Experimental Report

Proposal: **4-01-1064** **Council:** 4/2011

Title: Investigating the dependence of the spin gap and of the spin excitonsin the ground state of CeB6 on theexternal magnetic field

This proposal is continuation of: 4-01-912

Researh Area: Physics

Main proposer: **INOSOV Dmytro**

Experimental Team: JANG Hoyoung
INOSOV Dmytro
FRIEMEL Gerd
PORRAS PEREZ GUERRERO Juan Pablo
GHAMBASHIDZE Giorgi

Local Contact: IVANOV Alexandre

Samples: CeB(6) with 99.6 at. % isotope-enriched boron-11

Instrument	Req. Days	All. Days	From	To
IN14	10	5	04/12/2012	09/12/2012

Abstract:

Among the RB6 family of hexaborides (R being a rare earth), CeB6 stands out as the only compound that exhibits an exotic ordered phase in the narrow temperature range between 2.3 and 3.2 K, commonly referred to as an antiferroquadrupolar (AFQ) phase. Its true nature is not yet fully understood and is discussed controversially. In our recent experiment performed on the same sample at IN14, we have observed that the AFQ phase survives down to the lowest temperatures, where it coexists with the antiferromagnetic (AFM) order. We found new, very intense, resonance-like spin excitons in the ground state, maximized at the AFQ wavevector. Here, we propose to investigate the field-dependence of these new excitations at the lowest temperatures, far below the Neel transition (2.3 K), as it is known that the AFQ phase is strongly enhanced by the magnetic field, while the AFM order is suppressed by relatively small fields of the order of 1 Tesla. This experiment will allow us to pinpoint the microscopic origin of the excitons. We suggest to perform this experiment at the IN14 cold-neutron spectrometer on the same 8g single-crystalline sample grown from a 99.6 at. % isotope-enriched 11B powder.

Introduction.

The heavy fermion compound CeB_6 is known for exhibiting antiferroquadrupolar (AFQ) order (phase II) below $T_Q = 3.2\,K$ with the wave vector $\mathbf{q}_{AFQ} = R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$. This type of order can be understood in a localized model [1]. However, the compound also exhibits antiferromagnetic (AFM) ordering below $T_N = 2.3\,K$ with wave vectors $\mathbf{k}_1 = \Sigma(\frac{1}{4}\,\frac{1}{4}\,0)$ and $\mathbf{k}'_1 = S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$, which was not investigated by inelastic neutrons scattering so far. Our recent observation of a exciton-like mode at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ and $E = 0.5\,meV$ in this state indicates that an itinerant description might be more appropriate [2]. The exciton bears similarity to the resonant mode frequently observed in the superconducting state (SC) of unconventional superconductors [4]. The AFM state can be suppressed by applying an external magnetic field, upon which the system returns to the AFQ state.

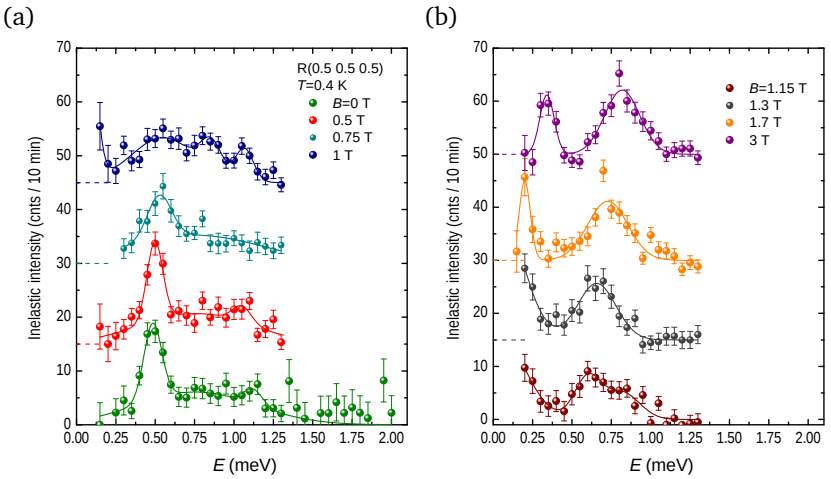

Fig. 1: **(a),(b)** Spectrum at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ for different magnetic fields at base temperature in CeB_6. The spectra are successively shifted by 20 units for clarity.

