Experimental report

15/02/2023

Proposal: **4-03-1754**

Council: 4/2021

Title: Quantum criticality from sequential Kondo destruction in Ce3Pd20Si6

Research area: Physics

This proposal is a resubmission of 4-03-1749

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Samples: Ce3Pd20Si6

Instrument	Requested days	Allocated days	From	To
THALES	8	8	31/08/2021	08/09/2021

Abstract:

Quantum criticality remains at the forefront of modern condensed matter physics, and is increasingly drawn upon to classify strongly correlated electron phases (ref. 1 of proposal). Of particular interest in heavy fermion compounds are Kondo destruction quantum critical points (QCPs), where electronic degrees of freedom are critical in addition to the (bosonic) order parameter fluctuations. The canonical situation that entails the spin Kondo effect has previously been observed by inelastic neutron scattering (refs. 2,3 of proposal), but it is unknown whether the orbital Kondo effect can undergo a similar transition. This is what we propose to study here, using the material Ce3Pd20Si6 where transport and thermodynamic data suggest such a situation to be realized (ref. 7 of proposal).

Introduction

The cage compound $Ce_3Pd_{20}Si_6$ shows an antiferromagnetic (phase III) and two distinct types of multipolar-ordered phases (II and II') separated by two field-tuned quantum critical points (QCP_1 and QCP_2) for **B** || [001] [1, 2]. In our recent experiment performed on the high-field QCP_2 at PANDA we could not pin down whether the dynamical structure factor $S(\mathbf{Q}, \omega)$ shows the quantum critical scaling expected for a Kondo destruction QCP because (a) the overlap in data from different temperatures was too small and (b) the resolution at low energies was insufficient. Therefore, in the proposal for the present beamtime we had suggested to collect more data using the cold-neutron three-axis low energy spectrometer ThALES.

Experimental configuration

Measurements were performed on two coaligned single crystals of $Ce_3Pd_{20}Si_6$ resulting in the total sample mass of ~ 5.9 g, which were grown by the floating-zone technique in a 4-mirror furnace. The sample was mounted on a copper sample holder for good thermalization in the 2.5 T cryomagnet with its crystallographic (001) axis aligned vertically. The resulting scattering plane was ($HK0$) and we fixed the final neutron wavelength k_f to 1.3 Å. We used a dilution refrigerator and managed to cool the sample down to 60 mK.

Summary and outlook

We started the measurement sequence by checking the sample alignment and Bragg scattering intensity for $\mathbf{Q}$ = (010). In addition, after reaching base temperature to accurately determine the quantum critical field, we investigated the inelastic neutron signal between 1.5 and 2.1 T in steps of 0.2 T for several wavevectors. The idea was to determine where we have the highest scattering intensities at low energies. From the raw data, we did not see any clear field dependence of the signal.

In a previous experiment conducted at the disk chopper time-of-flight spectrometer IN5 on the same sample, the constant-energy cuts integrated in energy ranges just above the elastic line show that in the vicinity of QCP_2 the quasielastic signal is essentially $\mathbf{Q}$-independent [2]. Moreover, by means of transport measurements the critical field was determined to be 1.73 T, consistent with the INS data [1].

Here we measured the full temperature dependence from 0.06 to 10 K at $\mathbf{Q}$ = (010) from -0.1 to 1 meV at 1.73 T. Firstly, it was crucial to measure the background from the empty cryostat. We can see that below 0.1 meV there is a small peak generated by the scattering from the instrumental set-up, which might mask the quantum critical signature (Fig. 1a and red line in b). We subtracted this background contribution from the raw data at every temperature to end up with the total scattering intensity due to the sample and sample holder (Fig. 1c). We are currently carrying out a very careful analysis of this signal, test several ways on how to disentangle the incoherent scattering from the dynamical structure factor $S(\mathbf{Q}, \omega)$ close to zero energy. We are in particular exploring different models to properly eliminate Bragg contamination from the quantum critical signal and to demonstrate whether there is the theoretically expected energy-over-temperature scaling behavior with a fractional critical exponent. We expect the results to be ready for publication in the next few months.

