Phenomena Near Quantum Criticality

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Experimental studies of quantum criticality quite often focus on compounds in the vicinity of a magnetic instability, while little is known about the behavior near a structural instability. We continued our work on heavy-fermion systems located close to magnetic quantum criticality. Our interest remained on the effect of geometrical frustration on the quantum-critical behavior. In addition, we started detailed investigations of compounds close to structural quantum criticality and followed the relevant phonon softening. In our studies we combined different spectroscopic measurements, namely elastic and inelastic neutron scattering, high-resolution inelastic x-ray scattering and muon spin rotation to gain new insight in to the intriguing properties of materials close to quantum criticality. We performed our experiments at different large-scale facilities and in collaboration with well-renowned scientists from research groups worldwide.

Continuous phase transitions occurring at absolute zero temperature, so-called quantum critical points (QCPs), remain in the focus of current solid-state physics. QCPs separate an ordered from a disordered ground state at $T = 0$. Usually magnetic QCPs are investigated where a magnetically ordered ground state can be tuned continuously to $T = 0$ as a function of an external control parameter. In recent years we mainly studied heavy-fermion compounds and focused on the influence of geometrical frustration on the quantum-critical behavior. We also looked at novel phases close to quantum criticality such as unconventional superconductivity. However, the concept of quantum criticality is not just limited to magnetic systems, but is also applicable to non-magnetic quantum criticality. In contrast to magnetic quantum criticality, remarkably little is known about systems exhibiting a structural quantum-critical point. As a consequence, we started to investigate the properties of structural quantum criticality. In particular, our measurements focused on the phonon softening near the structural quantum critical point in $Lu(Pt_{1-x}Pd_x)_2In$ [1, 2]. As a continuation of our previous studies on CePdAl we now focused on the influence of frustration on the quantum-critical behavior by performing muon spin rotation on pure and Ni-subtituted CePdAl when pressure and concentration tuning the QCP [3, 4]. We also studied the effect of frustration on the magnetism in $Ce_4Ni_6P_{17}$ [5] where clear signs of frustration are evidenced by an analysis of the magnetic entropy. In addition, we investigated the flux-line lattice in $CeCu_2Si_2$ [6], reviewed the important results on the unconventional superconductivity in $CeCu_2Si_2$ [7] and compared its spin resonance in the superconducting state with other heavy-fermion superconductors [8]. We mainly used different microscopic techniques, such as elastic and inelastic neutron scattering, high-resolution inelastic x-ray scattering and muon spin rotation, to address the above-mentioned issues.

Structural quantum criticality

Recently, we discovered the intermetallic alloying series $Lu(Pt_{1-x}Pd_x)_2In$ to be a realization of a structural or charge-density-wave (CDW) quantum critical point with the appearance of a superconducting dome in the vicinity of quantum criticality [9]. The second-order structural transition from a face-centered cubic Heusler structure to a body-centered cubic superstructure occurs in pure $LuPt_2In$ at $T_{CDW} = 490\,K$ and is linearly suppressed to zero temperature upon Pd substitution in $Lu(Pt_{1-x}Pd_x)_2In$ with a critical Pd concentration of $x_c \approx 0.6$ [9]. As shown in Figure 1, quantum criticality is accompanied by a strong enhancement of the superconducting transition temperature T_{sc}.

So far, the low-energy phonons, being at the origin of the quantum-critical behavior and the enhanced superconductivity in $Lu(Pt_{1-x}Pd_x)_2In$, have only been investigated indirectly. For instance, the phononic contribution β to the heat capacity at low temperatures displays a pronounced maximum near x_c [9]. Using the Bridgman technique we succeeded in growing large, high-quality

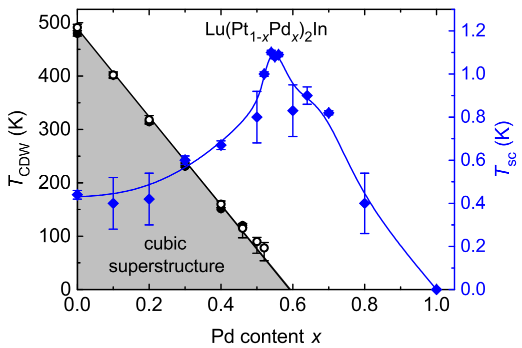

Fig. 1: Temperature – concentration (x, T) phase diagram of $Lu(Pt_{1-x}Pd_x)_2In$ showing the charge-density-wave (structural) transition temperature T_{CDW} and the superconducting transition temperature T_{sc} as function of Pd content x (adapted from [9]).

single crystals of $Lu(Pt_{1-x}Pd_x)_2In$ [1] and started a detailed characterization of the low-energy phonons in the system.