For this experiment we proposed to study the magnetic field dependence of the exciton mode, which we expect to become suppressed parallel to the AFM phase [3]. From our previous experiments we know that AFM phase vanishes at $B_c = 1\,T$, whereas the weak AFQ Bragg intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ [2] is field independent at low fields and then increases linearly with field above $B_q = 1.7\,T$.

In the meantime before the scheduled experiment we could obtain a Lanthanum-doped sample ($Ce_{1-x}La_xB_6, x = 0.28$). The La does not carry any f-electron and therefore suppresses the AFQ and AFM order. However, transport and magnetization showed the emergence of a presumably antiferro-octupolar (AFO) phase above the La-concentration of $x = 0.2$ [5], with an ordering wave vector of $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ as well [6]. This phase is sandwiched between AFM phase and paramagnetic phase in zero field. We have characterized the sample by specific heat, showing a strong anomaly at the transition temperature between the paramagnetic and the AFO state. Part of this beam time was used to study the magnetic field dependence of the AFO order parameter. We also checked the spectrum in the AFM state at lowest temperature at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ to look for the spin exciton mode.

The sample was mounted in a 10T asymmetric vertical cryomagnet. Lowest temperatures of $T = 0.1\,K$ could be accessed via a dilution insert. Both CeB_6 and $Ce_{1-x}La_xB_6$ are cubic with almost equal lattice parameter of $a = 4.14\,Å$. The rodlike samples were mounted vertically, so that the field is oriented along the rod axis $(1\bar{1}0)$ and the perpendicular (HHL)-vectors are lying in the scattering plane. The final neutron wave number was chosen to $k_f = 1.3\,Å^{-1}$.

Experimental results for CeB_6

First, we measured the field dependence of the spectrum at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$. The exciton in zero-field at $E = 0.5\,meV$ is suppressed in intensity in a field of $B = 1\,T$ (Fig. 1) and at the same time shifts towards higher energies. Above the critical field $B_c = 1\,T$, a new mode appears around $E = 0.65\,meV$, which reaches into the AFQ state at $B = 3\,T$. Here, an additional mode at $E_2 = 0.3\,meV$ enters the spectrum. The energy of the AFQ modes depends on the field strength. Therefore, measuring them at higher fields would allow a comparison with the previous measurements of Bouvet [7], who mapped out the AFQ excitations in a field of $B = 6\,T$ for various wave vectors in the Brillouin zone.

Experimental results $Ce_{1-x}La_xB_6$

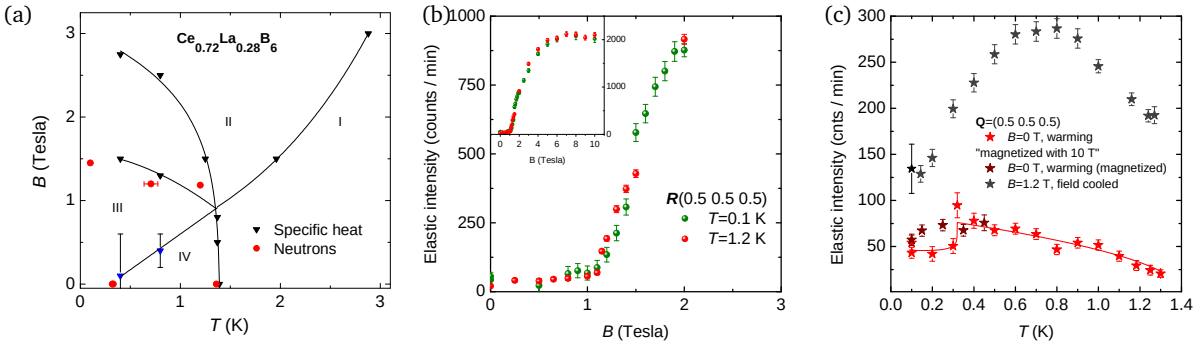

Fig. 2: **(a)** B-T-phase diagram with data points marking the phase transition observed by specific heat or by the elastic neutron intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$. **(b)** Field dependence of the Bragg intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ at $T = 0.1\,K$ and $T = 1.2\,K$. **(c)** Temperature dependence of the Bragg intensity at R in zero field and $B = 1.2\,T$. The former was measured in field cooling and zero field cooling mode.

