論 文 の 要 旨

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論 文 題 目　Field Induced Quadrupole Phase Transition in HoTr_2Al$_{10}$(Tr=Fe, Ru) under an Orthorhombic Crystal Electric Field
（HoTr_2Al$_{10}$ (Tr =Fe, Ru)の直方晶結晶場下における磁場誘起四極子相転移）

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, HoTr_2Al$_{10}$(Tr=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 $HoFe_2Al_{10}$, 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 $HoFe_2Al_{10}$ 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 $HoFe_2Al_{10}$. On the other hand, $HoRu_2Al_{10}$ 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(=2×2-1) at least. However, there has been no investigation about the quadrupole ordering in HoTr_2Al$_{10}$ so far. In the present study, to investigate the quadrupolar ordering in the orthorhombic compound HoTr_2Al$_{10}$, measurements of the specific heat, magnetization, and elastic modulus were carried out.

2. Research results

【$HoFe_2Al_{10}$】

In $HoFe_2Al_{10}$, 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.

【$HoRu_2Al_{10}$】

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 $HoRu_2Al_{10}$ 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 $(C_{11}+2C_{12}-4C_{13}+C_{22}-4C_{23}+4C_{33})/12$ which is corresponding to quadrupole O_2^0, the field induced softening in C_{11}, C_{22}, and C_{33} suggests that the O_2^0–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_2Al$_{10}$ with the orthorhombic symmetry to investigate the ordered state in HoTr_2Al$_{10}$. In the experiments, the enhancement of the elastic softening under H // a and c was observed in HoTr_2Al$_{10}$, suggesting that the FIQ ordering emerges by applying H // a and c. The order parameters of the FIQ phase in $HoFe_2Al_{10}$ and $HoRu_2Al_{10}$ are probably O_{zx} and O_2^0, respectively. This is the first report on the FIQ ordering in the compounds with the orthorhombic symmetry.