Experimental Report

15/07/2013

Proposal:	**4-01-1251**	**Council:**	10/2012

Title: Quantum criticality and suppression of the exciton mode in Ce{0.72}La{0.28}B{6}

This proposal is a new proposal

Research Area: Physics

Main proposer: **JANG Hoyoung**

Experimental Team: INOSOV Dmytro
FRIEMEL Gerd
JANG Hoyoung
GHAMBASHIDZE Giorgi
TSENG Kuo Feng

Local Contact: IVANOV Alexandre
PIOVANO Andrea

Samples: Ce(0.72)La(0.28)B(6) with isotopically enriched 11B

Instrument	Req. Days	All. Days	From	To
IN14	12	9	04/03/2013	13/03/2013
IN3	2	2	28/02/2013	02/03/2013

Abstract:

We have recently reported the discovery of a magnetic exciton mode in the heavy-fermion antiferromagnet CeB6, whose behavior bears similarities with that of the spin resonant mode in superconductors. We have argued that this mode, originating from fluctuations of the antiferroquadrupolar (AFQ) order parameter, is stabilized due to the opening of a gap in the particle-hole continuum associated with the antiferromagnetic (AFM) order that sets in below T_N = 2.3 K at a different wave vector. We suggest to verify this interpretation with measurements of a similar Ce{0.72}La{0.28}B{6} compound, in which partial dilution of the Ce sublattice by nonmagnetic La ions suppresses the AFM transition. The AFM order can be restored by applying magnetic field, 0 < B < 2.3 T, along the <110> direction, beyond which it is suppressed again, giving way to the AFQ phase. This represents ideal conditions for checking experimentally if the exciton mode is indeed linked to T_N. In addition, we would be also interested in establishing the so-called E/T scaling of dynamical susceptibility close to the quantum critical point.

Introduction.

The heavy fermion compound CeB_6 is known for exhibiting antiferroquadrupolar (AFQ, phase II) order 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 order (AFM, phase III) below $T_N = 2.3\,K$ with the propagation vectors $\mathbf{k}_1 = \Sigma(\frac{1}{4}\,\frac{1}{4}\,0)$ and $\mathbf{k}'_1 = S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$. In this phase the Fermi surface becomes gapped leading to a formation of an exciton-like mode at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ and $E = 0.5\,meV$ as observed in our recent inelastic neutron scattering study [2]. This mode, resembling the resonant mode in the superconducting state (SC) of high-T_c superconductors, indicates that the contribution of the conduction electrons to the spin dynamics should not be neglected.

In magnetic field the AFM phase is suppressed as determined by the vanishing of the Bragg peak $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$ around $B_c = 1\,T$. The AFQ phase sets on at $B_Q = 1.7\,T$, signified by a linear increase of the Bragg intensity at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$. We observed that the exciton mode vanishes as well, however, its energy evolves in a continuous fashion and the intensity shows reentrant behaviour upon reaching the AFQ state [3]. Another tuning parameter is the dilution of the magnetic Ce^{3+} ions with non-magnetic La^{3+} in $Ce_{1-x}La_xB_6$ solid solutions, upon which phase II and phase III become suppressed [4]. By means of neutron scattering we wanted to determine the $H-T-$phase diagram by measuring the field dependence of the respective order parameters. Furthermore, we wanted to investigate the doping evolution of the exciton mode, which, together with the scaling in magnetic field, would give important insight to the interaction leading to its formation. In our previous beam time we measured a $x = 0.28$ substituted sample, where the exciton mode was found to be absent. Also we could not detect any AFM Bragg peak at $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$. In this study we chose the doping level slightly lower ($x \approx 0.18$), so that the AFM phase is present and still resembles the undoped compound [6]. Again, we measured the field and temperature dependence of the AFM and AFQ order parameters, at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ and $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$, to verify the established magnetic phase diagram. In parallel, we measured the field dependence of the exciton mode at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, similar to the study done in the undoped compound [3].

