Experimental Report

Proposal: **4-01-1297** **Council:** 10/2012

Title: Evolution of the magnetic excitation spectra in Kondo insulator CeRu2Al10 upon Rh doping.

This proposal is a new proposal

Researh Area: Physics

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Samples: Ce(Ru0.95Rh0.05)2Al10

Instrument	Req. Days	All. Days	From	To
IN8	5	5	17/05/2013	22/05/2013

Abstract:

An intriguing phase transition has been observed at T0 = 27.3 K in the Kondo-insulator compound CeRu2Al10. Whereas long-range magnetic order does exist below T0, there is strong evidence that the transition cannot be explained in terms of standard AF exchange interactions alone. Further insight into this elusive phenomenon can be gained by varying the number of d electrons on the transition-metal sites. Bulk magnetic measurements have shown that a low concentration of Rh substituted for Ru can restores a more conventional magnetic behavior. The effect on the excitation spectrum must now be studied. Single crystals of this material have been produced, and preliminary experiments performed on the thermal-beam triple-axis spectrometer 2T at LLB. The INS signal is much weaker than in pure CeRu2Al10, despite the factor of two larger AF ordered moment. Measurements with the higher neutron flux of IN8 are needed to study the magnon dispersion with better accuracy.

The results are to be used as a check of RPA magnon calculations applied previously to pure CeRu2Al10.

Evolution of the magnetic excitation spectra in Kondo insulator $CeRu_2Al_{10}$ upon Rh doping.

An intriguing phase transition has been observed at T_0 = 27.3 K in the Kondo-insulator compound $CeRu_2Al_{10}$. Whereas long-range magnetic order does exist below T_0, there is strong evidence that the transition cannot be explained in terms of standard AF exchange interactions alone. A specific role of *c-f* hybridization along the *a* direction has been suggested. In $CeOs_2Al_{10}$, where the unstable valence character is more pronounced, evidence has been reported that a spin gap could open around 45 K, i.e. significantly above the onset of the magnetic order at T_0 = 29 K. The opening af an anisotropic gap has also been observed in optical conductivity measurements in the same temperature range. In the paramagnetic regime, the susceptibility exhibits a large anisotropy, with the easy direction parallel to the orthorhombic *a* axis but, in the AFM state, the ordered Ce moment derived from the neutron diffraction results is oriented along *c* [J.-M. Mignot *et al.*, J. Phys. Soc. Jpn. Suppl. **80SA**, SA022 (2011)]. This indicates the existence of strongly anisotropic exchange interactions, which has been confirmed by our previous unpolarized (LLB/2T, IN8: 4-01-1086) and polarized (IN20: 4-01-1086 & 4-01-1139) neutron scattering experiments. Results have been published in: J. Robert *et al.*, Phys. Rev. Lett. **109**, 267208 (2012).

To gain further insight into these intriguing properties, studies have been undertaken in which the number of *d* electrons on the transition-metal sites is varied by doping. Bulk magnetic measurements have shown that a low concentration of Rh substituted for Ru [A. Kondo *et al.*, J. Phys. Soc. Jpn. Suppl. **82**, 054709 (2013)] (or of Ir substituted for Os [J. Kawabata *et al.*, arXiv cond-mat.str-el, 1401.3414 (2014)]) restores a more conventional magnetic behavior in which, e.g., the AF ordered moment aligns along the easy *a* axis.

The aim of the present proposal was to study the resulting changes in the spin dynamics of a sample containing 5% Rh. Single crystals of this material had been prepared, and measured in a preliminary experiment on the thermal-beam triple-axis spectrometer 2T at the LLB. In comparison with the pure compound, the energy scans at T = 3 K showed a decrease of the gap, and suggested the existence of at least one weak excitation branch with reduced dispersion. However, the inelastic magnetic signal was much weaker, and also broader, than in $CeRu_2Al_{10}$, despite a larger AFM moment (by about a factor of 2). As a result, it was difficult to characterize the magnon branch, and we thus hoped that the higher flux available on IN8 would lead to more conclusive results.

