Experimental report

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Title: Low-energy magnetic excitations inRh-substituted $CeRu_2Al_{10}$

Research area: Physics

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Samples: $CeRu_{1.9}Rh_{0.1}Al_{10}$

Instrument	Requested days	Allocated days	From	To
THALES	5	5	04/11/2015	09/11/2015

Abstract:

An intriguing phase transition has been observed at T_0 = 27.3K in the Kondo-insulator compound $CeRu_2Al_{10}$. Whereas long-range magnetic order exists below T_0, there is strong evidence that the transition cannot be explained in terms of standard AF exchange interactions alone. Further insight into this physics can be seeked 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 restores a more conventional magnetic behavior. The effect on the excitation spectrum must now be characterized. Single crystals have been produced, and experiments performed on the thermal-beam triple-axis spectrometers 2T and IN8. However, the results remain inconclusive because the signal is much weaker than in pure $CeRu_2Al_{10}$, despite the factor of two larger AF ordered moment, and the excitation branch tentatively identified on 2T could not be confirmed on IN8. The higher resolution and high flux of Thales (optionally IN12) should provide better measuring conditions for observing the magnetic response at low energy.
The results will serve to test the RPA calculations applied previously to $CeRu_2Al_{10}$

4-01-1458 – Low-energy magnetic excitations in Rh-substituted $CeRu_2Al_{10}$

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The orthorhombic compound $CeRu_2Al_{10}$ (*Cmcm*, space group no 63), exhibits a Kondo-insulator behavior below room temperature, but orders in a simple AFM structure below T_0 = 27.3 K. The ordered phase is unconventional in several respects, with *i)* a weak AF Ce moment (m_{AF} = 0.32–0.34 μ_B) oriented parallel to *c*, rather than to the paramagnetic easy *a* axis associated with the single-ion anisotropy, *ii)* an anomalously high Néel temperature, in comparison with other RRu_2Al_{10} compounds, and *iii)* a magnetic susceptibility which decreases below T_0 for *all* directions of the applied field, contrary to behavior of a classical antiferromagnet. Numerous experiments have been performed to understand the origin of these peculiarities, but a comprehensive interpretation has not yet emerged. In this work, we consider the effect of electron doping, through Rh substitution, on the magnetic dynamical response.

Bulk measurements [1] have shown that, for a Rh concentration of only 5%, the material recovers a more conventional behavior, with a larger ordered Ce moment (about 1 μ_B), now aligned parallel to *a* (as subsequently confirmed by neutron powder diffraction [2]), and the normal magnetic anisotropy of an antiferromagnet below T_0, between the temperature dependence of the susceptibility measured with $H \parallel a$ (drop in $\chi_\parallel$) or $H \parallel b$ or *c* (weak variation in $\chi_\perp$). However, T_0 remains anomalous, with only a moderate decrease from 27.3 K to 23-24 K.

The first studies of the spin dynamics were performed on thermal beam TAS spectrometers (LLB/2T and ILL/IN8) for the Rh 5% composition. The most striking observation was that, despite the three-fold increase in the Ce moment, the inelastic signal was *considerably weaker* than in pure $CeRu_2Al_{10}$. In the 2T data, a magnon-like excitation still seemed to exist at the minimum temperature, but its energy was reduced to about 3.5 meV (4.8 meV in $CeRu_2Al_{10}$), and the dispersion was weak. On IN8, however, these results could not be confirmed, possibly because of a contamination of unknown origin occurring in the same energy range.

In the present experiment, we have increased the Rh concentration to 10% in order to enhance the “normal” character of the AFM order, hoping to observe well-defined AF magnon branches. From the previous data, a cold-neutron spectrometer seemed more appropriate in view of the energy range of interest and required resolution. Thales was selected for its high incoming neutron flux.

Sixteen single-crystal pieces, with masses ranging from 30 to 70 mg, were prepared at Hiroshima university by Hiroshi Tanida using an Al flux method. They were co-aligned on a thin Al sample holder machined by spark erosion, and glued using Cytop glue, a fluoropolymer with a low hydrogen content (Fig. 1). The mosaic spread determined from the neutron rocking curves did not exceed 3 degrees (FWHM).

Fig. 1. Sample mount with 16 co-aligned single crystals, for a total of 611 mg.

The measurements were performed in the (*a**, *c**) scattering plane, using the PG 002 monochromator and analyzer of Thales, at the fixed final wavevector $\boldsymbol{k_f}$ **= 1.55 Å^{-1}**. The velocity selector in the incident beam and the Be filter on k_f were used to suppress higher order contaminations, providing a low featureless background on the order of 4.5 counts/mn.