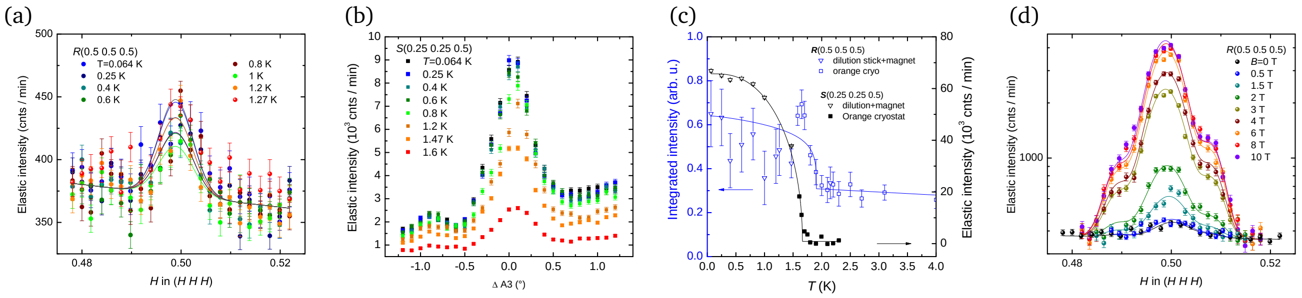

Fig. 1: (a) Longitudinal scans through $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ for different temperatures. **(b)** Rocking scans through the AFM Bragg peak $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$ for different temperatures. **(c)** Temperature dependence plot of background-corrected integrated intensity of the AFQ and AFM Bragg peaks. **(d)** Longitudinal scans through $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ for different magnetic fields, $\mathbf{B} \parallel (1\,\bar{1}\,0)$.

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 parameters 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 be $k_f = 1.3\,Å^{-1}$.

Field dependence of AFQ and AFM order parameter

Since La-doped CeB_6 has been rarely studied by elastic neutron scattering, we first checked the existence of the AFQ and AFM parameters. Both Bragg peaks could be observed as a peak in longitudinal scans through $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ [see Fig. 1(a)] and in rocking scans through $S(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{2})$ [see Fig. 1(b)]. We checked their temperature [Fig. 1 (a),(b)] and field dependence [Fig. 1 (d)].

Fig. 1 (c) shows the integrated intensity for both parameters *vs.* temperature. Both show an onset at T_Q and T_N, respectively. The AFQ intensity in Fig. 1 (c) has a constant offset for $T > T_Q$, which is due to a 2nd order contamination of the incoming neutrons. The ratio $I_{AFQ}/I_{AFM} = 300$ between AFM and AFQ intensities is comparable to the undoped compound. In order to outline the magnetic phase diagram of this compound, we also measured the AFQ and AFM Bragg intensity as a function of magnetic field by performing the same momentum scans while changing the field in steps. These measurements were repeated for the AFQ Bragg peak at several higher temperatures and are shown in Fig. 2 (a). The AFM Bragg peak becomes suppressed upon application of magnetic field and van-

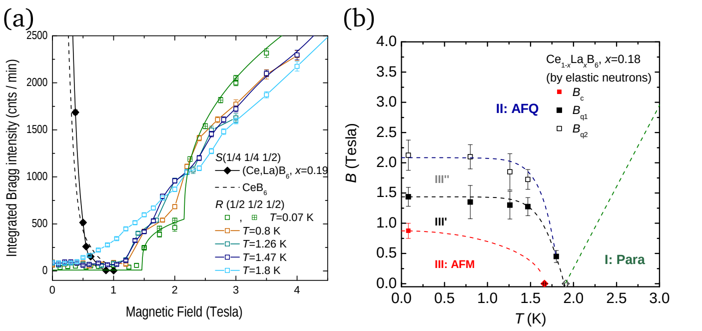

Fig. 2: (a) Field dependence of the AFM and AFQ Bragg peak amplitudes. The latter was measured for several temperatures. **(b)** Phase diagram of $Ce_{1-x}La_xB_6$ showing the transition points determined from the transition temperatures in Fig. 1 (c) and transition fields in Fig. 2 (a).

ishes around $B_c = (0.88 \pm 0.13)$ T. Interestingly, as seen in Fig. 2 (a), the AFQ intensity (green curve) does have an onset at a distinct higher field of $B_{q1} = (1.44 \pm 0.16)$T. We associate the intermediate range $B_c < B < B_{q1}$ with phase III′ [7].

Different to the parent compound there seems to be another phase transition at $B_{q2} = (2.13 \pm 0.25)$T, reflected by a kink in the $I_{afq}(B)$-curve [Fig. 2 (a)]. This additional kink, which is absent in CeB_6, is also observed for the other temperatures. We cannot say what kind of order parameter is present in this field range $B_{q1} < B < B_{q2}$ and denote it as phase III″. Comparison with other experimental techniques for the same doping level might clarify if the nature of this new phase is intrinsic.

These qualitative new observations show that the transition between phase III and II in field fractionalizes in 3 intermediate phases, which cannot be explained by domain formation. Above B_{q2} it seems that the true AFQ order is present. We measured the AFQ Bragg intensity up to $B = 10$ T, where it saturates, similarly to the $x = 0.28$ doped sample [5].

We summarized all transition temperatures and fields in the phase diagram in Fig. 2 (b).

