Experimental Report

21/04/2015

Proposal: **4-01-1347** **Council:** 4/2014

Title: Spin gap and resonance mode in thenew Kondo insulator compound CeFe2Al10

This proposal is a new proposal

Researh Area: Physics

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Samples: CeFe2Al10

Instrument	Req. Days	All. Days	From	To
IN20	7	7	03/11/2014	10/11/2014

Abstract:

The prior observation of resonance-like magnetic excitons in archetype Kondo insulators (KI) such as YbB12 or SmB6 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 CeFe2Al10, we have 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.
In order to better single out the magnetic signal, to confirm the unusually strong polarization suggested by the unpolarized data, and to trace the transfer of spectral weight to a weakly Q-dependent quasielastic signal with increasing temperature, we propose to take advantage of the polarization analysis available on IN20.
Crystals previously studied at LLB and ISIS by two different teams will be combined to optimize measuring conditions for this experiment.

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

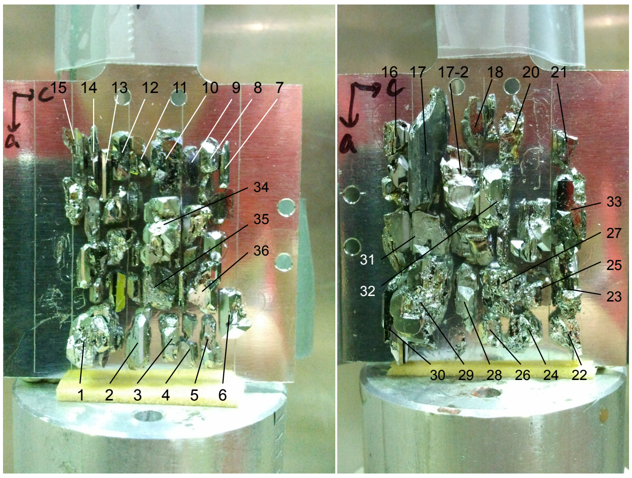
Background

An intriguing phase transition occurring at T_0 = 27.3 K has been discovered a few years ago in the compound $Ce\underline{Ru}_2Al_{10}$. Whereas long-range magnetic order definitely exists 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. The existence of strongly anisotropic exchange interactions has been confirmed by previous unpolarized (IN8: 4-01-1086) and polarized (IN20: 4-01-1086 & 4-01-1139) neutron scattering experiments performed at the ILL, following early measurements as at the LLB and ISIS. However, the nature of the electronic states at low temperature in that material, as well as in the corresponding Os and Fe compounds, is still an open question.

$Ce\underline{Fe}_2Al_{10}$ does not exhibit magnetic order, but belongs to the class of mixed-valence semiconductors (also often termed “Kondo insulators”, hereafter KI), as evidenced by its low-temperature electrical transport properties. Inelastic neutron scattering (powder) spectra measured at T = 7 K on the TOF spectrometer MERLIN at ISIS pointed to the existence of a spin gap, with an inelastic magnetic peak located at about 13 meV [1]. More detailed information on the $\boldsymbol{Q}$ dependence of the magnetic response was obtained from a single-crystal study performed on 2T (LLB). At low temperature, the results reveal an excitation branch with a positive dispersion starting from the AFM reciprocal lattice *Y*-point, $\boldsymbol{q}$ = (0, 1, 0), at an energy slightly above 10 meV. The energy reaches 12-13 meV at the top of the branch, which accounts, at least partly, for the position of the peak observed on powder. With increasing temperature, this magnetic excitation shifts to higher energies while losing intensity, and is no longer observable at T = 95 K. In Ref. [2], this spectral component was interpreted as a “resonance mode”, reminiscent of that found in HTC cuprates and other superconductors [3]. The possibility for such a “magnetic exciton mode” to form at low temperature in KI materials was first pointed out by Riseborough [4], in connection with INS studies of SmB_6 [5] and YbB_{12} [6]. As with superconductors, the effect is directly related to the opening of an electronic gap resulting, in the KI case, from the “*c-f*” hybridization between local 4*f* and conduction 5*d*-6*s* states [7]. $CeFe_2Al_{10}$ is to date the only Ce-based KI compound in which convincing evidence for this effect has been reported.

The aim of the present proposal was to gain further insight into the above phenomenon by *i)* properly separating out the magnetic signal, especially above 12 meV where a strong sloping nuclear background exists in the 2T data; *ii)* studying the anisotropy of the correlations from a full polarization analysis; *iii)* searching for a quasielastic signal at higher temperatures, where the exciton mode is suppressed.