Spectra were recorded at $T_{\min}$ = 1.6 K for several AFM $\boldsymbol{Q}$ vectors (*Y* points), (1, 0, -1), (3, 0, -1), (1, 0, -3), as well as for the nuclear zone centers $\boldsymbol{Q}$ = (2, 0, -1) and (2, 0, -3) (*Γ* points). In all cases, the intensities measured were quite weak. However, at the former two AFM $\boldsymbol{Q}$ vectors, an extra signal was found to exist at energies comprised between 2 and

6 meV. For $\boldsymbol{Q}$ = (2, 0, -1) (Γ point), on the other hand, the intensity practically reduces to the background. For $\boldsymbol{Q}$ = (2, 0, -3) (another Γ point), a sloping contribution was measured at higher energies but, since it was found to persist in the paramagnetic phase (T = 25 K), even gaining intensity as temperature increases to 90 K, it was concluded to be of non-magnetic origin.

The temperature dependence of the magnetic signal at low energy was studied by measuring the $\boldsymbol{Q}$ = (3, 0, -1) spectrum at 4 different temperatures: T = 1.6, 12, 19, and 25 K. Fig. 2 shows the resulting transformation from a spin-gap response at $T_{\min}$ to a quasi-elastic-like shape above T_0 = 23 K.

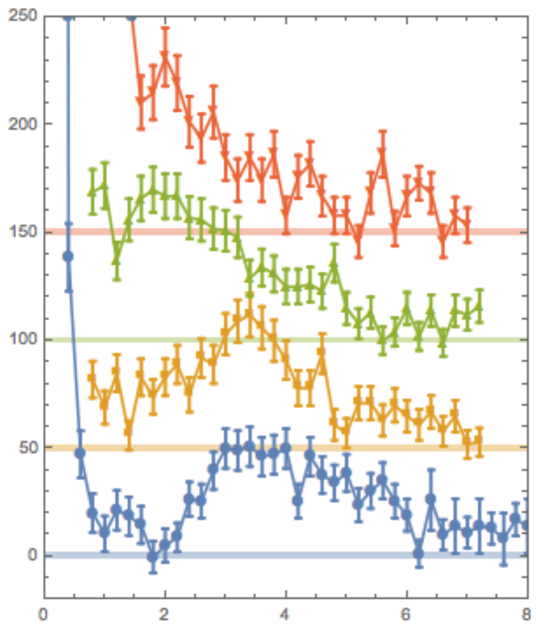
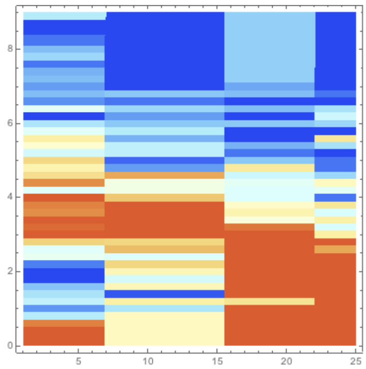
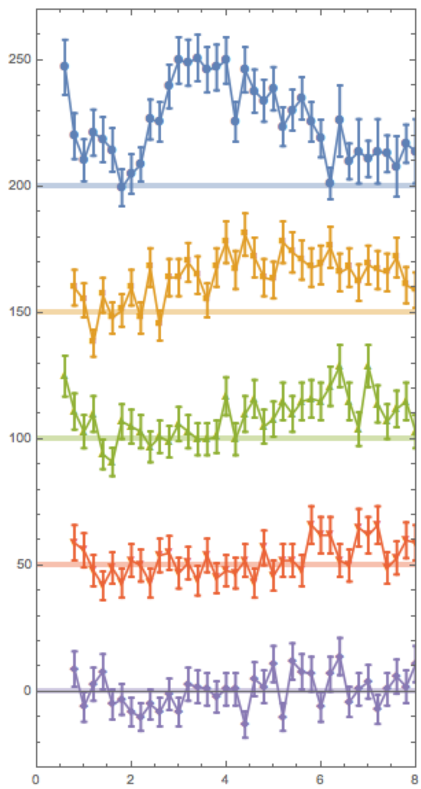


***Fig. 2**. (left)* Temperature dependence of the magnetic excitation spectrum measured for $\boldsymbol{Q}$ = (3, 0, -1). From bottom up: T = 1.7, 12, 19, and 25 K. A constant background of 40 counts has been subtracted, and each curve is shifted vertically by 50 units with respect to the previous one.
(below) Intensity map showing the closing of the spin gap



The dispersion of the excitation was traced along the (3-h, 0, -1) direction with h varying from 3.0 (Y point) to 2.0 (Γ point). The center of the peak shifts from E = 4 ± 0.2 to 7 ± 0.4 meV, while the integrated intensity decreases by about a factor of 2. The results are summarized in Fig. 3.



