Experimental report

09/02/2018

Proposal: **4-01-1537** **Council:** 10/2016

Title: Magnetic-field dependence of the ferromagnetic excitation in the heavy-fermion metal CeB6

Research area: Physics

This proposal is a new proposal

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Samples: CeB6

Instrument	Requested days	Allocated days	From	To
THALES	6	4	20/02/2017	24/02/2017
IN12	6	0		

Abstract:

The heavy fermion material CeB6 has a complex phase diagram combining antiferro-quadrupolar (AFQ) and antiferromagnetic (AFM) phases, and its underlying physics is still poorly understood in spite of extensive studies. In our recent inelastic neutron scattering experiments, we have discovered several novel features in the magnetic excitation spectrum in the parent CeB6. Intense ferromagnetic (FM) low-energy collective mode, that dominates the magnetic excitation spectrum of CeB6 in the AFM phase, is suppressed by the weak magnetic field to zero together with the AFM order parameter. At higher fields inside the hidden-order AFQ phase this excitation reappears following the energy of an electron spin resonance (ESR). To fully understand the complex phase diagram, further INS measurements are essential. In this experiment we expect to find the second spin exciton seen in ESR experiment. We will observe the evolution of low-energy magnetic excitations at the FM wave vector, as a function of the applied magnetic field above 8 T, which can be directly compared with the existing ESR data on CeB6.

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Introduction

The heavy fermion material CeB_6 has a complex phase diagram combining antiferro-quadrupolar (AFQ) and antiferromagnetic (AFM) phases [1], and its underlying physics is still poorly understood in spite of extensive studies. Sharp resonant mode was initially revealed below T_N at the propagation wave vector of the AFQ phase [2], while a strong FM mode in the magnetic excitation spectrum of CeB_6 at the zone center Γ was discovered later [3]. Magnetic field dependence of the FM resonance was measured at different instrument up to 14.5 T. It is initially suppressed with the magnetic field within the AFM phase, but reappears upon entering the AFQ phase, following the energy of an electron spin resonance (ESR). ESR measurements, which probe zone-center excitations, have shown that the frequencies of the two observed resonances A and B [4] change linearly with field within phase II. We compared resonance energies obtained from ESR with the field-dependent energy of the zone-center INS excitation and realized, that its energy matches that of the A resonance seen in ESR. The second ESR line observed in high fields (mode B) was interpreted as the result of a crossover of the excited state to the free-ion limit, as the field at which it appears is comparable with the condensation energy of the AFQ phase, $\sim 1.75 k_B T_Q$ [5].

Our last experiments at FLEXX spectrometer showed the absence of the second resonance in the INS spectrum. We realized that at higher fields above 8 T, excitation starts to deviate towards lower energies. Results observed during the experiment questioned the accuracy of the magnetic field and temperature calibration (see experimental report No. 17105302). Since we observed signal at the position which was not in accordance with the predicted, our first assumption was that the field calibration is wrong. In order to check this hypothesis, as well as to reveal second spin exciton, seen in ESR experiment, we performed Thales experiment.

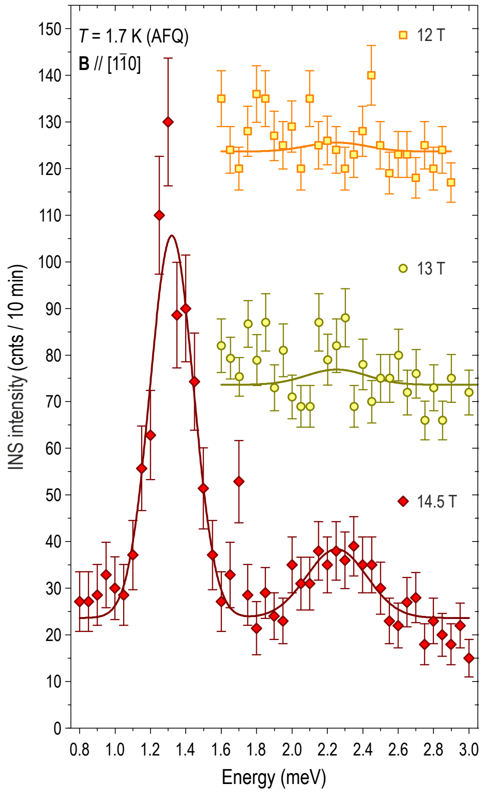

Fig. 1: INS spectra measured at Thales, near the zone center $\Gamma(110)$, which shows the appearance of the second resonance. The spectra are shifted vertically for clarity

