Experimental Report

Proposal: **7-02-148** **Council:** 4/2014

Title: Structural Quantum Critical Point in Lu(PtPd)2In

This proposal is a new proposal

Researh Area: Physics

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Samples: LuPt2In
LuPd2In
Lu(Pt0.45Pd0.55)2In

Instrument	Req. Days	All. Days	From	To
IN6	4	4	14/10/2014	18/10/2014
IN4	6	6	08/10/2014	14/10/2014

Abstract:

Quantum critical points (QCPs) are second order phase transitions that occur at T=0; they involve unusual critical fluctuations, which give rise to interesting phenomena such as unconventional superconductivity and non Fermi-liquid behaviour. So far QCPs have mostly been studied for magnetic phase transitions.
Recently, we have discovered that the phase diagram of the substitution series Lu(PtPd)2In shows a QCP of a structural phase transition. We observe superconductivity with the highest transition temperature at the QCP, suggesting a link between the unusual critical fluctuations and the appearance of superconductivity. We would like to investigate this relation further by measuring the phonon density of states by inelastic neutron scattering. The comparison of different samples of the alloying series would allow to study the development of soft phonons.

Structural Quantum Critical Point in $Lu(Pt_{1-x}Pd_x)_2In$

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Quantum critical points (QCPs) occur if second order phase transitions are suppressed to T=0. Close to zero temperature, quantum fluctuations become more relevant than thermal fluctuations, and therefore they strongly influence the critical behaviour. For magnetic transitions, this has been and continues to be extensively studied, the interest being driven by the large variety of unusual physical phenomena that are observed in the vicinity of QCPs, such as non Fermi-liquid behaviour or unconventional superconductivity [1,2]. To access a QCP, one needs to be able to tune the ordering temperature in a controlled manner, for example by pressure, magnetic field or element substitution. This has been done successfully for many antiferromagnetic compounds.

On the other hand, QCPs of *structural* transitions have been studied far less, even though they can also be expected to involve unusual fluctuations which give rise to interesting unconventional phases. Recently, Klintberg *et al.* reported on the observation of a superconducting phase related to a structural QCP in $(Sr,Ca)_3Ir_4Sn_{13}$ [3]. However, the maximum of the superconducting transition temperature T_C and the QCP do not occur at the same position in the phase diagram so that the relation between superconductivity and QCP is uncertain. We have now found that $Lu(Pt_{1-x}Pd_x)_2In$ presents a very good example of superconductivity related to a structural QCP.

While $LuPd_2In$ crystallises in a cubic Heusler structure [4] at all temperatures, we observed the formation of a cubic superstructure in $LuPt_2In$ below 490 K. The high-temperature phase has the same structure as $LuPd_2In$. We succeeded in tuning the transition temperature of this second-order transition by synthesising different compounds of the alloying series $Lu(Pt_{1-x}Pd_x)_2In$, reaching $T_{str} = 0$ at around 55 % Pd substitution. We also found that the samples become superconducting at $T_C \approx 1$ K, with a striking coincidence of minimum T_{str} and maximum T_C [5]. Furthermore, measurements of the specific heat of the substitution series indicate a softening of the phonon modes around the critical concentration. This gives further evidence that critical phonon modes related to the structural QCP are involved in the formation of the superconducting state.

The aim of our inelastic neutron scattering experiment was to perform a more direct measurement of the phonon softening. We choose three samples out of the series, pure $LuPt_2In$ and $LuPd_2In$ as well as the critical concentration $Lu(Pd_{0.55}Pt_{0.45})_2In$. Two different sample environments were necessary, a heating loop to access the transition temperature $T_{str} = 490$ K of $LuPt_2In$ and a ^{4}He cryostat to measure the low-temperature behaviour of $Lu(Pd_{0.55}Pt_{0.45})_2In$ and $LuPd_2In$. Due to the large absorption coefficients of Lu and In we chose flat circular aluminum cans with large diameter as sample holders; thus, the sample thickness for our 10 g powder samples was ≤ 1 mm.