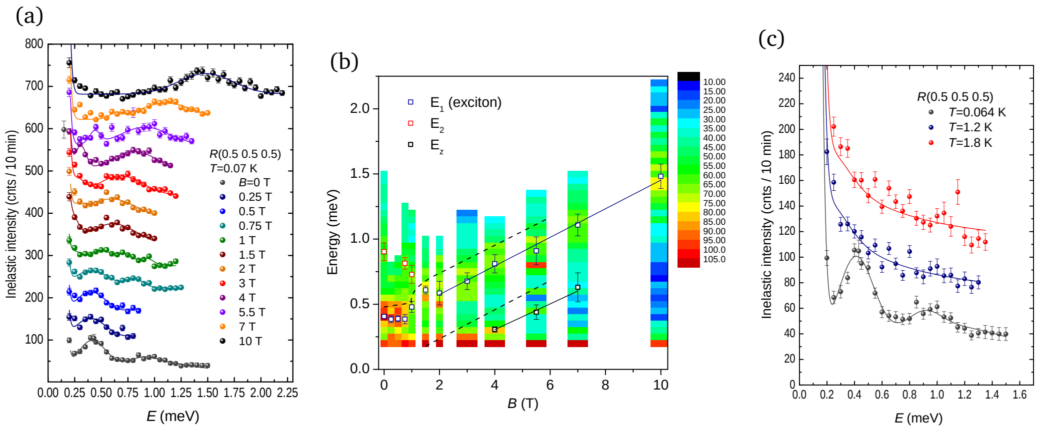

Fig. 3: **(a)** Spectra at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$, measured at base temperature and in different magnetic fields $\mathbf{B} \parallel (1\,\bar{1}\,0)$. The curves are shifted by 60 counts for clarity. **(b)** Colormap representation of the $E-B$-parameter space covered in (a). The symbols correspond to the center position of the Gaussian fits to the peaks in the spectra of (a). The dashed lines are the corresponding $E_i(B)$ functions, determined for the parent compound CeB_6[3]. **(c)** Spectra at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ in zero field for different temperatures.

Spin exciton and its field dependence at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$

One of the central objectives was to measure the spin exciton mode in this doped compound. Fig. 3 (a) shows the raw energy scans at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ for different magnetic fields $\mathbf{B} \parallel (1\,\bar{1}\,0)$. In zero field a clear excitation is visible at $E_1 = 0.41$ meV, which we associate with the spin exciton mode. There is another excitation visible at $E_2 = 0.91$ meV. Upon applying magnetic field the exciton mode energy is slightly enhanced, whereas E_2-mode is suppressed in field. Both merge at $B = 1.5$ T. Interestingly, the intensity of the spin exciton is only slightly suppressed towards the critical field $B_c = 0.88$ T, contrary the clear decrease of intensity, observed for the undoped compound. Above $B_{q2} \approx 2$ its energy scales linearly with magnetic field. For CeB_6 we could not obtain that many data points for $B > 3$ T, but if we fit a linear increase in the corresponding field range (dashed line), its slope is almost equal for undoped and x=0.18 doped compounds. However, the energy seems to be offset by a constant value compared to CeB_6. Also the onset of the linear scaling seems to be already happening at $B = 1$ T, close to the critical field of the AFM phase. Only the overlapping with the approaching E_2 mode causes this shoulder around $B = 1.5$ T. Also we could discern a mode E_3 which appears at low energies for $B > 2.5$ T, increases in energy, and quickly loses intensity towards higher fields. A similar mode was also observed in CeB_6[5]. Its $E_3(B)$ relation is indicated by the lower dashed line.

We also measured the temperature dependence of the exciton mode in zero field. Fig. 3(c) shows the spectra in zero field. When reaching the AFQ state the spectrum takes a quasielastic lineshape, in accordance with the observations made in the undoped compound [2].

Summary

The $Ce_{1-x}La_xB_6$(x=0.18) compound studied here is a crucial doping level, sharing properties with the undoped compound like the low temperature AFM phase, as well as the spin exciton mode at $R(\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$. However, there are also differences like the shift in the mode energy and the existence of a phase III″ as seen by the kinks in the field dependence of the AFQ Bragg intensity. Together with the transition temperatures we could outline the $B-T$-phase diagram, which can be compared with the phase diagram determined from transport and magnetization measurements [8].

As for higher La-doping the phase diagram gets more complicated with the appearance of phase IV, it is crucial to first clarify the spin dynamics of phase III and phase II for the moderately doped compounds, where the contribution of the conduction electrons were shown to play a role, but were omitted in persisting models of the system [1].

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