We combined inelastic neutron scattering and high-resolution inelastic x-ray scattering to record the phonons at various $\boldsymbol{Q}$ and to obtain the phonon dispersions along different high-symmetry directions [2]. Figure 2a shows as an example typical inelastic neutron scattering scans across the phonons along $[1\bar{1}0]$ direction around $\boldsymbol{Q} = (2\ 2\ 0)$ in $Lu(Pt_{0.6}Pd_{0.4})_2In$ at room temperature. Well-dispersing acoustic phonons are visible. As a result of the measurements the dispersions of the low-energy acoustic phonons along [110] and along [100] are displayed in Figure 2b and c. While the transverse-acoustic (TA) and longitudinal-acoustic (LA) phonon branches along [100] look normal without any anomaly, the low-energy TA phonon branch along [110] (with polarization along $[1\bar{1}0]$) exhibits a clear dip in the dispersion around (0.5 0.5 0), i.e., a phonon softening occurs at $\boldsymbol{q}_{CDW} = (0.5\ 0.5\ 0)$ at room temperature well above the structural transition at ≈ 140 K for a $x = 0.4$ sample (cf. Figure 1).

For a detailed study of the phonon softening we focused on a $x = 0.5$ sample, i.e., on $Lu(Pt_{0.5}Pd_{0.5})_2In$, and carried out high-resolution inelastic x-ray measurements [2]. Figure 3a and 3b visualize the dispersion of the low-energy TA phonon branch along [110].

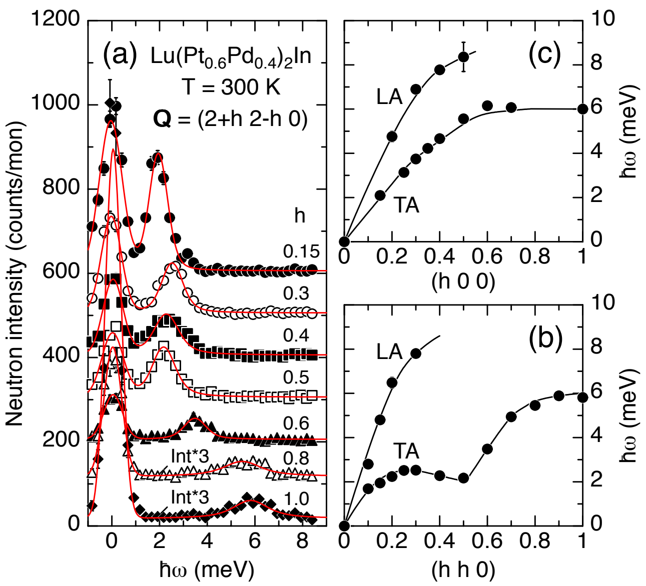

Fig. 2: (a) Inelastic neutron scattering measurements of the low energy acoustic phonons in $Lu(Pt_{0.6}Pd_{0.4})_2In$ along $[1\bar{1}0]$ around $\boldsymbol{Q} = (2\ 2\ 0)$ at room temperature. Data for different $\boldsymbol{Q}$ are shifted vertically with respect to each other. Solid lines are fits to the data. (b,c) Phonon dispersion at $T = 300$ K in $Lu(Pt_{0.6}Pd_{0.4})_2In$ along [110] and [100] as a result of fits to the data shown in (a) and other measurements [2].

