Experimental report

02/02/2017

Proposal: **4-01-1506**

Council: 4/2016

Title: Spin gap and resonance mode in thenew Kondo insulator compound $CeFe_2Al_{10}$

Research area: Physics

This proposal is a continuation of 4-01-1347

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Samples: $CeFe_2Al_{10}$

Instrument	Requested days	Allocated days	From	To
IN20	5	5	23/11/2016	28/11/2016

Abstract:

The prior observation of resonance-like magnetic excitons in archetype Kondo insulators (KI) such as YbB_{12} or SmB_6 raised the possibility of a common physical background for this phenomenon and the celebrated "magnetic resonance" in high-TC superconductors. However, evidence for this behavior was missing so far for Ce-based Kondo insulators. In recent INS measurements on $CeFe_2Al_{10}$, we had observed a dispersive mode near 10 meV, which develops only in the low-T, KI regime and exhibits some of the features expected for this type of excitation.
Subsequent polarization analysis on IN20 confirmed the magnetic origin of the signal, as well as the dominant contribution of magnetic correlations between the m_a components of the Ce magnetic moments. However, the results remained incomplete because of a failure of one power supply during the measurements. The data thus need to be complemented, especially regarding the temperature dependence of the magnetic excitation spectra, both at low energies (appearance of a quasielastic signal in the region of the spin gap) and near 17 meV where a second branch was suspected to exist, but failed to be confirmed in subsequent unpolarized measurements.

Report on experiment 4-01-1506 (proposal #75544)

Spin gap and resonance mode in the new Kondo insulator compound $CeFe_2Al_{10}$

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IN20, linear polarization analysis, 23–28 Nov. 2016

This experiment was the continuation of exp. 4-01-1347 previously performed on IN20. For lack of time, due to the failure of a power supply, information was missing as to the temperature dependence of the magnetic exciton peak observed in this compound at $T = 2$ K, as well as on the appearance of a quasielastic (QE) signal with increasing temperature. The possible existence of a second excitation at higher energy, suspected from the previous data but not confirmed in unpolarized neutron experiments on 2T at LLB, also deserved further investigations.

Characterizing the properties of this "resonance" mode at $E = 10$ meV is important because $CeFe_2Al_{10}$ is to date the only Ce-based Kondo insulator (KI) in which such a feature, previously studied in SmB_6 and YbB_{12}, was convincingly established. Furthermore, interesting anisotropy effects single out this orthorhombic material with respect to its forerunners, both of cubic structure.

The experimental conditions were the same as previously, except for a reduced sample mass (from 8 to 5 grams) due to the loss, in transportation, of some of the co-aligned crystals, which could not be replaced.

The measurements, performed with the <u>*a* axis normal to the scattering plane</u>, were focused on the scattering vector $\boldsymbol{Q} = (0, 3, 0)$, corresponding to the AFM Y point, known from the previous experiment to provides a rather high intensity of the 10-meV peak, with no sizable contamination.

Background correction

At base temperature, $T = 1.8$ K, the main features of the previous measurements could be reproduced, at the cost of longer counting times. Because some of the questions to be addressed concern rather weak signals (QE and hypothetical "second mode"), special care must be exerted to ensure proper background subtraction. In principle, polarization analysis provides a straightforward estimate of the background, obtained as a linear combination of three intensities measured in the spin-flip channel: $bkg = SF_y + SF_z - SF_x$ (1). Unfortunately, the resulting values (Fig. 1, left frame) are affected by prohibitively large statistical errors.

