Valence Fluctuations in PuCoGa5

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Introduction

There is a long-standing problem in condensed mater physics of how to treat electrons that are well described neither as itinerant nor localized. This problem especially relevant in plutonium and its compounds, where the outermost 5f electrons straddle the boundary between itinerant and localized behaviour.

Elastic moduli, which are thermodynamic quantities, have long been used to differentiate different regimes of mixed-valence behavior. We use resonant ultrasound spectroscopy (RUS) to measure the temperature dependence of all six elastic moduli in PuCoGa5, and are able to relate the observed ultrasonic anomalies to valence fluctuations.

Experimental

A single crystal of flux-grown PuCoGa5 was polished to a cuboid of dimensions 2.208x2.204x0.641 mm. The RUS apparatus consists of two parallel transducers, one fixed and one on a free pivot, held in a low-vibration assembly and placed in a 4He flow cryostat. At each measurement temperature one of the transducers is driven from 200 kHz to 2 MHz, and power is transmitted to the other transducer at each mechanical resonance of the sample (65 resonances were recorded in this experiment). These resonance positions are then decomposed into the elastic moduli, and this procedure repeated from 300 K down to 12 K.

Results and Discussion

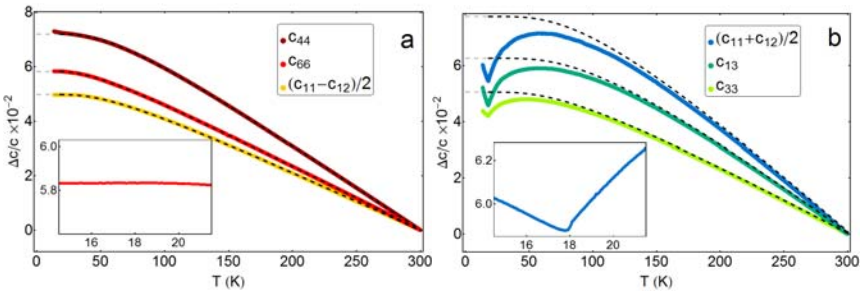

Figure 1: Elastic moduli of PuCoGa5 from 300 K to 12 K. a) The three shear moduli, corresponding to volume-preserving strains. No feature is seen at Tc = 18.5 K. b) The three compressional moduli, corresponding to volume-changing strains. Unlike the typical behavior of the shear, these moduli begin to soften well above Tc, and this behavior is truncated by superconductivity.

We find that the three shear moduli (Fig. 1a) show behaviour typical of most metals, while the compressional moduli soften well above Tc. Furthermore this softening diverges from the background as $1/T-T_0$, with T_0~9 K, and the appearance of superconductivity truncates this softening.

The three compressional moduli are all of scalar A1g symmetry, and this constrains the fluctuations responsible for their softening to also be A1g and time-reversal even (non-magnetic). Softening in this symmetry channel is reminiscent of other strongly mixed-valence materials, such as $Sm_{1-x}Y_xS$[1], except that in the case of PuCoGa5 superconductivity intervenes before the valence transition. This leads us to believe that the valence fluctuations have a strong connection the Fermi surface, and therefore have long-range spatial correlations.

Conclusions

We have used resonant ultrasound spectroscopy to measure fluctuations associated with the Fermi surface of PuCoGa5, including their symmetry. This provides the first direct experimental evidence for valence fluctuations in PuCoGa5, and a possible pairing mechanism in PuCoGa5 that does not solely rely on spin fluctuations or phonons.

Acknowledgements

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References

[1] Mock, R. *et al.* PRL, **57,** 1040 (1986)