ESRF	Experiment title: Understanding the nature of metamagnetic transitions in Kondo lattices	Experiment number: HE-3893
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Report:

Aim of the experiment was to better understand the origin of metamagnetic transitions in 4f Kondo lattices which is subject of a dispute ongoing since several years. Essentially two competing explanations have been given by theorists as to why a sudden jump in magnetization at the metamagnetic transition is observed. The first argues that the metamagnetism is induced by a localization of 4f states which would be reflected in a change in valence of the magnetic ions. The second argues that pure Fermi surface effects are at play which should leave the 4f states unaffected.

Typically, these metamagnetic transitions are hard to access with X-ray spectroscopies because the changes in magnetization around the transition are small and the required magnetic fields are high. We recently found a very pronounced metamagnetic transition in CeTiGe of about 0.75 μ_B at a field of only 12.5T which makes this compound the most promising candidate to discriminate between the two proposed scenarios. A respective proposal to study the Ce valence and magnetism across the metamagnetic transition by means of Ce $L_{2.3}$ XAS and XMCD at ID12 was awarded beamtime and the results are reported here.

The magnetisation curve as seen in SQUID measurements was well reproduced in Ce L_2 XMCD measurements as a function of field confirming the presence of the strong metamagnetic transition in the samples (Fig. 1a). The opening of the hysteresis loop is consistent with the expected sample temperature at ID12. We then followed the complete shape of the XMCD signal at both the Ce L_2 and L_3 edges across the metamagnetic transition. A change in the Ce valence consistent with the scenario of 4f localization should lead to notable changes in the shape of the XMCD spectrum, in particular, in the pre-edge region that is dominated by 2p-4f quadrupole transitions (E2). We did not detect respective changes in shape of the XMCD but a simple scaling of the full XMCD spectrum with the sample magnetization (Fig. 1b). The analysis of the shape of the XAS spectrum as a function of temperature was severely complicated by the magnetostriction effects of the sample which caused changes in the spectral shape as a function of field because of diffuse diffraction peaks moving with the field. To overcome these issues we powdered the samples down to $20\mu m$

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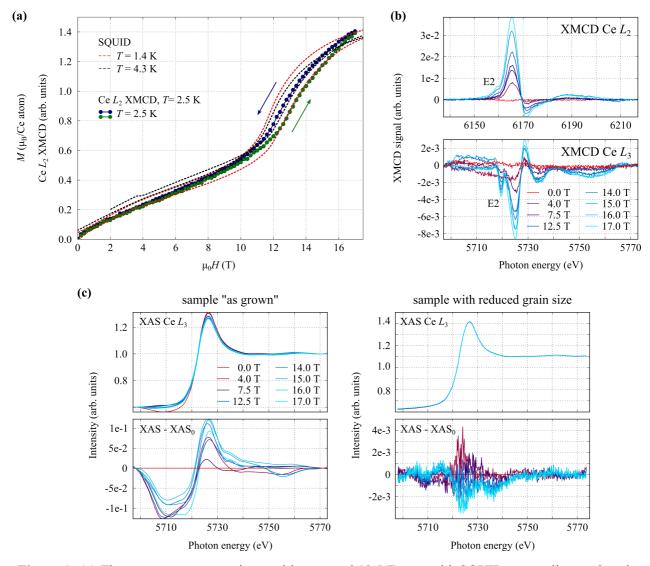


Figure 1: (a) The strong metamagnetic transition around 12.5 T seen with SQUID was well reproduced looking at the Ce L2 XMCD intensity as a function. **(b)** The shape of the XMCD at both the Ce L2 and L3 edge did not change across the metamagnetic transition. In particular, the strong quadrupolar part of the XMCD, marked with E2, remains unaffected across the transition which contradicts the proposed scenario of 4f localization at the transition. **(c)** Magnetostriction effects led to field-dependent changes in the Ce L2 and L3 XAS spectra which are however not related to the metamagnetism. Reducing the grain size of the polycrystalline sample those effects were averaged out and no field dependence of the XAS was observed in agreement with the XMCD data. The magnetisation curve of both type of samples agrees well with that shown in (a).

grain size and prepared them as pellet. The so prepared showed the same magnetization curve and XMCD spectrum and allowed to measure the XAS without being affected by magnetostriction anymore. The respective XAS spectra did not change across the metamagnetic transistion (Fig 1c), again proposing that the metamagnetic transition in CeTiGe cannot be due to 4*f* localization.

Hence both our XAS and the XMCD results are in favour of the scenario that changes in the Fermi surface might be at the origin of pronounced jump in the magnetisation. However, a few weeks before our experiment, results of a similar study on another Kondo lattice, CeRu₂Si₂, were reported where a metamagnetic transition was studied using X-ray absorption and pulsed magnetic fields. A notable field dependence of the XAS was found which the authors interpreted as a 4*f* localization across the metamagnetic transition, in contrast to what we found in CeTiGe. Hence more experiments are needed to clarify the nature of the metamagnetism in Kondo lattices. We propose an additional study of the Ce valence in CeTiGe as function of temperature at fixed magnetic fields in order to see whether the Kondo lattice survives above the transition. This would give a second, independent proof that the metamagnetism at least in CeTiGe is not due to 4*f* localization as a localization of 4*f* states should suppress Kondo behaviour.