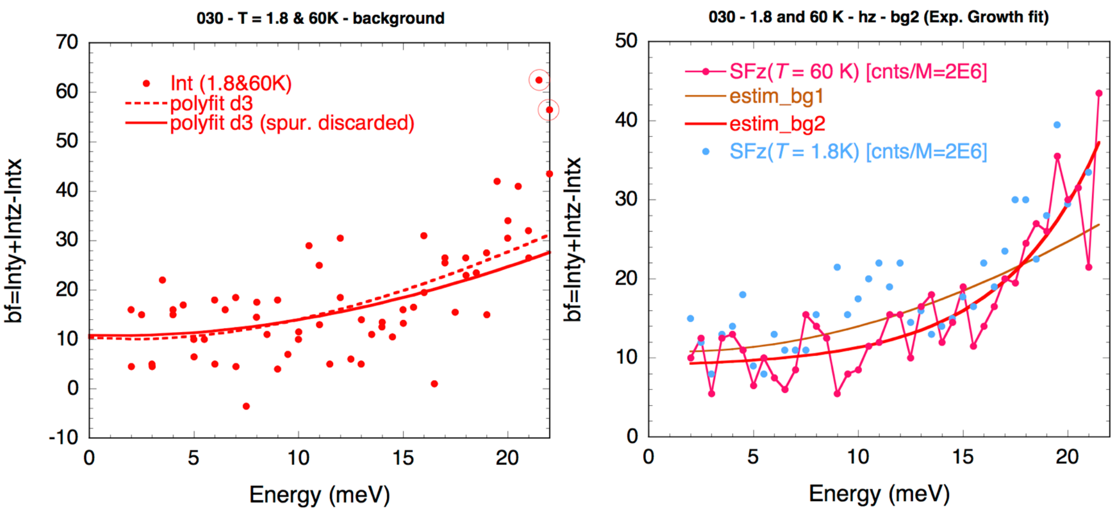

Fig. 1. Estimation of the background: (left) 3rd degree polynomial fit to the *bkg* data [eqn. (1)] for $T = 1.8$ and 60 K; (right) Comparison of the resulting curve (brown trace) with the spectrum measured in the SF channel with h_z polarization at 60 K (red markers), fitted by an exponential law (red trace). The *SFz* data at $T = 1.8$ K (blue markers) are shown as a consistency check.

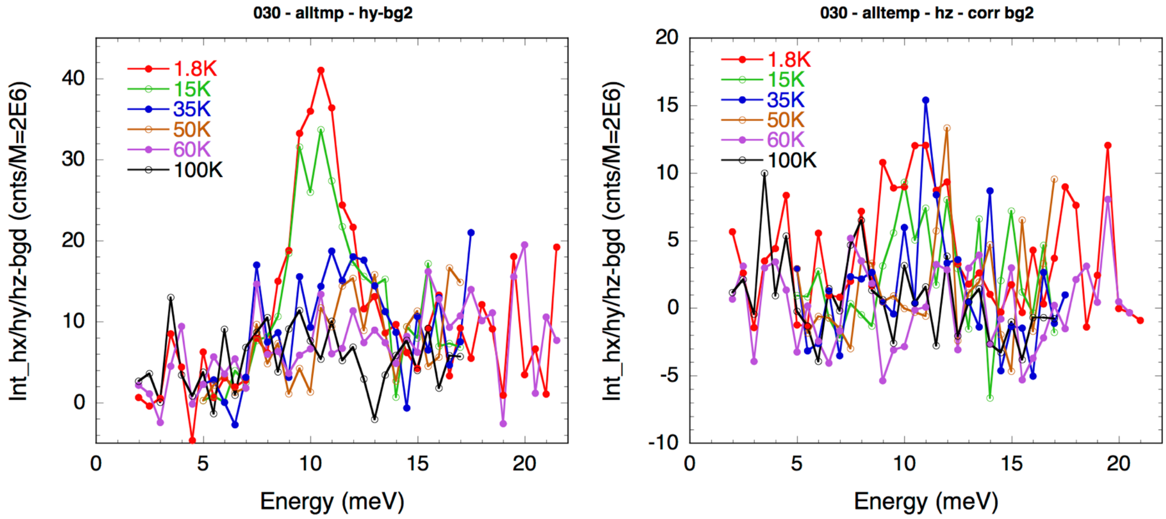

Fig. 2. Temperature dependence of the exciton mode measured at $\boldsymbol{Q} = (0, 3, 0)$ in the spin-flip channel, corrected for the background as explained in the text and shown in Fig. 1. Incident neutron polarization along the *c* axis (left) and the *a* axis (right).

We have nonetheless attempted to fit them (interval $-5 \le E \le 26$ meV), assuming a smooth energy variation, in order to derive an analytical representation of the background (red trace). This approach proved unsuccessful because, over a large fraction of the experimental energy range, the resulting "background" intensity was actually higher than the total spin-flip signal measured at $T = 60$ K for the direction of polarization h_z (Fig. 1, right). Therefore, since the latter *SFz* spectrum is smooth and has low intensity, it provides a reasonable approximation for the background, and will be used hereafter for correcting other spectra measured at the same $\boldsymbol{Q}$ vector for different temperatures and/or polarizations.

Experimental temperature dependence at $\boldsymbol{Q} = (0, 3, 0)$

The spectra measured in the spin-flip channel are shown in Fig. 2 for two different incident neutron polarizations, h_y and h_z, as a function of temperature between 1.8 and 100 K. Note the different intensity scales. All spectra have been corrected for an energy dependent (but temperature and polarization independent) background, as discussed above. The exciton peak at 10 meV reported previously [1,2] is clearly visible at the lowest temperature for the h_y polarization (left frame). At $T =$ 35 K, its intensity is already strongly reduced, with a moderate shift to higher energies (~11.5 meV), and the peak becomes undetectable already at 60 K. It can be noted that the a.c. susceptibility maximum ($H \parallel a$), generally ascribed to Kondo fluctuations, is reached only around 70 K.

In the similar spectrum obtained with h_z polarization (right frame), the excitation peak is still visible but its intensity is drastically reduced. This anisotropy was already observed in the previous experiment [2]. Since, for $\boldsymbol{Q} = (0, 3, 0)$, the spectra measured with h_y and h_z polarizations (incident neutron spin parallel to *c* and *a*) reflect two-site magnetic correlations between the M_a and M_c Ce moment components, respectively, this implies that the excitation mainly involves AFM fluctuations of the *a* components (*a* is also the easy magnetic axis observed in the bulk susceptibility measurements).