Measuring conditions



The experiment on IN20 was carried out in the linear polarization configuration. The sample consisted of a large number of small single crystals prepared by Y. Muro, fixed onto an Al plate by means of hydrogen-free Cytop glue and co-aligned with their orthorhombic *a* axis normal to the scattering plane. The resulting mosaic spread was relatively large, of the order of ±5° from the profile of nuclear rocking curves. Measurements were performed down to T_{min} = 2 K in a standard ILL cryostat. Some tests were performed at k_f = 4.1 Å^{-1}, but the best conditions, used for all data presented hereafter, were obtained for k_f = 2.662 Å^{-1}, with a PG filter placed on the scattered beam to remove second-order contamination.

Results

1. Base temperature, T_{min} = 1.8 K

The spectra measured at the AFM *Y* point, $\boldsymbol{Q}$ = (0, 3, 0) confirm the existence of the excitation at 10 meV. The corresponding component is found in the SF channel, with a large intensity for P_x and P_y polarizations, and a much weaker one for P_z. Polarization analysis reveals that the latter signal arises mainly from a contamination, also detected by the second monitor, and that the M_y component, obtained as SF_x–SF_z and associated with magnetic correlations along *c*, is quite weak in comparison with the M_z component (correlations along *a*). This predominance of $\langle m_i^a m_j^a \rangle$ correlations supports the conclusion drawn from the unpolarized measurements of Ref. [2] by comparing the magnetic response at equivalent *Y* points, *e.g.*, $\boldsymbol{Q}$ = (3, 0, 0) vs. (0, 3, 0), or (1, 2, 0) vs. (2, 1, 0). It is also consistent with the easy *a* axis derived from bulk magnetization studies. The present results further suggest the existence of a second mode, also polarized along *a*, at an energy of about 17 meV.

For $\boldsymbol{Q}$ = (0, 0, 3) (nuclear zone center, forbidden according to the symmetry condition: *l* even if *k* = 0) the difference SF_x–SF_z shows a weak maximum at about 15 meV but, surprisingly, some residual intensity seems to exist down to 10, or even 7 meV.

The dispersion was studied along the (0, 3, *l*) direction. The data confirm the *q* dependence observed in the unpolarized measurements. In addition, the upper mode near 17 meV is enhanced as *l* increases, and becomes dominant (without a significant shift in energy) for *l* = 0.5.

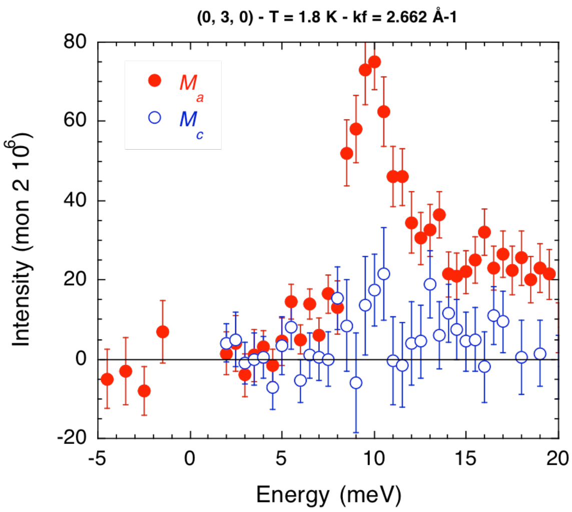


Fig. 1. *Polarization analysis of the INS spectrum for* $\boldsymbol{Q}$ *= (0, 3, 0) measured at T = 1.8 K. Correlations between the a and c components of the Ce magnetic moments, obtained as the differences* SF_x–SF_z *and* SF_x–SF_y*, are plotted using closed (red) and open*

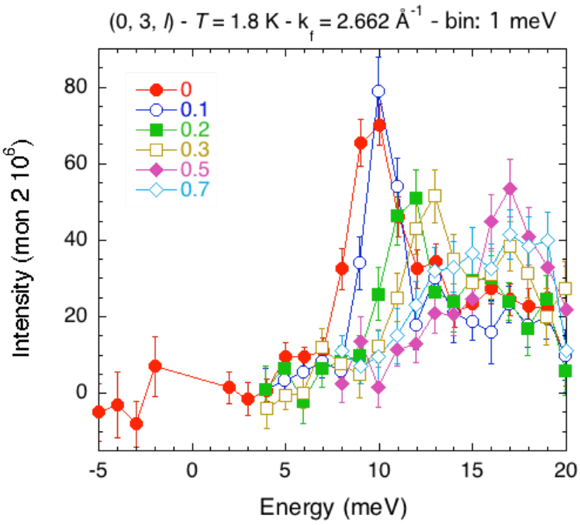


Fig. 2. *Dispersion of the magnetic excitation branches along the (0, 3, l) direction at T = 1.8 K. Data measured with a step of 0.5 meV have been rebinned to 1 meV. The plot shows the difference* SF_x–SF_z*, associated with correlations between a components.*

