Observation of Field Induced Magnetic Octupole in CeB_6

By resonant X-ray diffraction, it has been verified that magnetic octupole moment is induced in the Ce-4*f* orbital of CeB_6 by magnetic fields. The clue to the successful detection was the asymmetric field dependence of the E2 resonance intensity when the field direction was reversed. The observation provides firm evidence for the theory of mysterious antiferro-electric-quadrupole ordered phase of CeB_6, which has interested researchers for more than 30 years.

Most magnetic materials become magnetized when a magnetic field is applied. This state is microscopically described as being that a magnetic dipole moment on each magnetic ion is orientated to the field direction. However, in a rare-earth compound CeB_6, it has been theoretically predicted that quite an exotic magnetization state appears (inset of Fig. 2(b)). This type of magnetic moment is called a magnetic octupole (MO), representing an anisotropic magnetization density, which may be classically illustrated as consisting of four N and four S poles. Although MO does not contribute to the net magnetization, the theory states that the inter-ionic interaction between MOs plays an important role in various macroscopic properties of CeB_6 [1].

The appearance of MO in CeB_6 has its origin in the antiferro electric quadrupole (AF-EQ) order (inset of Fig. 2(a)), where an anisotropic charge density is ordered in an alternating arrangement. When a magnetic field is applied here, the magnetization density in a Ce ion will become anisotropic because the magnetization appears only where the charge density is. The magnetization in the area with no charge density may effectively be considered as oppositely directed. This simplified picture may capture the essence of MO. To be accurate, however, the MO in Fig. 2(b) is an independent quantum mechanical degree of freedom represented by the operator $T_{xyz} = (J_xJ_yJ_z + J_yJ_zJ_x + J_zJ_xJ_y - J_xJ_zJ_y - J_zJ_yJ_x - J_yJ_xJ_z)$, where J_x, J_y, and J_z represent magnetic dipoles in the *x*, *y*, and *z* directions. All the terms are equally induced and interact with those of neighboring ions to stabilize the AF-EQ ordered phase [1]. It is noted that the MOs are alternately arranged as well, resulting in an antiferro magnetic octupole (AF-MO) ordered state (inset of Fig. 2(b)). It is also noted that the even rank multipoles (mono-, quad-, etc.) are electric and the odd rank multipoles (di-, oct-, etc.) are magnetic, but the opposite state does not exist as far as the electrons are in a potential with inversion symmetry, which is the case in CeB_6.

To observe this AF-MO order, the resonant X-ray diffraction method was utilized. When the incident X-ray energy is tuned to the energy difference between the *2p* core level and the unoccupied *5d* or *4f* levels, the scattering amplitude is largely enhanced by the resonant process. When there is an ordering of *4f* orbitals, resonant diffraction occurs at the corresponding superlattice spots. An experiment was performed at BL-3A, using a vertical-field 8-Tesla superconducting magnet equipped on a two-axis diffractometer. The sample was cooled to 2.5 K below the AF-EQ ordering temperature of 3.3 K. We carefully investigated the superlattice reflections corresponding to the periodicity of the AF-EQ order.

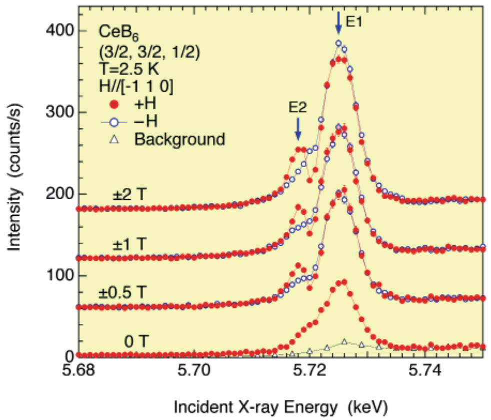

Figure 1
X-ray energy spectra of the (3/2 3/2 1/2) superlattice reflection in magnetic fields with reversed directions. The E1 (*2p–5d*) resonance at 5.724 keV reflects the AF-EQ moment. The E2 (*2p–4f*) resonance at 5.718 keV reflects both the AF-EQ and AF-MO moments. From the asymmetric behavior of the E2 peak with respect to the field reversal, the information on the AF-MO moment can be extracted.

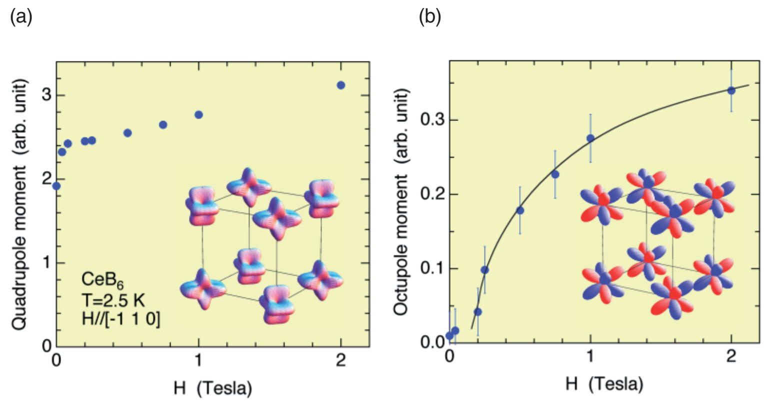

Figure 2
Magnetic-field dependences of (a) AF-EQ and (b) AF-MO moments deduced from the symmetric E1 and asymmetric E2 components. The solid line is a guide for the eye.

In the course of the present study, we discovered that the resonance peak exhibited different energy spectra when the field direction was reversed, as shown in Fig. 1 [2]. For fields in the plus direction, the E1 (*2p–5d*) and E2 (*2p–4f*) resonance peaks became stronger and well resolved, whereas the E2 peak became obscure when the field direction was reversed.

We analyzed the results in terms of the interference effect between the E1 and E2 resonances. As theoretically demonstrated, the E1 and E2 resonances are sensitive to multipole moments up to quadrupole (rank 2) and hexadecapole (rank 4), respectively [3]. In CeB_6 it is considered that the E1 and E2 peak reflect EQ and MO moments, respectively, since these two types of moments are the main order parameters in CeB_6. First, from the magnetic-field dependence of the intensity measured at 5.718 keV (E2) and 5.724 keV (E1), symmetric and asymmetric components with respect to the field reversal were extracted. The E1 peak consists mostly of the symmetric part, indicating that the peak indeed reflects the EQ moment, which does not change its sign with the field reversal. On the other hand, the E2 peak has a large contribution from the asymmetric part, which is due to the *4f* moment whose direction is reversed with respect to the field reversal.

The magnetic-field dependences of the EQ and MO moments deduced from the symmetric and asymmetric field dependences are shown in Fig. 2(a) and 2(b). The field dependence of the MO moment agrees well with that of the transferred hyperfine field at the boron site as deduced from NMR. In addition, it exhibits a convex dependence like a Brillouin function as theoretically predicted. This is quite a contrast to the concave field dependence of the induced antiferro-magnetic-dipole component as measured by neutron diffraction in the past. The field reversal method used in this study has the potential to be widely applied to other multipole ordering systems.

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