Sample mount

The 6 single crystals previously measured on 2T were supplemented by 9 additional pieces prepared for the present experiment. All crystals were co-aligned visually under the binocular with the orthorhombic *b* axis vertical (according to previous x-ray diffraction data) and glued with silver paint on a new, lighter, sample holder prepared by spark erosion to minimize the amount of Al in the beam (Fig. 1).

A quick check on IN3, confirmed by the final alignment on IN8, revealed that the mosaic spread was somewhat degraded, with groups of satellites lying as far as 4° from the center of mass. However, this was difficult to correct with the present setup and it was considered acceptable for our purpose.

The sample holder was connected to the sample stick of an orange cryostat and cooled down to 2 K.

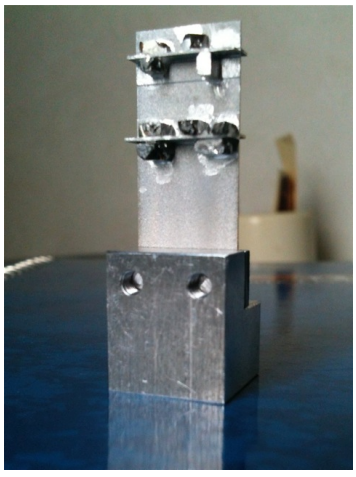

Fig. 1. Sample mount

Experimental conditions

We first tried to reproduce the data collected previously on 2T, starting from the (1, 0, 3) peak, which showed a peak at about 3 meV.

The results did not agree with our expectations. Namely, no excitation could be clearly resolved from the elastic peak. Attempts made to improve the measuring conditions were unsuccessful:

- with the Si monochromator, the spectral shape did not improve, while the counting time increased by a factor of 3.

- with the PG monochromator, working at $k_f = 1.97$ Å^{-1} produced too many spurious contaminations, as revealed by the counting rate in monitor M2.

We thus returned to the initial conditions and measured constant-$\boldsymbol{Q}$ scans at $T_{\min} = 2$ K along high-symmetry directions, as well as at several zone centers. Counting times were on the order of 1 min, for a total of about 1 hour per scan.

The results did not indicate any well-behaved magnetic excitation branch (Fig. 2). Furthermore, a peak-like structure observed in the 2-4 meV region for some $\boldsymbol{Q}$ vectors looked dubious.

To sort out the problem, we heated up the sample to $T = 30$ K > T_N and repeated most of the scans. The results clearly show the appearance of a quasielastic-like scattering. Its intensity (determined by integrating the difference between the signals measured at 30 K and 2K) is in general largest at the position of the strongest magnetic Bragg peaks, suggesting that the signal arises from critical AFM fluctuations. On the other hand, the low-energy inelastic signal, where it exists, remained unchanged upon heating (see, e.g. Fig. 3). Furthermore, for $\boldsymbol{Q} = (3, 0, 1.2)$, no corresponding peak was observed at 30 K on the energy-gain side, although the intensity ratio between positive and negative energy given by the Bose factor at this temperature should be only on the order of 3 for $E = 3$–4 meV $\approx kT$. The fact that the detailed balance factor is not fulfilled confirms that the observed peak does not correspond to a