Looking first at the diffractograms, we observe the appearance of superstructure peaks in the low-temperature phase of $LuPt_2In$, in both the IN4 and the IN6 data. The strongest one is at $Q = 2.42$ Å^{-1}; its intensity decreases gradually as the temperature is inceased and then vanishes around $T_{str} = 490$ K, as expected for a second-order transition (see also inset figure 1, right). These data are in agreement with x-ray data. Moreover, due to the good signal-to-background ratio in the relevant Q-range at IN6, the second-order nature of the transition can be seen more clearly in the neutron data than in our previous x-ray data.

To get an overview of the phonon density of states, we have measured time-of-flight spectra at $\lambda = 2.22$ Å at IN4. For the comparison of the three samples they were all measured at 320 K in the heating loop. In the left panel of figure 1, the measured

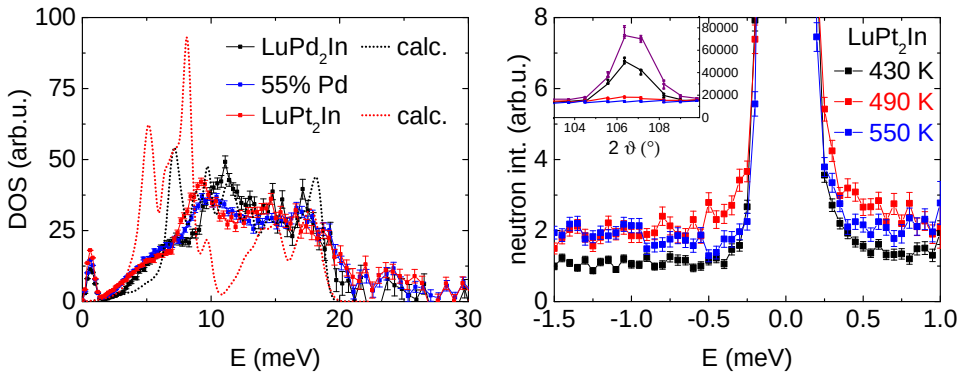

Figure 1: *Left:* The phonon density of states of $LuPt_2In$, $Lu(Pt_{0.45}Pd_{0.55})_2In$ and $LuPd_2In$, measured in the whole angular range available at IN4 at 2.22 Å. They are compared with calculated DOS of $LuPt_2In$ and $LuPd_2In$. The data are rescaled such that the area under all curves is the same. Below 1 meV the measured data are not meaningful. *Right:* Spectra of $LuPt_2In$ at Q =2.42 ± 0.05 $Å^{-1}$. Data were measured at IN6 at 430 K, 490 K and 550 K. The inset shows the respective section of the diffractogram, where also data at 320 K are added in purple.

phonon density of states is shown and compared with results from DFT calculations, which did not include the superstructure. This comparison suggests that all differences between the samples are simply due to the mass difference of Pd and Pt, while the superstructure formation plays a minor role in this energy range. This conclusion is supported by the observation that there is no temperature dependence in the phonon density of states for $\Delta E > 2$ meV for any of the samples, not even for $LuPt_2In$ around the transition temperature.

To gain better insight into the behaviour at low energies, the same samples (also with the same sample environments) were measured at $\lambda = 4.1$ Å at IN6. Again, phonons at generic Q-transfers seem temperature-independent. However, at $Q = 2.42$ $Å^{-1}$, where a superstructure Bragg peak appears in the low-temperature phase, we observe an enhanced inelastic signal in $LuPt_2In$. This is shown in the right panel of figure 1: Clearly, the intensity below $\Delta E < 1$ meV is stronger at $T_{str} = 490$ K than at 430 K and 550 K. In the spectra of $Lu(Pd_{0.55}Pt_{0.45})_2In$, where enhanced fluctuations are expected on approaching $T = 0$, we fail to observe a similar signal due to an unfavourable signal-to-background ratio: The signal is masked by strong cryostat scattering for positive energy transfers, and by the Bose-factor for negative energy transfers.

In summary, we have observed enhanced fluctuations associated with the second-order phase transition of $LuPt_2In$ with a characteristic energy scale of around 1 meV. The development of this signal in the subsitution series $Lu(Pt_{1-x}Pd_x)_2In$ remains a subject for further investigations.

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