1. Research background

Recently, physical properties originating from the higher multipole degrees of freedom have been received much attention. Usually, researches of the higher multipole are performed by using compounds with the higher crystal symmetry such as a cubic symmetry, because an orbitally degenerate state can exist in the crystal electric field (CEF) state. On the other hand, compounds with a lower symmetry than cubic have attracted less attention because the orbitally degenerate state splits by the CEF. However, several compounds with the lower symmetry show the multipolar ordering. In the compounds, electric quadrupole degrees of freedom originate from a formation of a ground quasi–degenerate state. The fact suggests a possibility that the multipolar ordering may occur in the lower symmetric system than tetragonal if there is the quasi-degenerate state.

In this context, Ho$\text{Tr}_2\text{Al}_{10}$($\text{Tr}=\text{Fe, Ru}$) with the orthorhombic structure is a good candidate which shows the quadrupolar ordering in the low symmetric system. The 4$f$-electronic state of Ho$^{3+}$ under the orthorhombic CEF splits to 17 singlets. About HoFe$_2$Al$_{10}$, there is no magnetic ordering at 0.1 K under zero magnetic field, which is reported from a neutron scattering experiment. In addition, a specific heat between 0.25 and 3 K shows no sharp peak indicating a phase transition. However, the temperature $T$ dependence of the inverse magnetic susceptibility of polycrystalline HoFe$_2$Al$_{10}$ shows a decrease below 0.25 K, inferring that an ordered state develops at 0.25 K in a magnetic field $H$. Note that the magnitude of the magnetic field $H$ in the experiment was not reported. From the results, there is a possibility of a field induced phase transition due to a quasi-degenerate state in HoFe$_2$Al$_{10}$. On the other hand, HoRu$_2$Al$_{10}$ shows an antiferromagnetic (AFM) phase transition at $T_N=5.0$ K at zero magnetic field. The phase transition at $T_N$ arises from a quasi-degenerate state. Considering the multipole degrees of freedom of the quasi-degenerate state, the quadrupole degrees of freedom may be allowed in the quasi-degenerate state because the number of the degrees of freedom in the quasi-degenerate state is 3($=2\times2-1$) at least. However, there has been no investigation about the quadrupole ordering in Ho$\text{Tr}_2\text{Al}_{10}$ so far. In the present study, to investigate the quadrupolar ordering in the orthorhombic compound Ho$\text{Tr}_2\text{Al}_{10}$, measurements of the specific heat, magnetization, and elastic modulus were carried out.
2. Research results

[HoFe$_2$Al$_{10}$]

In HoFe$_2$Al$_{10}$, transverse modulus $C_{55}$ shows an elastic softening down to 0.5 K without any anomaly under zero magnetic field, suggesting that no phase transition above 0.5 K. However, under $H$ along the $a$- and $c$-axes, the softening of $C_{55}$ in 0.6(0.4) T stops at $T_Q$=0.8(0.75) K for $H//a(c)$. With further increasing $H//a$ and $c$, an enhancement of the softening toward $T_Q$ in $C_{55}$ was observed. From the results, the magnetic field induced phase transition (FIP) has been confirmed in the $H$-$T$ diagrams for $H//a$ and $c$. $T_Q$ exhibits a reentrant behavior with respect to $H//a$ and $c$. Since $C_{55}$ is the modulus corresponding to quadrupole $O_{zx}$, the enhancement of the elastic softening toward $T_Q$ in $C_{55}$ by $H$ suggests that the FIP is the field induced $O_{zx}$-type quadrupolar ordering. The CEF calculation for the specific heat, magnetization, and elastic modulus shows the approach between the CEF ground and first excited singlet with increasing $H//a$ and $c$, suggesting a formation of a quasi-degenerate state. Here, there is the interlevel transition of $O_{zx}$ between them. In this sense, the field induced quadrupolar(FIQ) ordering probably originates from the enhancement of the expected value for $O_{zx}$ by the formation of the quasi-degenerate state.

[HoRu$_2$Al$_{10}$]

The step like softening at $T_N$ in longitudinal moduli $C_{11}$, $C_{22}$, and $C_{33}$, and hardening of all moduli below $T_N$, which can be explained by the thermodynamic relation between the elastic moduli and the magnetic ordering, were observed in HoRu$_2$Al$_{10}$ at zero magnetic field. In addition, $H$-$T$ diagrams for $H//a$, $b$, and $c$ were determined from the measurements of the specific heat, magnetization, and elastic modulus. In $H//b$, $T_N$ decreases monotonically with increasing $H$, and a phase boundary closes around 1.0 T, which is a usual behavior originating from a competition between the Zeeman effect and spin interaction. In contrast, under $H//a$ and $c$, although $T_N$ decreases with increasing $H$, the phase boundary shows an inflection point around 5.0 T. Meanwhile, the remarkable softening of $C_{11}$, $C_{22}$, and $C_{33}$ toward $T_N$ is induced under $H//c$ above 8 T, which is not observed in the low $H$ range. Since $C_{11}$, $C_{22}$, and $C_{33}$ are included in an elastic mode $(C_1+2C_{12}+4C_{13}+C_{22}+4C_{23}+4C_{33})/12$ which is corresponding to quadrupole $O_{20}$, the field induced softening in $C_{11}$, $C_{22}$, and $C_{33}$ suggests that the $O_{20}$–type quadrupolar ordering emerges in high $H//c$. The FIQ ordering may be expected in the high $H//a$ because the magnetic anisotropy between the $a$- and $c$- axes is small. From the view point, the inflection point of the phase boundary in $H//a$ and $c$ is assumed to result from the phase transition from the AFM ordering in the low $H$ range to the FIQ ordering in the high $H$ range. However, for $H//a$ and $c$, any clear anomaly which indicates the phase boundary between the AFM ordering and FIQ ordering were not observed in this study, suggesting that there is a crossover between the higher and lower $H$ range.

3. Summary
In the present study, the measurements of the specific heat, magnetization, and elastic modulus were performed on HoTr$_2$Al$_{10}$ with the orthorhombic symmetry to investigate the ordered state in HoTr$_2$Al$_{10}$. In the experiments, the enhancement of the elastic softening under $H \parallel a$ and $c$ was observed in HoTr$_2$Al$_{10}$, suggesting that the FIQ ordering emerges by applying $H \parallel a$ and $c$. The order parameters of the FIQ phase in HoFe$_2$Al$_{10}$ and HoRu$_2$Al$_{10}$ are probably $O_{zx}$ and $O_{z}^{0}$, respectively. This is the first report on the FIQ ordering in the compounds with the orthorhombic symmetry.