Experimental configuration

Measurements were performed on single-crystalline sample of CeB_6 with a mass of 4 grams, prepared from 99.6 % isotopically enriched ^{11}B to minimize neutron absorption. The sample was mounted in the cryomagnet with a maximal field of $B = 15$ T with its crystallographic $\langle 1\bar{1}0 \rangle$ axis aligned vertically. Using the published lattice parameters $a = b = c = 4.137$ Å, $\alpha = \beta = \gamma = 90°$ and measured in-plane reflections we aligned on the most intense (110) and (001) reflections.

Results

As a first step we tried to repeat same 14.5 T scan, as we did at FLEXX, to check if the excitation's energy can be reproduced. Luckily, the energy of the resonance within the errorbar was the same at both experiment. However, to our great surprise, we realized that the signal intensity at FLEXX ant Thales experiments was exactly the same. Despite the fact that the reactor is ~ 2.5 times more powerful, background level as well as the signal itself could be averaged just like it was measured on a same instrument. This seriously worried our experimental team. Luckily, one day later we found the reason: previous users closed the virtual source slit before the monochromator to 3.5 mm, instead of 35 mm and forgot to tell us. After changing back the slit size the status quo was restored, and as expected the intensity increased almost ~ 3 times. As compensation for partially lost time, as well as as a bonus, since detailed mapping (see text below) took more time then we expected, we received two additional days, which were originally intended to be used for internal tests.

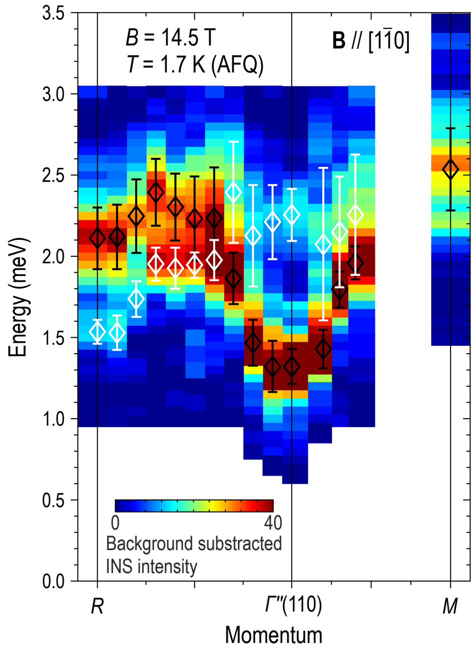

Fig. 2: Energy-momentum profile, which shows dispersion of the magnon spectrum along straight segments connecting the $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, $\Gamma(110)$, $M(\frac{1}{2}\,\frac{1}{2}\,0)$ points.

Immediately after this, we managed to observe the onset of the second branch. To our surprise, energy of the second resonance was mach higher than we expected, and did not overlap with the second resonance observed by ESR. Since we were restricted by the maximal available field of 14.5 T on Thales, we followed field dependence of the second excitations by lowering the field. As shown in Fig. 1, second mode appears at fields above 12 T. Its strong field dependence indicates that it is magnetic.

Another question, that we tried to answer during this experiment was bandwidth determination. Within the AFM phase a strong FM mode at the zone center is hybridized together with the maximum of intensity at the R point and the spin-wave modes emanating from the AFM wave vectors. Together they form a continuous dispersive magnon band in a narrow energy range between 0.2 meV and 0.7 meV [3]. A magnetic field of 2.5 T does not change the excitation energy at the zone center significantly but increases the magnon bandwidth twofold, as the dispersion now reaches ∼ 1.4 meV at the M point in contrast to 0.7 meV in zero field [7]. Assuming linear dependence for the M we can expect that the bandwidth will also increase significantly for the high field. Not the least, we wanted to see if there is a connection between the second resonance, which we found at the Γ point, with the previously known excitations at the R (0.5 0.5 0.5) point. To check this we did detailed mapping of the magnon dispersion at the maximum avaliable field of 14.5 T, as shown in Fig. 2. As expected at the R point we found both resonance that follows previously observed field dependence [7], but the peak at 2.5 meV observed at the M (0.5 0.5 0) point suggest that the bandwidth remains more or less constant within the AFQ phase.