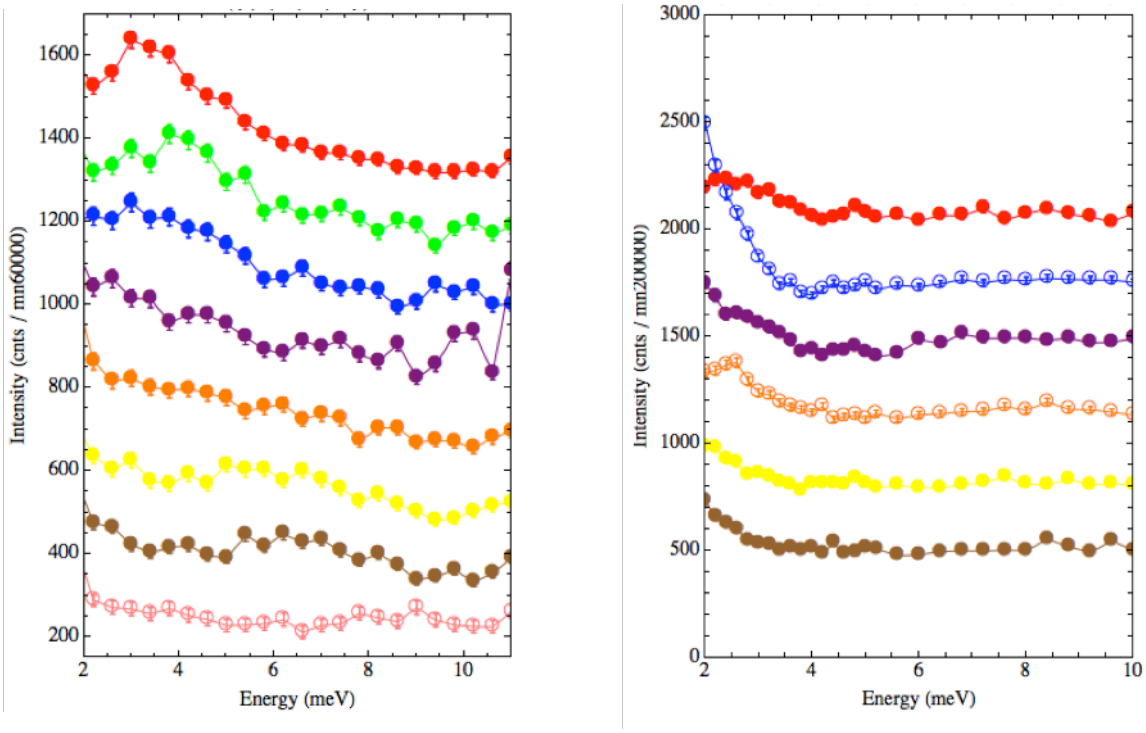

Fig. 2. Left frame (LLB/2T): $\boldsymbol{Q} = (1, 0, l)$, PG monochromator, $k_f = 2.662$ Å^{-1}, $T = 3$ K. From top to bottom : $l = 3.0$, 2.9, 2.8, 2.6, 2.4, 2.2, 2.0, and 1.0. Each curve shifted by 150 cnts.

Right frame (these measurements): $\boldsymbol{Q} = (1, 0, l)$, PG monochromator, $k_f = 2.662$ Å^{-1}, $T = 2$ K. From top to bottom : $l = 3.0$, 2.8, 2.6, 2.4, 2.2, and 2.0. Each curve shifted by 500 cnts.

real excitation. We now suspect that it may be the same signal which was ascribed to a weakly dispersive excitation in the 2T measurements.

The possibility of a Bragg contamination due to the large mosaic spread of the sample was not confirmed because the intensity did not change in a rocking scan performed through the peak position. Another source of contamination could be the incoherent scattering from hydrogen contained in the silver paint. This would explain why the same type of signal already existed on 2T, but the tiny amount used to fix the crystals makes this explanation dubious. At this stage, we have found no satisfactory interpretation. The main problem is actually the small sample volume, and the reduced inelastic scattering cross section with respect to $CeRu_2Al_{10}$ measured previously.

Finally, since no appropriate conditions could be achieved to study dispersive modes in the AFM state, we used the rest of the beam time to measure the temperature dependence of the QE signal (Fig. 4). We also took the opportunity to record the T dependence of the magnetic Bragg intensity (Fig. 5: counting at the nominal position of the 101 peak), which, was not previously determined for this Rh concentration.

