Experimental report

24/02/2017

Proposal: 4-01-1552 **Council:** 10/2016

Title: Geometrical frustration induced quantum phase transition in CeRh1-xPdxSn: Low energy E/T scaling

Research area: Physics

This proposal is a new proposal

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Samples: CeRh1-xPdxSn x=0.1, 0.3, 0.75, LaRh1-xPdxSn x=0.1, 0.75

Instrument	Requested days	Allocated days	From	To
IN6	6	4	19/01/2017	24/01/2017

Abstract:

The quasi-kagome Kondo lattice CeRhSn exhibits intriguing properties, which ascribe to quantum criticality induced by geometrical frustration. Divergent Gr¿¹neisen ratios provide evidence for a zero-field quantum critical point (QCP). The anisotropy of thermal expansion, which only displays quantum critical behavior along a-axis and not along the c-axis, indicates that geometrical frustration induced QCP in this system. Our recent heat capacity study of CeRh1-xPdxSn series reveals magnetic transitions for x ¡Ý 0.2 (TN=0.2K for x=0.2 and 2.5K for x=0.75), but non-Fermi-liquid type behavior down to 0.09K. We have found that the weak NFL/QCP state of CeRhSn is easily transformed into strong and clear NFL/QCP state with larger C/T in x=0.1 and finally to an AFM ordered state with Pd doping x ¡Ý 0.2. For further study of NFL/QCP of CeRh1-xPdxSn system, we proposed to investigate low energy E/T scaling in x=0.1 and spin waves in 0.3 and 0.75.

Geometrical frustration induced quantum phase transition in CeRh1-xPdxSn: Low energy E/T scaling

Scientific Background: Geometrical frustration describes situations where interactions are incompatible with the lattice geometry and stabilizes exotic phases such as spin liquids. Whether geometrical frustration of magnetic interactions in metals can induce unconventional quantum critical points is an active area of research. Recently the hexagonal heavy fermion metal CeRhSn, where the Kondo ions are located on distorted kagome planes stacked along the *c*-axis. Low-temperature specific heat, thermal expansion, and magnetic Grüneisen parameter measurements prove a zero-field quantum critical point [1-16]. The linear thermal expansion, which measures the initial uniaxial pressure derivative of the entropy, displays a striking anisotropy. Critical and noncritical behaviors along and perpendicular to the kagome planes, respectively, prove that quantum criticality is driven be geometrical frustration. It is also reported a spin flop–type metamagnetic crossover. This excludes an itinerant scenario and suggests that quantum criticality is related to local moments in a spin liquid–like state.

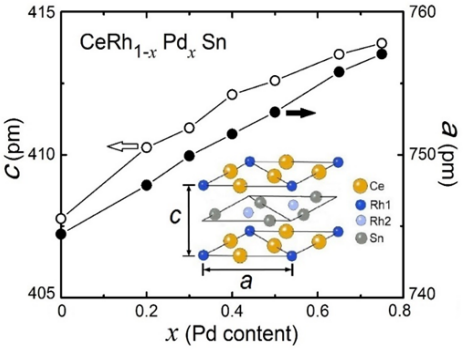

Fig 1. Lattice parameters of $CeRh_{1-x}Pd_xSn$ as a function of x. The insert shows the unit cell of CeRhSn [11].

The equiatomic cerium transition metal stannides CeTSn (T =transition metals) have thoroughly been studied in the last twenty years with respect to their broadly varying magnetic properties [1-5]. CeCuSn, CeAgSn, CeAuSn, and CeZnSn crystallize with hexagonal ordering variants of the AlB_2 type [Fig. 1]. The transition metal and tin atoms form puckered T_3X_3 networks which are separated by the cerium atoms. These four stannides contain stable trivalent cerium and exhibit magnetic ordering at low temperatures. In case of hexagonal CeRhSn compound, layers composed of Ce and Rh1 atoms alternate along the *c*-axis with layers composed of Rh2 and Sn atoms as shown in insert of Fig. 1 [11]. This structural aspect suggested that the Ce 4f state is strongly hybridized with the 4*d* band derived from the Rh2 atoms, leading to the valence-fluctuating state with a high Kondo temperature $T_K \approx 200$ K. Below 7 K, however, non-Fermi liquid (NFL) behavior appear [17]: (a) Magnetic susceptibility $\chi(T)$ shows a power-law behavior, (b) Resistivity $\rho(T) \sim T^n$ ($n \leq 1.5$), and (c) the specific heat divided by temperature C/T shows an upturn and saturation to a large value of 200 mJ / K^2mol [Figs. 2]

