Thermal expansion and ultrasonic measurements of ferroquadrupole ordering in HoB_6

Takashi Yamaguchi [a,1], Mitsuhiro Akatsu [a], Yoshiyuki Nakano [a], Takehito Washizawa [a], Yuichi Nemoto [a], Terutaka Goto [a], Andreas Dönni [a], Shintaro Nakamura [b], Satoru kunii [c]

[a] *Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan*
[b] *Center for Low Temperature Science, Tohoku University, Sendai 980-8577, Japan*
[c] *Department of Physics, Tohoku University, Sendai 980-8578, Japan*

Abstract

Rare-earth hexaboride HoB_6 shows a ferroquadrupole ordering of O_{yz}, O_{zx} and O_{xy} with Γ_5 symmetry at $T_Q = 6.1$ K accompanied by a structural change from cubic to trigonal. In order to examine the order parameter in HoB_6, we have performed ultrasonic and thermal expansion measurements. The lattice length along [001] expands $\Delta L/L_{[001]} = 2.5 \times 10^{-4}$ just below T_Q, while the length along [111] shrinks appreciably $\Delta L/L_{[111]} = -4.5 \times 10^{-3}$. This result confirms the cubic-trigonal distortion observed by neutron scattering experiments.

Key words: HoB_6; ferroquadrupole ordering; ultrasound; thermal expansion

Rare-earth hexaborides RB_6 with the cubic CaB_6 type structure show various properties due to the spin, orbit and charge degrees of freedom for 4f-electrons [1]. Heavy rare-earth compound HoB_6 with Γ_5 triplet ground state show a ferroquadrupole (FQ) ordering at $T_Q = 6.1$ K and an antiferromagnetic (AFM) ordering at $T_N = 5.6$ K. The transverse C_{44} mode of HoB_6 shows a huge softening of 70% around T_Q. Neutron scattering indicates the crystal structure of HoB_6 changes from cubic to trigonal at T_Q [2]. These results are described in terms of quadrupole-strain interaction. In order to characterize the ferroquadrupole ordering in HoB_6, we have performed ultrasonic measurements in magnetic fields and thermal expansion measurements. Phase comparison method was used for the sound velocity measurements. A dilatmetric cell based on capacitance method with an accuracy 10^{-8} was employed.

Fig. 1 shows the temperature dependence of the elastic constant C_{44} of HoB_6 in magnetic fields for $H//[111]$. The C_{44} is measured by transverse ultrasonic wave propagating along $\boldsymbol{k}//[110]$ with polarization $\boldsymbol{u}//[001]$. Under zero field, the elastic constant C_{44} shows a pronounced softening about 70% above T_Q. This softening arises from the coupling of quadrupole O_{yz}, O_{zx} and O_{xy} to elastic strain with Γ_5 symmetry as, $H_{QS} = -g_{\Gamma 5}(O_{yz}\varepsilon_{yz} + O_{zx}\varepsilon_{zx} + O_{xy}\varepsilon_{xy})$. Increasing magnetic field, the softening of C_{44} is suppressed and the minimun point indicating ferroquadrupole transition shifts to higher temperatures. The softening of C_{44} in HoB_6 is influenced easily by magnetic fields. The small magnetic field effect in softening of C_{44} in DyB_6 is contrast to the present results of HoB_6 in Fig. 1 [3].

The inset of Fig. 1 shows a magnetic phase diagram of HoB_6. Under high fields up to 6 T, the antiferromagnetic transition temperature T_N decided by specific heat in magnetic fields for [110] shifts to lower temperatures, while the FQ ordering temperature T_Q shifts to higher temperatures [4]. These behavior are typical phenomena in other material showing FQ ordering [5].

Fig. 2 shows the temperature dependence of thermal expansion in HoB_6 along the [001] and [111] directions. The thermal expansion along [001] expands