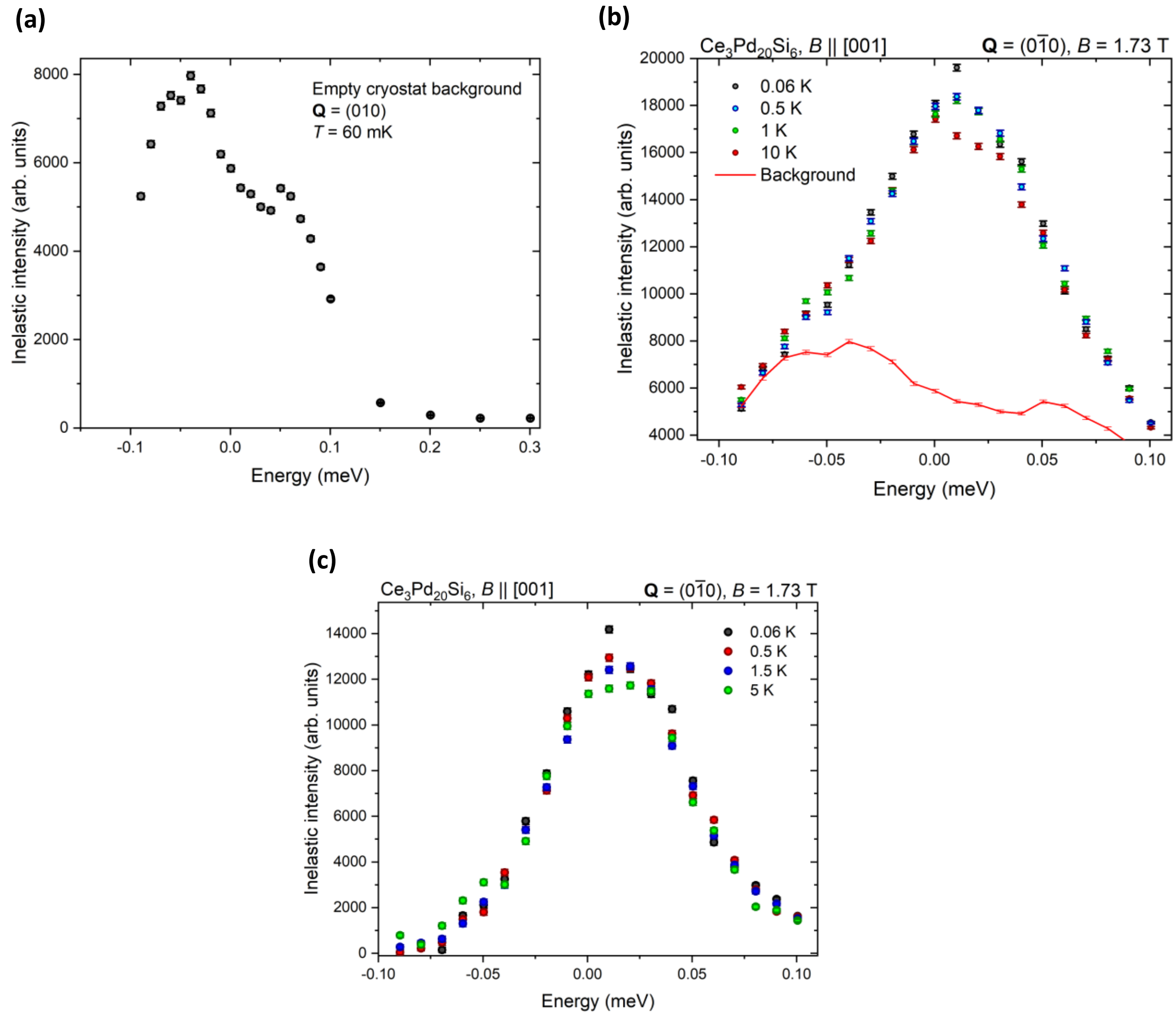

Fig. 1: **(a)** The scattering cross-section from the empty cryostat measured for Q = (010) at 60 mK. **(b)** The total scattering intensity measured at different temperatures. The red line indicates the background signal of the empty cryostat, with calculated error bars by means of standard deviation. **(c)** Constant-**Q** energy scans after instrumental background removal measured at 0.06, 0.5, 1.5 and 5 K.

[1] Martelli, V., Cai, A., Nica, E. M., Taupin, M., Prokofiev, A., Liu, C.-C., Lai, H.-H., Yu, R., Ingersent, K., Küchler, R., Strydom, A. M., Geiger, D., Haenel, J., Larrea, J., Si, Q. & Paschen, S. Sequential localization of a complex electron fluid. *Proc. Natl. Acad. Sci. U.S.A.* **116**, 17701 (2019).

[2] Portnichenko P. Y., Nikitin S. E., Prokofiev A., Paschen S., Mignot J.-M., Ollivier J., Podlesnyak A., Meng S., Lu Z., and Inosov D. S. Evolution of the propagation vector of antiferroquadrupolar phases in $Ce_3Pd_{20}Si_6$ under magnetic field *Phys. Rev. B* **99**, 214431 (2019).