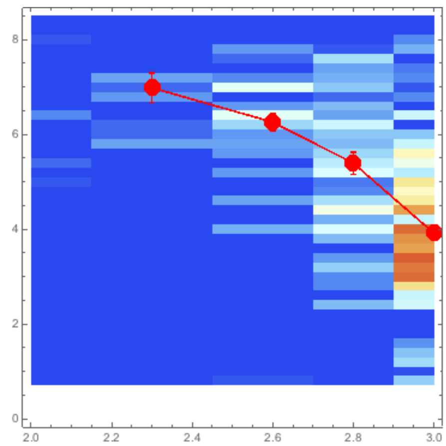


***Fig. 3**. (left)* Dispersion of the magnetic excitation spectrum measured for $\boldsymbol{Q}$ = (3-h, 0, -1). From top to bottom: h = 0, 0.2, 0.4, 0.7, 1. A constant background of 40 counts has been subtracted, and each curve is shifted vertically by 50 units with respect to the previous one.
(above) Intensity map showing the positive dispersion and decrease in intensity along the a^* direction, starting from the Y point (3, 0, -1). Red circles are the result of Lorentzian fits.

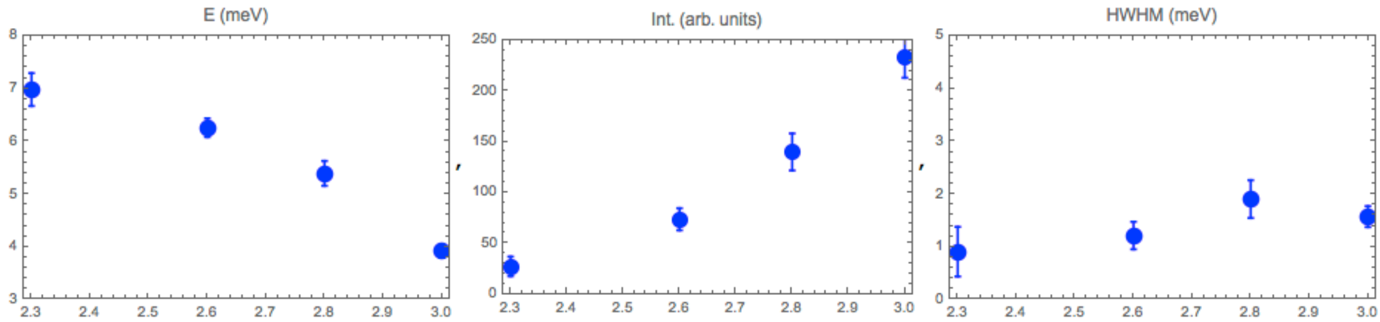


Fig. 4. *h dependences of the energy, integrated intensity, and linewidth (from left to right) of the excitation peak at T = K derived from Lorentzian fits.*

Conclusion

The present measurements have been remarkably successful in view the low magnetic intensity and limited sample volume, emphasizing the spectacular boost in performance achieved after the instrument upgrade from IN14 to Thales. In particular, the low, featureless, background proved to be essential here for singling out the weak signal from the sample.

Clear evidence was found, at base temperature in the AFM phase ($T_{\min}$= 1.65 K << T_0 = 23 K), for a dispersive magnetic excitation branch with a spin gap of about 4 meV, reaching ≈ 7 meV near the Γ point. This branch disappears on heating to 25 K > T_0. It is reminiscent of that observed previously in pure $CeRu_2Al_{10}$ but the spin gap is a little smaller, and the peak intensity is *considerably reduced*. This strong suppression, achieved by substituting a minor amount of Rh for Ru, was already pointed out in the 2T experiment for x = 0.05, but the weak magnetic signal could not be properly identified in the thermal beam measurement.

What can be suggested from the present results is that the inelastic response is not directly related to the static AFM (Bragg) component, as would be the case for conventional magnons, but likely arises from the fluctuations of other magnetic degrees of freedom. Such excitations were found to dominate in pure $CeRu_2Al_{10}$, where the static AFM moment was very small (≈ 0.34 μ_B) and the inelastic peak quite intense. They now appear to be steeply depleted by electron doping with Rh substitution, just as a more conventional local-moment behavior in the static susceptibility (in particular with respect to its anisotropy), and a larger ordered moment (≈ 1 μ_B), are recovered.

It is still somewhat surprising that we could not observe a distinct “antiferro-magnon” branch, associated with the transverse excitations of the AFM structure. One should recall, however, that for x = 0.10 (and even up to 0.20, according to Ref. [2]) the ordering temperature remains pathologically high, indicating that the AFM state cannot be regarded as “normal” (in the sense of a simple, local-moment, RKKY + CEF picture).

Unfortunately, it will be difficult to extend the present study much further in this series because of the Rh solubility limit. In fact, a suitable material is still lacking to serve as a reference for a “normal” AFM spin-wave spectrum, especially since, up to now, the same simple $\boldsymbol{k}$ = (0, 1, 0) magnetic structure existing in $CeRu_2Al_{10}$ (and $CeOs_2Al_{10}$) has not been found elsewhere in the 1-2-2 family.

[1] Tanida, H. *et al. Collapse of spin gap by Ru-site substitution in the antiferromagnetic Kondo semiconductor $CeRu_2Al_{10}$. Phys. Rev. B* **90,** 165124 (2014).

[2] Kobayashi, R. *et al. Influence of Electron Doping on Magnetic Order in $CeRu_2Al_{10}$. J. Phys. Soc. Jpn.* **83,** 104707 (2014).