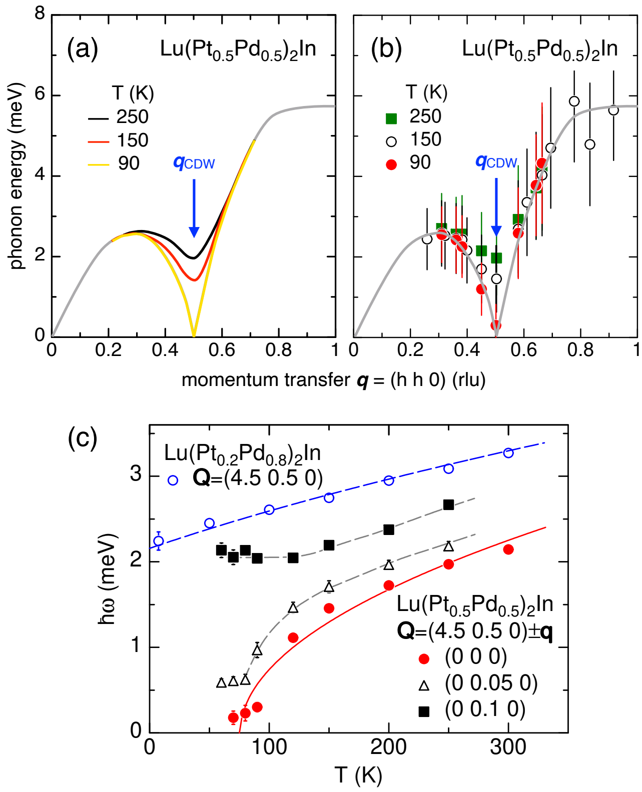

Fig. 3: (a) Schematic view and (b) measured dispersion of the low-energy TA phonon branch along [110] indicating the full softening at $\boldsymbol{q}_{CDW} = (0.5\ 0.5\ 0)$ when lowering the temperature (adapted from [2]). Vertical bars in (b) denote the energy width of the phonon. (c) The lowest-lying phonon energies in $Lu(Pt_{0.5}Pd_{0.5})_2In$ and $Lu(Pt_{0.2}Pd_{0.8})_2In$ at and around $\boldsymbol{Q} = (4.5\ 0.5\ 0)$. The solid lines denotes a mean-field fit to the data, while dashed lines are guides to the eye [2].

Only around $\boldsymbol{q}_{CDW} = (0.5\ 0.5\ 0)$ a pronounced phonon softening is observed when lowering the temperature, while away from $\boldsymbol{q}_{CDW}$ the phonon branches do not show any change with temperature. At $T = 90$ K an almost full phonon softening has taken place in $Lu(Pt_{0.5}Pd_{0.5})_2In$. The phonon energies at certain momentum transfers $\boldsymbol{Q}$ are plotted versus temperature in Figure 3c for $Lu(Pt_{0.5}Pd_{0.5})_2In$ and $Lu(Pt_{0.2}Pd_{0.8})_2In$. A full phonon softening occurs only for $\boldsymbol{q}_{CDW}$, the further away from $\boldsymbol{q}_{CDW}$ the less temperature dependence of the phonon is seen. As visible by the solid line in Figure 3c clear deviations from a mean-field behavior of the soft phonon, i.e., $\omega_{CDW} \propto (T - T_{CDW})^{0.5}$, are detected especially close to T_{CDW}. Compared to theoretical calculations, the observed phonon dispersion relations are quite similar to those predicted. However, the softening occurs in a much narrower momentum range than expected [2].

In line with the unusual temperature dependence of the soft phonon, the superstructure intensity also does not follow a simple mean-field behavior. In particular, the superstructure intensity saturates only at very low temperatures and shows a large tail extending far above the CDW transition. This indicates the importance of critical order-parameter fluctuations which might be related to the pronounced enhancement of the superconducting T_{sc} around the QCP in $Lu(Pt_{1-x}Pd_x)_2In$.

Effect of frustration on quantum-critical behavior

In the hexagonal heavy-fermion compound CePdAl there exists a coexistence of long-range antiferromagnetic order below $T_N = 2.7$ K with 1/3 of the cerium moments remaining disordered due to geometrical frustration. As seen in Figures 4a and 4b a QCP can be reached either by applying hydrostatic pressure in pure CePdAl with a critical pressure of $p_c \approx 0.9$ GPa or by substituting Ni for Pd in $CePd_{1-x}Ni_xAl$ with a critical Ni content of $x_c \approx 0.14$. We performed extensive muon spin rotation (µSR) experiments to study the pressure- and concentration-tuned QCP in CePdAl on a microscopic level [3, 4].