Concerning the two issues raised in the introduction:

- The present measurements fail to reveal any sizable QE signal up to $T = 100$ K, at least for the $\boldsymbol{Q}$ vector investigated. Exploratory scans (Fig. 3) performed at other $\boldsymbol{Q}$ vectors: (0, 1, 2)—another zone-boundary *Y*-point, and (0, 2, 0.5)—arbitrarily chosen point inside the Brillouin zone, led to the same negative result. For completeness, we also checked that no QE contribution occurs at $T = 100$ K in the NSF channel. The reason for the discrepancy with the unpolarized neutron measurements performed on LLB/2T on the same sample thus remains unclear. It can be noted that, from the Curie-Weiss behavior observed in the static susceptibility above T~100 K, one would indeed expect such

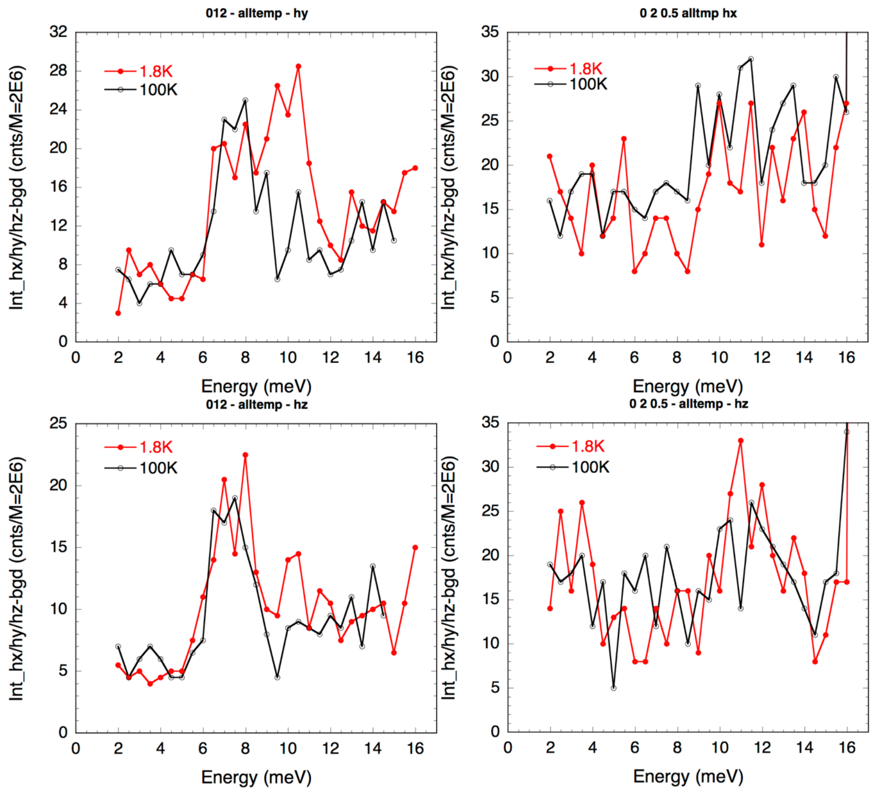

Fig. 3. Spectra measures at T = 1.8 and 100 K for two different $\boldsymbol{Q}$ vectors (0, 1, 2) (Y point at zone boundary - left column) and (0, 2, 0.5) (inside Brillouin zone – right column) for different incident beam polarizations. In the first case, the exciton peak is observed with h_y polarization*, but a strong contamination exists near E = 7 meV, making the position (0, 1, 2) unsuitable for a study of the temperature dependence like that reported above. No clear evidence for an extra QE signal at 100 K is seen in any of these scans.

*Contrary to the measurements performed for $\boldsymbol{Q}$ = (0, 3, 0), the intensity in the SF channel for h_y polarization now contains contributions from both M_a and M_c correlation terms.

low-energy fluctuations to exist, at least in principle, though the magnitude of their contribution to the INS signal at a given $\boldsymbol{Q}$ vector is difficult to predict.

- As to the possibility of a second mode, some rather weak intensity seems to remain, after subtracting the background, at energies above the peak position. However, its lack of temperature dependence, in contrast with the steep suppression of the main peak upon heating, is difficult to reconcile with the assumption of a second branch of AFM excitations arising from the same magnetic moments and exchange couplings. This conclusion is consistent with lack of evidence for a second branch, both on 2T at Orphée-LLB and in single-crystal measurements on MERLIN at ISIS.

In conclusion, the present experiment allowed us to clarify some of the points left unsolved after the previous run on IN20. We have now collected sufficient information to prepare a report on the unconventional spin dynamics of this Kondo insulator compound.

[1] J.-M. Mignot, P. A. Alekseev, J. Robert, S. Petit, T. Nishioka, M. Matsumura, R. Kobayashi, H. Tanida, H. Nohara, and M. Sera, Phys. Rev. B **89**, 161103 (2014).

[2] ILL experimental report 4-01-1347.