In order to understand the relation between the exciton mode and the AFM phase better, we changed the sample to a La-doped compound $Ce_{1-x}La_xB_6$ ($x = 0.28$). According to the magnetic field phase diagram in Fig. 2 (a), which is based on specific heat measurements, the AFM transition temperature is suppressed to $T_N \approx 0.3\,K$. At higher temperatures, between $T_N < T < 1.4\,K$, the AFO order (phase IV) exists. The $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ shows Bragg intensity and can be identified as the order parameter for this phase [6]. We measured the field dependence of the Bragg intensity at two temperatures $T = 0.1\,K$ and $T = 1.2\,K$, shown in Fig. 2 (b). Both curves are very similar showing an almost constant intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ up to a field of $B = 1.1\,T$. The rapidly increasing intensity for higher fields marks the AFQ phase consistent with the observation for the parent compound CeB_6. Such an increase is not observed in the AFO phase, which exists at $T = 1.2\,K$.

The temperature dependence, shown in Fig. 2 (c) of the $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ Bragg intensity also reveals interesting aspects. In zero field it apparently becomes slightly suppressed below T_N, pointing to a competition between AFO and AFM phase, similar to the observation at the AFQ-AFM phase transition in CeB_6[2]. This effect is even stronger when crossing the phase boundary between AFQ and AFM phase in field of $B = 1.2\,T$. Here, the intensity (grey symbols in Fig. 2 (c)) is halved. A similar behaviour was observed in the parent compound [8]. However, these experiments used a coarser energy resolution ($k_f = 2.662\,Å^{-1}$), whereas the scattering in our experiment was truly elastic. Future experiments should complement the temperature dependence of the Bragg intensity to higher temperatures, to observe the onset temperatures $T_Q(B)$ and $T_O(B)$ of the AFQ and AFO phase, respectively.

The AFM phase is specified by a magnetic ordering with the wave vector $\Sigma(\frac{1}{4}\,\frac{1}{4}\,0)$ and $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$ in the parent compound. We searched for magnetic intensity at these point in $Ce_{1-x}La_xB_6$, but could not find Bragg peaks. Since the specific heat measurements clearly show the transition into an AFM state we suspect that the magnetic order is incommensurate with the corresponding wave vectors shifted out of the scattering plane.

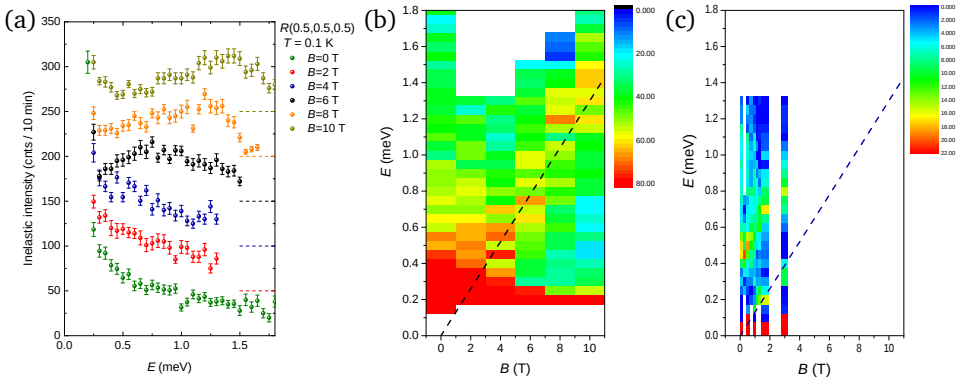

Fig. 3: **(a)** Spectra at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ for various external fields in $Ce_{1-x}La_xB_6$, each spectrum is shifted by 50 units with respect to the preceding spectrum for clarity. **(b)** Colormap of intensity for the $E-B-$parameter space covered by the measurements in (a). **(c)** Equivalent colormap intensity for the $E-B-$parameter space measured for CeB_6.

Next we measured the spectrum at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ to see how the exciton mode changes upon doping. Figure 3(a) shows spectra for different external fields. In zero field the exciton is clearly replaced by a broad quasielastic response. For higher fields this lineshape develops a shoulder and a broad peak appears at $B = 4\,T$, centered at $E = 0.5\,meV$, which seems to move to the right, for the highest applicable field of $B = 10\,T$. Plotting this data in a two-dimensional color map in Fig. 3(b) over the $E-B-$parameter space shows that this mode energy scales linearly with magnetic field reaching $E = 0$ in zero field, signified by the dashed line. This is similar as observed for the mode $E_2(B)$ in CeB_6 in Fig. 1. There, it is much sharper in energy but follows the same scaling *vs.* field as shown in the color map in Fig. 3(c). It is interesting to note that the AFQ mode at $E_1 = 0.8\,meV$ is either absent or much broadened in $Ce_{1-x}La_xB_6$, so that it overlaps with the E_2 mode.