[1] E-mail: gucci@phys.sc.niigata-u.ac.jp

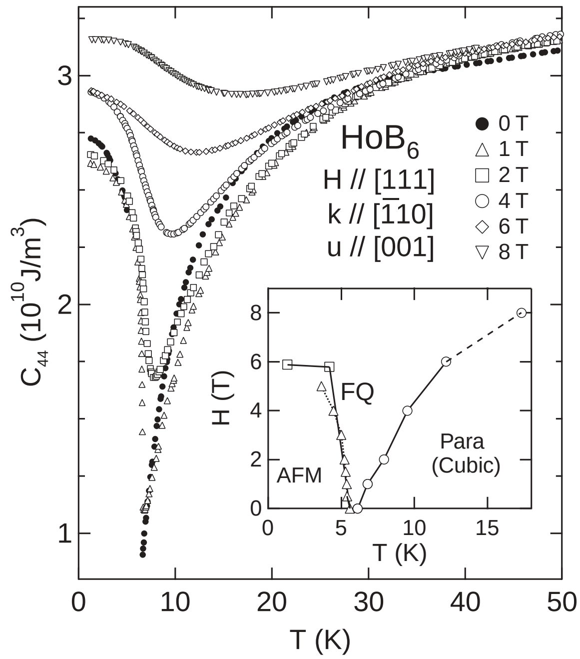

Fig. 1. Temperature dependence of the elastic constant C_{44} in HoB_6 under several magnetic fields for $H//[111]$. Inset is the magnetic phase diagram of HoB_6. The circles (T_Q) in phase diagram are decided by temperature dependence of the elastic constant, the squares (T_N) by field dependence of the elastic constant and the triangles (T_N) by the specific heat in magnetic fields for H//[110] [3].

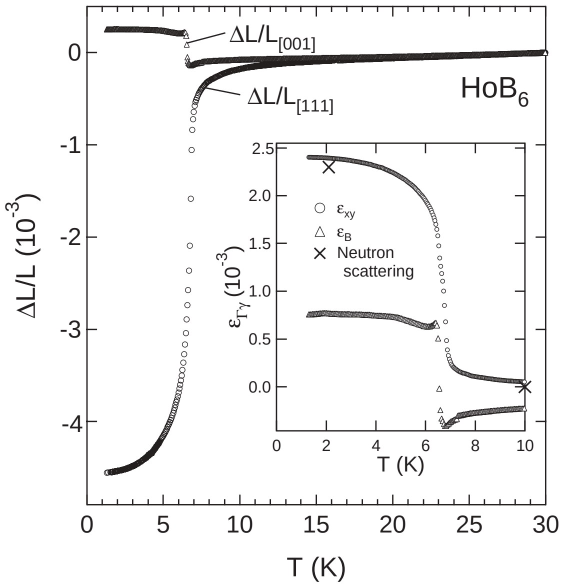

Fig. 2. Thermal expansion of HoB_6 along the [001] and [111] directions. Inset is the temperature dependence of the spontaneous strains ε_{xy} and ε_B below T_Q. The present result is well consistent with the previous neutron results[2].

$\Delta L/L_{[001]}$=2.5×10^{-4}, while along [111] shrinks appreciably $\Delta L/L_{[111]}$=-4.5×10^{-3} with decreasing temperature far below T_Q. The inset of Fig. 2 is the temperature dependence of the spontaneous strains ε_{xy}, which means changing the angle between x and y axes, and ε_B, which means expanding volume, caluculated by the experimental results in Fig. 2 using equations $\Delta L/L_{[001]} = \varepsilon_{zz} = \varepsilon_B/3 + \varepsilon_u/\sqrt{3}$ and $\Delta L/L_{[111]} = \varepsilon_B/3+2(\varepsilon_{yz}+\varepsilon_{zx}+\varepsilon_{xy})/3$. In the case of cubic-trigonal transition in HoB_6, it is naturally expected that tetragonal distortion is irrelevant $\varepsilon_u = 0$. Consequently the volume strain is described as $\varepsilon_B = 3\times\Delta L/L_{[001]}$, and the trigonal strain is written as $\varepsilon_{xy} = (\Delta L/L_{[111]} - \Delta L/L_{[001]})/2$. The temperature dependence of strains ε_{xy} and ε_B shows a jump indicating a first order transition at T_Q. The spontaneous strain $\varepsilon_{xy} = 2.4\times10^{-3}$, $\varepsilon_B = 7.5\times10^{-4}$ at 2 K far below T_Q means the trigonal distortion in addition to the volume expansion. The spontaneous strain $\varepsilon_{yz} = \varepsilon_{zx} = \varepsilon_{xy} \neq 0$ for trigonal distortion below T_Q is proportional to the quadrupole moment such as $<\varepsilon_{\Gamma5}>= Ng_{\Gamma5} < O_{\Gamma5} > /C^0_{44}$. The distorted angle $\alpha = 90^\circ+2\theta$ in the trigonal lattice from $\alpha = 90^\circ$ in orthogonal axes of cubic is described in terms of the spontaneous strain as $\varepsilon_{xy} = \sin\theta$. The neutron scattering of HoB_6 observed a change of the angle from $\alpha = 90^\circ$ at 10 K above T_Q to $\alpha = 90.264^\circ$ at 2.1 K below T_Q [2]. This result leads to $\varepsilon_{xy} = 2.3\times10^{-3}$, which is consistent with the present thermal expansion in Fig. 2.

In this paper, we show the cubic-trigonal distortion in FQ ordering of HoB_6. It is now recognized the dilatometry by the capacitance method is a very sensitive probe for the lattice distortion associate with FQ ordering. We are now in progress the thermal expansion under magnetic fields to elucidate field effect for the order parameter of HoB_6.

References

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