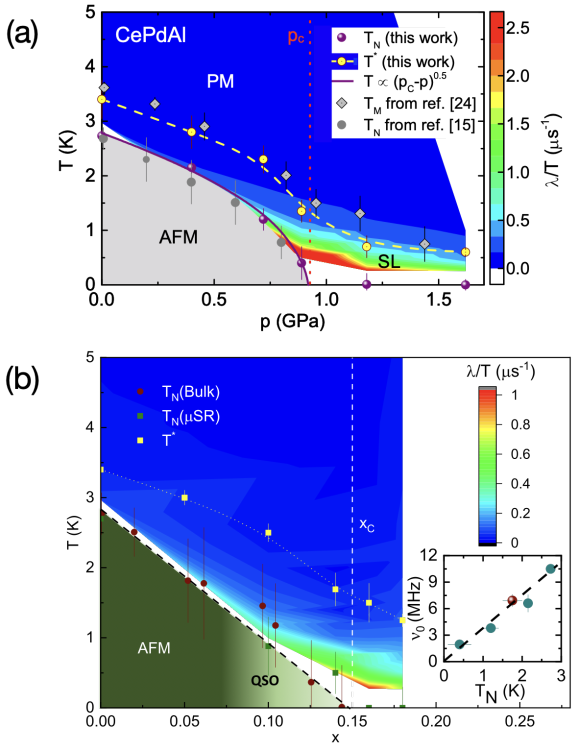

Fig. 4: (a) Temperature – pressure (p,T) and (b) temperature – concentration (x,T) magnetic phase diagram of CePdAl and $CePd_{1-x}Ni_xAl$ together with contour plots of the pressure/concentration and temperature dependent µSR relaxation rates $\lambda(p,T)$ or $\lambda(x,T)$ plotted as λ/T. Inset in (b): Evolution of the ordering temperature T_N and muon oscillation frequency ν_0 with pressure (closed symbols) and concentration (open symbols) [3, 4].

First, the µSR measurements fully confirm the phase boundaries of the antiferromagnetic state (cf. Figure 4). In particular, the linear dependence of the order parameter, here the muon oscillation frequency ν_0 as revealed by zero-field measurements, on the ordering temperature T_N, as displayed in the inset of Figure 4b, corroborates the results obtained previously by neutron scattering [10]. Hence, both techniques nicely demonstrate the equivalence of pressure- and concentration-tuning the QCP in CePdAl.
Second, the µSR results indicate the existence of a temperature scale above T_N, denoted as T^* in Figure 4, which marks the transition from pure exponential relaxation in the zero-field µSR signal above T^* to stretched exponential behavior below T^*. As seen in Figure 4a this crossover temperature scape agrees quite well with the temperature scale T_M of the magnetic entropy accumulation above T_N as a result of magnetization measurements at ambient pressure [11] and electrical resistance measurements under pressure [12]. Furthermore, power-law dependences of the µSR relaxation rates and quantum-critical time-over-magnetic-field scaling hint at the existence of an extended quantum-critical region beyond the QCP, i.e., above p_c or x_c, with spin-liquid behavior [3, 4].

Unconventional superconductivity

$CeCu_2Si_2$ is the first discovered heavy-fermion superconductor located close to a magnetic QCP. The unconventional superconductivity is not mediated by phonons, instead magnetic excitations are at the origin of Cooper pairing in $CeCu_2Si_2$. Recent small-angle neutron scattering (SANS) served us to study the vortex lattice in

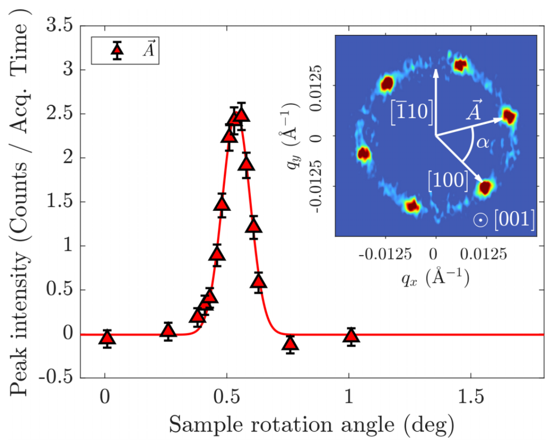

Fig. 5: Rocking scan across a peak (labeled $\overline{A}$ in the inset) of the vortex lattice in $CeCu_2Si_2$ at $T = 0.13$ K and $B = 1.5$ T. The inset displays the full vortex lattice with field applied along the c-axis after summing appropriate scans [6].

this topical heavy-fermion superconductor [6]. As displayed in Figure 5 the vortex lattice in $CeCu_2Si_2$ at $T = 0.13$ K and $B = 1.5$ T is hexagonal without any changes at other fields/temperatures. This finding agrees well with results obtained before by scanning tunneling spectroscopy [13]. The temperature dependence of the SANS intensity points to a fully-gapped multiband superconductivity with a large and a small gap without nodes in the gap structure.

Inspired by the wealth of experimental and theoretical work on the heavy-fermion superconductor $CeCu_2Si_2$ and its unique position among unconventional superconductors, we reviewed the experimental results on the superconducting state in $CeCu_2Si_2$ and presented the different theoretical scenarios, including fully gapped two-band s-wave superconductivity and effective two-band d-wave superconductivity [7]. The latter is able to explain the existing experimental findings in $CeCu_2Si_2$. In addition, we compare in a separate review the neutron spin resonance in the superconducting state of $CeCu_2Si_2$ with the spin resonances in other heavy-fermion superconductors and present reasons why such a resonance in the spin excitations spectrum is only seen in very few heavy-fermion superconductors [8].

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