In conclusion, the spin exciton in CeB_6 shows a rather complex behaviour in magnetic field, getting suppressed in intensity and enhanced in energy. In the AFQ state multiple modes start to appear, where the lowest mode scales linearly with magnetic field. This complex behaviour delivers input for theoretical description of the AFM and AFQ states. A continuation of the mapping of spin excitations in higher fields is strongly required. The results on $Ce_{1-x}La_xB_6$ ($x = 0.28$) offer new insights into the complex electronic phase diagram, which is extended with the AFO phase (phase IV). It seems that the AFM phase is competing with both AFQ and AFO phase, as inferred by the temperature dependence of the intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, the wave vector of the order parameter for both AFQ and AFO phase. No spin exciton mode is observed in the spectrum at R. Instead we observe the same low energy AFQ mode as observed in CeB_6, but strongly broadened. No AFM Bragg peak could be observed near $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$ or $\Sigma(\frac{1}{4}\,\frac{1}{4}\,0)$. An investigation, whether it is truly absent or shifted to an incommensurate position, should be addressed in future experiments.

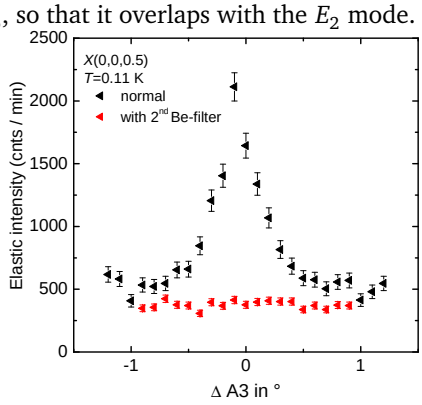

Fig. 4: Rocking scan in elastic channel through $\mathbf{Q} = (0\,0\,\frac{1}{2})$, which shows second order contamination. Applying the 2nd (hot) Be filter in the incoming beam can suppress these second order neutrons.

Comments on experimental conditions

The technical support at ILL is generally of high level and the majority of our experiments in the past were conducted under stable conditions, e.g. sample environment and instrument control. However, in this beam time we faced several technical difficulties which led to a considerable loss of beam time. The standard Be-filter at In14 happened to be defect at the time of the experiment. Another Be-filter was installed which suppressed 2nd order scattering insufficiently as shown in Fig. 4. We detected scattering at $(0\,0\,\frac{1}{2})$, which is not a superstructure reflection. The 2nd order scattering also contaminated the superstructure peak at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, measuring which we spent a large amount of time. We could suppress this by putting an additional piece of hot Be in the incoming beam, later in the experiment.

However, this reduced the flux by at least 25%. Consequently, we had to rescale the data taken without 2nd filter, which introduced unnecessary error in the data. Due to the strong absorption of the incoming beam, we had to remove the hot Be filter before each inelastic scattering measurement. We estimate that at least half a day was lost because of this problem, as we had to run several test and calibration measurements that were not planned initially.

Another complication was the unstable temperature, set by the dilution refrigerator, enabling us to measure within the AFM phase at $T = 0.1\,\mathrm{K}$. During the beam time an outage of cooling occurred with a subsequent rise of temperature to 1.3 K, which made the removal of the refrigerator insert necessary on the next day. The check and reinsertion plus cooling cost 1.5 days of beam time. In total, we would like to ask for consideration of 2 days of beam time compensation, which can be connected to the IN14 experiment 4-01-1251, scheduled in the upcoming cycle (Februar 2013-July 2013).

[1] H. Shiba *et al.*, J. Phys. Soc. Jpn. **68**, 1988 – 1993 (1999); F. J. Ohkawa *et al.*, J. Phys. Soc. Jpn. **54**, 3909–3914 (1985).
[2] G. Friemel *et al.*, Nat. Commun. **3**, 830 (2012).
[3] T. Fujita *et al.*, Solid State Commun. **35**, 569 – 572 (1980).
[4] D. S. Inosov *et al.*, Nat. Phys. **6**, 178–181 (2010).
[5] S. Kobayashi *et al.*, J. Phys. Soc. Jpn. **72**, 2947–2954 (2003).
[6] K. Kuwahara *et al.*, J. Phys. Soc. Jpn. **76**, 093702 (2007).
[7] A. Bouvet, PhD thesis, Université Grenoble, (1993).
[8] V. P. Plakhty *et al.*, Phys. Rev. B **71**, 100407 (2005). M. Sera *et al.*, Phys. Rev. Lett. **86**, 1578–1581 (2001).