2. Temperature dependence

The second part of the experiment was devoted to studying the temperature dependence of the magnetic response. Unfortunately, one of the power supplies of the polarization coils failed in the last days of the experiment. The problem was not immediately detected and caused the loss of about 15 hours of beam time. As a result, the data collected as a function of temperature remain incomplete. For $\boldsymbol{Q}$ = (0, 3, 0) (SF_x–SF_z plotted in Fig. 3), the intensity loss and shift to higher energies exhibited by the low-energy mode is in line with the behavior reported in Ref. [2], whereas the intensity in the energy range of the second branch (~ 17 meV) remains essentially unchanged.

For $\boldsymbol{Q}$ = (0, 0, 3) (Fig. 4), the most surprising issue is the lack of detectable quasielastic (QE) signal at T = 100 K. This is in contrast with the observation made in Ref. [2] for $\boldsymbol{Q}$ = (0.5, 3, 0). In the latter case, the QE contribution was already clearly visible at T = 50 K, whereas, in the present experiment, no extra intensity appears in the few data points that could be measured at T = 55 K for $\boldsymbol{Q}$ = (0, 3, 0). It is difficult to draw a conclusion at this point for lack of experimental data for other $\boldsymbol{Q}$ vectors and temperatures. However, this apparent discrepancy would deserve further investigations since it may reflect some peculiarity of the $\boldsymbol{Q}$ dependence and/or polarization of the correlations in the high-temperature spin-fluctuation regime.

In summary, the present results substantiate the conclusions drawn from the previous unpolarized studies and provide evidence for a second excitation branch near 17 meV. The anisotropy of the correlations, predominantly occurring along a, is in agreement with the easy magnetic axis derived from the bulk susceptibility. The observation of two modes near the spin-gap edge in the KI regime, with different temperature and $\boldsymbol{Q}$ dependences bares some similarities with the situation observed previously in YbB_{12}, though it is too early to claim a common origin.

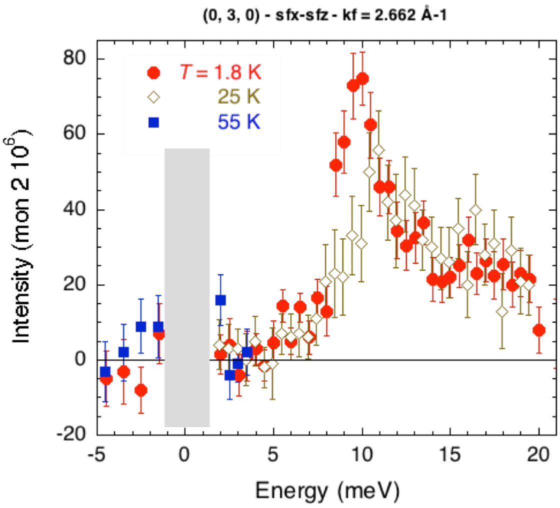


__Fig. 3.__ Temperature dependence of the magnetic signal SF_x–SF_z for $\boldsymbol{Q}$ = (0, 3, 0) (Y point).

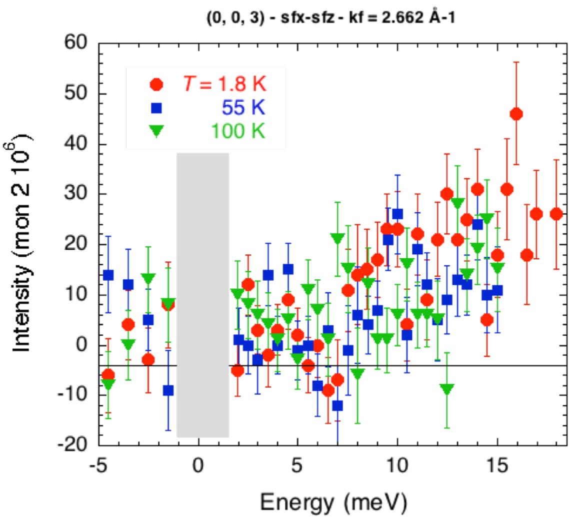


__Fig. 4.__ Temperature dependence of the magnetic signal SF_x–SF_z for $\boldsymbol{Q}$ = (0, 0, 3) (Γ point).

References

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