The observed NFL behaviors is thought to be manifestation of the frustration of Ce ions moments which hinders the long-range order and leads to a QCP. Furthermore, a zero-field QCP in CeRhSn was indicated by the measurements of specific heat, uniaxial thermal expansion, and magnetic Grüneisen parameter [17]. We have studied how the ground state changes with the substitution of Pd for Rh in $CeRh_{1-x}Pd_xSn$ ($0 \leq x \leq 0.75$) by measuring the specific heat, magnetic susceptibility. With increasing x, the lattice parameters *a* and *c* increase almost linearly by 1.6%, the paramagnetic Curie temperature changes from -450 K to 16 K, and M(B = 5 T) at T = 1.8 K increases from 0.05 μB/Ce (x=0) to 1.4 μB/Ce (x=0.7). The evolution of a long-range AFM order manifests in sharp peaks in heat capacity at 0.2, 0.3 0.7, 1.0, 1.5, 2.5 K for x =0.2, 0.3, 0.4, 0.5, 0.65, 0.75 K respectively (Fig.2). Interestingly x=0.1 does not exhibits a clear sign of long range ordering 0.09K, but reveals a clear sign of NFL behavior, i.e. C/T ~ -ln(T). Our results indicate that the doping of 4*d* electrons in the NFL system CeRhSn destroys the coherence of the quasi-kagome lattice and weakens the hybridization between the 4*f* state and conduction bands, leading to the evolution of first NFL state and then AFM order.

Inelastic neutron scattering study on IN: We had been given 5 days of beam time on IN6 to investigate temperature and composition dependence of quantum fluctuations in $CeRh_{1-x}Pd_x$ (x=0.1 to 0.8) samples in polycrystalline form using low energy inelastic neutron scatter. All the measurements were performed with the incident neutrons energy of 3.1 meV. The low energy INS study is crucial to investigate E/T scaling near QCT as well as, the spin wave energy. During this allocated beam time we have investigated x=0.1 alloy down to 0.07 K using a dilution fridge and x=0.2 and 0.8 alloys between 1.5 K and 150 K (Fig.3). The powder sample of x=0.1 was mounted in an annular Cu-sample holder with an outer diameter of 16 mm, inner diameter of 14mm and the sample thickness of 2 mm and measurements were performed using a dilution fridge between 0.07 K and 100 K. The Cu-can was filled with He-gas to thermalize the sample temperature. We also measured the

empty Cu-can at 1.5 K to subtract the background. The powder samples of x=0.2 and 0.8 were mounted in a thin Al-foil envelope and using a Cd-sample holder, 30mm (wide) x 40mm (height). The scattering was very weak in the x=0.1 sample and hence we collected data for 6 hours per temperature point. Fig.3a shows that the Q-integrated, between 0.2 and 2 $Å^{-1}$, back ground subtracted (the from Cu-can) scattering intensity at various temperatures. The data do not show clear change with temperature between 0.07 K and 50 K on the neutron energy loss side, but follows the population factor on the neutron energy gain side. This is a typical response observed in many NFL/QCP systems, for example $CeRh_{0.8}Pd_{0.2}Sb$. On the other hand the data above 50 K reveal a deviation from the NFL behaviour. The INS data of x=0.2 and 0.8 also exhibit clear low energy scattering between 1.5 K and 150 K. The response in x=0.2 also exhibits broadening at 100 K. On the other hand the x=0.8 shows spin wave scattering at 1.5 K with the zone boundary energy of 1 meV, but NFL type response between 5 and 100 K. Due to limited time available on IN6 we could not investigate x=0.5 alloy. A detailed analysis of the data is in progress.