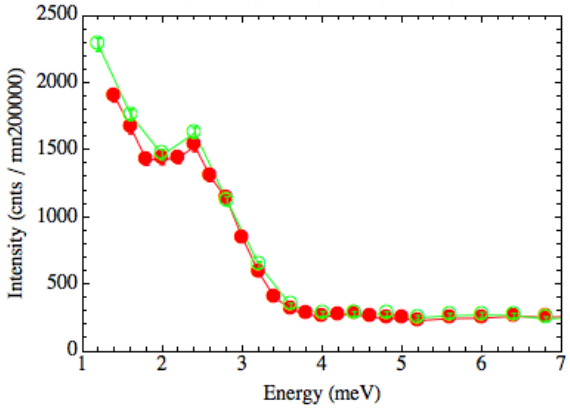

Fig. 3. $\boldsymbol{Q}$ = (3, 0, 1.4) PG monoc., k_f = 2.662 A^{-1}. T = 2.0 K (red), 30 K (green)

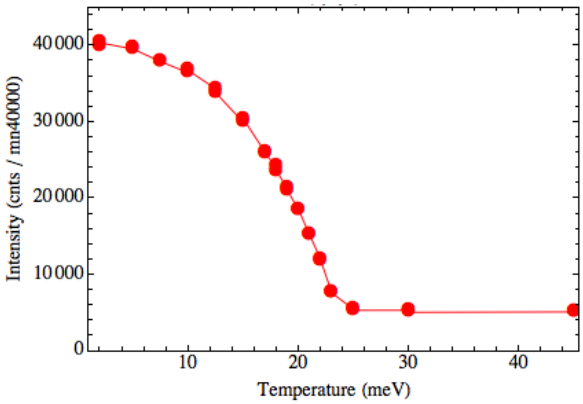

Fig. 5. Magnetic Bragg intensity for $\boldsymbol{Q}$ = (1, 0, 1) PG monochromator, k_f = 2.662 $Å^{-1}$.

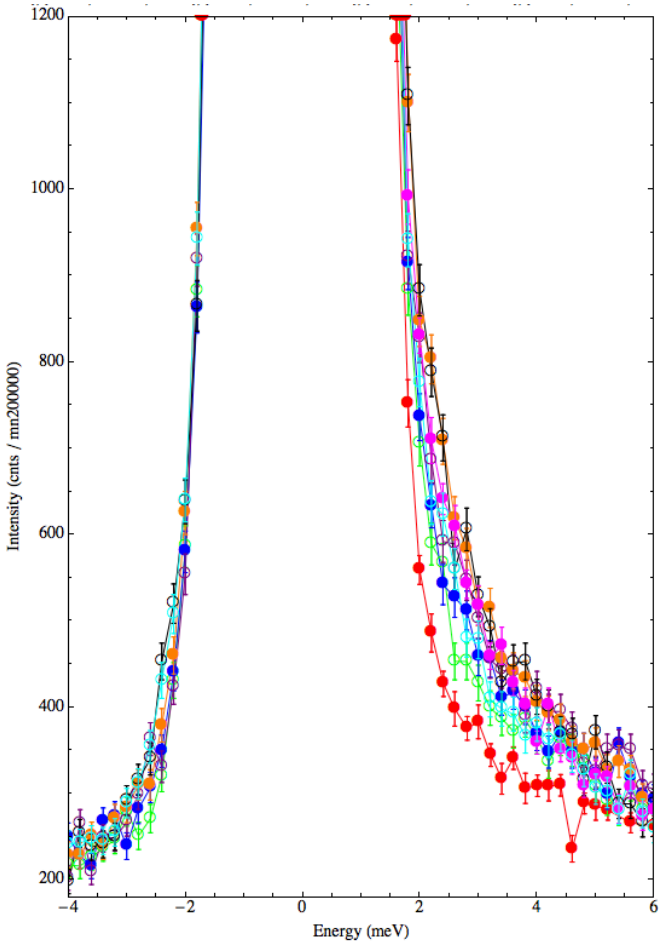

Fig. 4. Quasielastic signal for $\boldsymbol{Q}$ = (1, 0, 1) at T = 2 K (red), 17 K (green), 19 K (blue), 21 K (violet), 23 K (orange), 26 K (black), 30 K (purple), and 43 K (cyan). PG monochromator, k_f = 2.662 $Å^{-1}$.