Ressouche *et al*, Phys. Rev. Lett. **109**, 067201 (2012).
- [6] N. P. Butch *et al*, arXiv:1212.6238 (2012).
- [7] J. Buhot *et al*, J. Kor. Phys. Soc. **10**, 1427 (2013); D. Lampakis *et al*, Physica B **378**, 578 (2006).
- [8] S. J. Clark *et al*, Zeit. Krist. **220**, 567 (2005); K. Refson *et al*, Phys. Rev. B **73**, 155114 (2006).
- [9] S. Tonegawa *et al*, Nat. Comms **5**, 4188 (2014).
- [10] N. Butch *et al*, Phys. Rev. B **91**, 035128 (2015).
- [11] J. Buhot *et al*, Phys. Rev. B **91**, 035129 (2015).