ESRF	**Experiment title:** Observation of antiferroquadrupolar ordering in CeB_6	**Experiment number**: HE-594
Beamline: ID20	**Date of experiment**: from:April 28, 1999 to:May 3, 1999	**Date of report**: 4/09/2000
Shifts: 17	**Local contact(s)**: F. Yakhou	*Received at ESRF:*

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Report:

CeB_6 is known as a dense Kondo lattice in which the Kondo effect competes with exchange interactions. Its unusual phase diagram shows three phases at zero field: the paramagnetic phase above T_Q=3.3 K, an antiferromagnetic state below T_N=2.3 K that has been clearly identified by neutron diffraction and an intermediate phase which displays a field-induced antiferromagnetic component with wavevector (1/2 1/2 1/2). It was attributed to antiferroquadrupolar ordering but no direct experimental evidence was given so far.

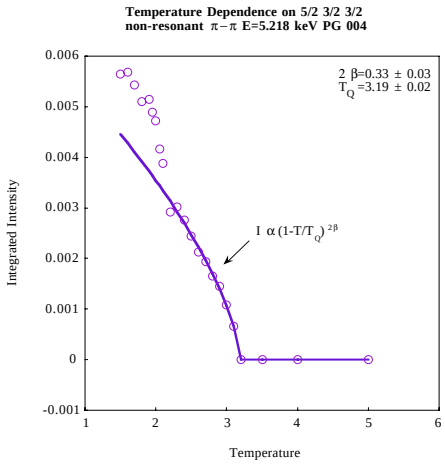
X-ray scattering should be sensitive to the asphericity of the 4*f* electronic distribution and superstructure reflections arising from the broken translational symmetry of these *f*-orbitals should appear. The 4*f* form factor has been calculated and the (1/2 1/2 1/2) wavevector appears to be the most favourable. The proposal was originally intended to measure those reflections at 20 keV in transmission geometry to lessen the effects of beam heating and maximize the reciprocal space accessibility.

Several unsuccessful attempts have been run on ID20 as in-house research (IHR) before the HE-594 experiment was carried out in May 99. This experiment had to be completed in July 99 and March 00 (IHR). Two additional attempts failed, once because of a cryostat failure and the second time due to a poor surface state.

Two single crystals with [100] (A) and [110] (B) surfaces were prepared at CENG with a good surface state (mechamical polishing down to 6 mm). They were mounted on the end cone of a cylindrical Cu holder and glued with Wood metal to insure a good thermal contact. The standard "orange" ^{4}He cryostat was cooled to its base temperature (1.45 K).

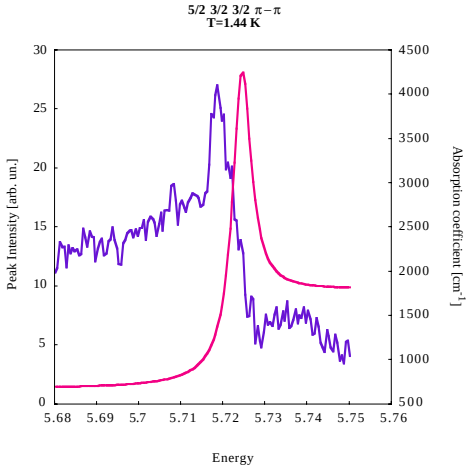
The experiments were performed in horizontal scattering geometry at 20, 6.164 (Ce LII absorption edge) and 5.73 keV (Ce LIII edge). A LiF 220 crystal was used as polarisation analyser at both edges. The crosstalk corrections between the two polarisation channels in each case did not execede 3%.

Four reflections with wavevector (1/2 1/2 1/2) were found: (the 3/2 3/2 1/2) on crystal B and the (5/2 1/2 1/2), (5/2 3/2 3/2) and (7/2 1/2 1/2) on crystal A. They all display similar temperature dependences (Fig. 1) with a second-order-like phase transition around 3 K and an enhancement of the intensity around 2 K, that we attribute to the appearance of the antiferromagnetic order at T_N=2.3 K. The temperature at which the signal disappears strongly depends on the attenuation factor indicating strong beam heating effects. Indeed, the beam had to be attenuated by a factor 150 at 200 mA for the reflection (3/2 3/2 1/2) to appear.



The peak intensities as a function of energy were measured for each of these reflections in both polarisation channels π–π and π–σ, below and above TN. They all display resonant features, with a marked difference for the (5/2 3/2 3/2): it is the only one for which a considerable non-resonant contribution was observed (Fig. 2). From the position in energy of the resonant peaks, it appears that both dipolar and probably quadrupolar contributions are present. The same features were observed below T_N.

To confirm the occurence of a non-resonant signal, further measurements were performed at 5.218 keV that matches exactly the graphite (004) polariser energy. The (5/2 3/2 3/2) reflection is present in the π–π channel while the (5/2 1/2 1/2) appears only and weakly in the π–σ channel. These were the only accessible reflections at this energy.



No signal at all could be detected at 20 keV. Scans were also performed at the reciprocal lattice points corresponding to the antiferromagnetic phase. No intensity was detected either.

These results remain yet to be interpreted though the peculiar temperature and *q*-dependences of the observed intensities are strongly reminiscent of 3*d* orbital and 4*f* quadrupolar ordering. Further experiments are required to observe the known antiferromagnetic structure and compare the intensities to the above-measured ones. Special care should be taken for the sample mounting to insure the best possible thermal contact. A definitive answer as to the occurence of the quadrupolar ordering could be given by the azimuthal dependence of the resonant intensities but it requires the development of a special device allowing these measurements in an “orange” cryostat. This ideally opens the way to a long-term proposal.