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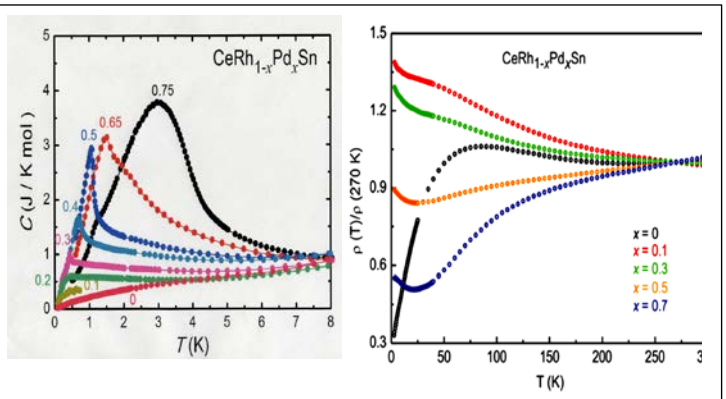

Fig 2. Specific heat of $CeRh_{1-x}Pd_xSn$ plotted as C /T vs logT. Temperature dependence of the reduced electrical resistivity of CeRhSn and samples from the solid solution $CeRh_{1-x}Pd_xSn$with x = 0.1, 0.3,0.4, 0.5,0.65 and 0.75.

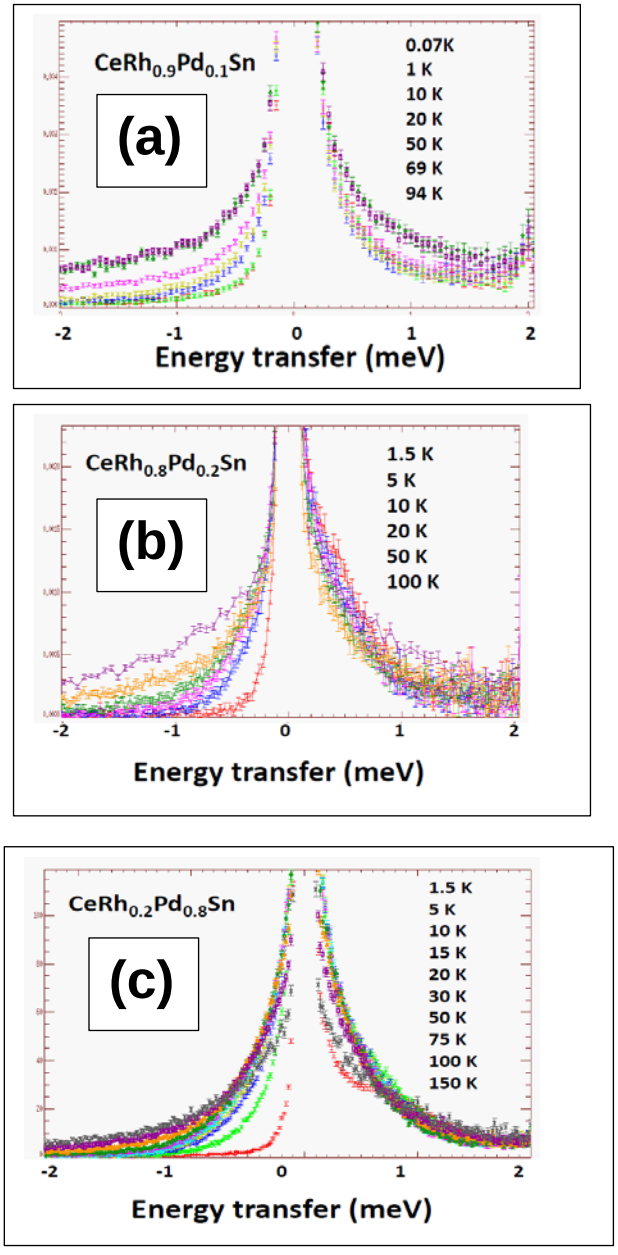

Fig 3. Q-integrated, between 0.2 and 2 $Å^{-1}$, inelastic neutron scattering from $CeRh_{1-x}Pd_xSn$ (top) from x=0.1, middle x=0.2 and (bottom) from x=0.8.