We also tried to study dependence of the g-factor as a function of momentum. Combining our last experiments at FLEXX and Thales triple-axis spectrometers, measured at the zone center Γ, we see that the previously observed excitation that followed the energy dependence of the A resonance starts to deviate slightly towards lower energies, and the corresponding g-factor decreases to the value of 1.56 shown with orange solid line in Fig. 3(a). Previous measurements of the resonance peak at the R point showed that increasing the field within the AFM phase keeps the resonance energy constant while it decreases in amplitude and broadens, transferring a significant part of its spectral weight to the second low-energy mode whose tail can be seen above the elastic line already above ∼ 0.5 T. Upon crossing through the phase III–III′ transition, the amplitude of the low-energy mode is maximized, whereas the higher-energy mode shifts up in energy. Both excitations then follow a linear trend with the same slope in phase II, in agreement with our earlier report of g-factor of 1.90 at the R point [8]. In this experiment we extended observed dependence up to 14.5 T, and we can confirm that the linear dependence persists, as shown in in Fig. 3(b). At the M point, we observe similar behavior but with the g factor of 1.65 only (within the AFQ phase), as shown in in Fig. 3(c). These results clearly demonstrate significant changes in the g-factor as a function of momentum.

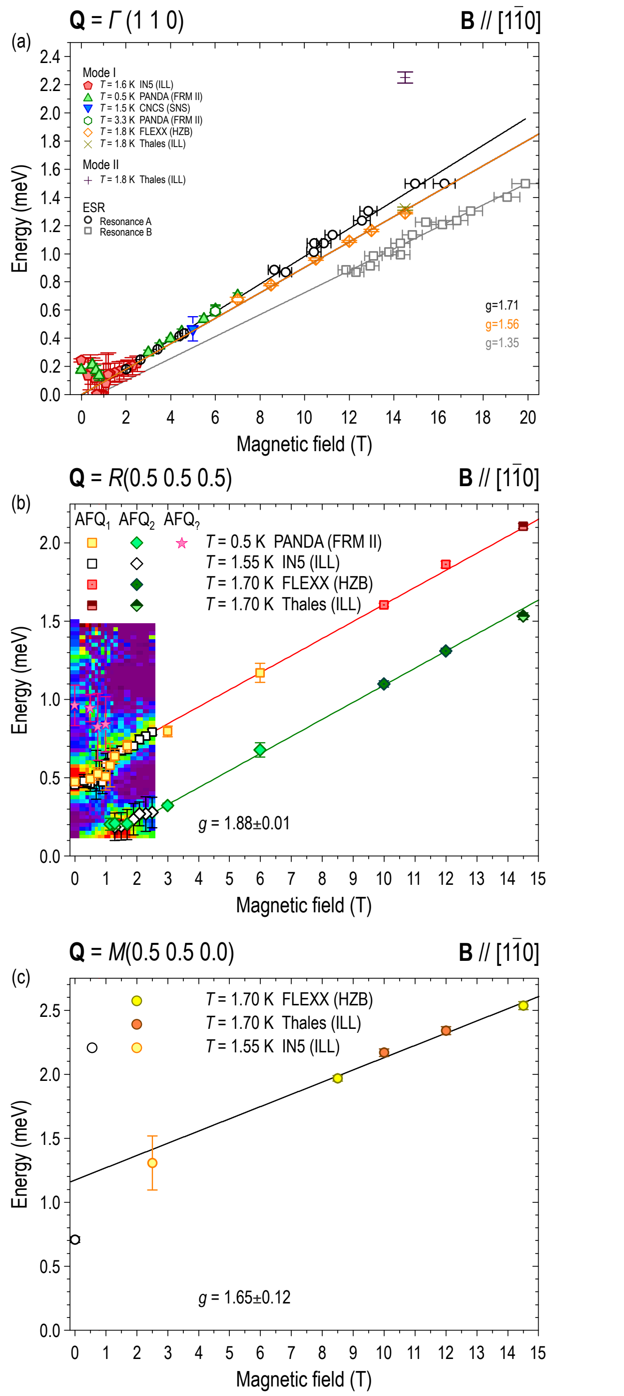

Fig. 3: (a) Summary of the magnetic field dependence of zone-center excitations obtained from both INS and ESR spectra. Solid lines are linear fits of resonances A, B, and INS data. (b) Summary of the magnetic field dependence of the $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ and (c) $M(\frac{1}{2}\,\frac{1}{2}\,0)$ points.

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