Elastic Constants of DyRhIn$_5$

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Abstract. The components-separated magnetic transition in DyRhIn$_5$ was investigated by measuring the magnetic susceptibility and elastic constants. The magnetic susceptibility along the [001] direction indicates the antiferromagnetic ordering of c-component of the magnetic moments at $T_N = 28.1$ K whereas the susceptibility along those of [100] and [110] direction show the paramagnetic behavior. The results of the elastic constant measurements suggest that the degeneracy of quadrupolar degrees of freedom exists in spite of the formation of magnetic order because the elastic softenings are observed below $T_N$. The quadrupolar effect in DyRhIn$_5$ is discussed in terms of the symmetry classification.

1. Introduction
"Successive component-separated magnetic transitions", which is unique successive transitions characterized by the independent magnetic ordering of the c- and ab-components of the magnetic moments at different temperatures, is known as one of the interesting magnetic behavior caused by the existence of a competing interaction in an f-electron system. DyB$_4$ [1] and TbCoGa$_5$ [2] are well known examples in rare-earth compounds which show such successive transitions. More recently, this type of successive transitions is reported in HoRh$_2$Si$_2$ [3].

In our study of successive components-separated magnetic transitions, we focused on DyRhIn$_5$ that crystallizes into the tetragonal HoCoGa$_5$-type structure ($D_4h^1-P4/mmm$). N. V. Hieu et al. reported that the magnetic susceptibility of DyRhIn$_5$ for $B||[001]$ shows a clear peak as $T_N = 28.1$ K, whereas that for $B||[100]$ shows no anomaly at $T_N$ and increases even below $T_N$ [4, 5]. This magnetically ordered phase in DyRhIn$_5$ is quite similar to the intermediate phase of DyB$_4$ or TbCoGa$_5$. In the case of DyB$_4$ and TbCoGa$_5$, we suggested that the presence of the degeneracy of quadrupolar degrees of freedom is an important key to the occurrence of the successive components-separated magnetic transitions. Therefore, the research on quadrupolar degrees of freedom is essential to clarify the magnetic transition in DyRhIn$_5$. In order to investigate the magnetic transition in DyRhIn$_5$, we have performed measurements of the magnetic susceptibility and elastic constants on the single crystals. The results and discussion are reported below.

2. Experimental
Single crystals of DyRhIn$_5$ were grown by the self-flux method using In as flux. The starting materials were inserted in a quartz tube in the ratio of Dy:Rh:In=1:1.30 and sealed under high vacuum. This tube was heated up to 1000°C and cooled down to room temperature at a rate of $-10^\circ$C/hr. The direction of the single crystals was determined by the X-ray back Laue method. The magnetic susceptibility of DyRhIn$_5$ was measured using a SQUID magnetometer in the temperature range from 1.8 to 300 K under a magnetic field of 0.1 T. The elastic constants
were examined by means of the ultrasonic measurement in the temperature range from 1.8 K to 50 K using the single crystalline sample. The measurement of the elastic constants is an effective technique to investigate the quadrupolar effect in an f-electron system with an orbitally degenerate ground state, because the quadrupole moment of the f-electron couples to the elastic strain. A relative change in the elastic constant, \( \Delta C / C \), of the sample was obtained using \( \Delta C / C = 2 \Delta v / v + (\Delta v / v)^2 \), which is derived from \( C = \rho v^2 \), where \( v \) and \( \rho \) represent the sound velocity and the density of the crystal, respectively. \( \Delta v / v \) was measured with a phase comparator using double-balanced mixers.

3. Experimental results and discussion

Figure 1. (a) Temperature dependence of the magnetic susceptibility \( \chi \) measured for magnetic field \( B = 0.1 \) T applied parallel to the main symmetry directions in DyRhIn\(_5\) single crystal. (b) Temperature dependence of inverse magnetic susceptibility \( 1/\chi \) along the main symmetry directions in DyRhIn\(_5\) single crystal. The solid lines show the Curie-Weiss fit to the data.

Figure 1 shows the temperature dependence of the magnetic susceptibility \( \chi \) for the single crystal of DyRhIn\(_5\). No difference between the field cooling (FC) and zero-field cooling (ZFC) susceptibility in any direction is observed throughout the whole temperature range. Therefore, only ZFC data are shown for clarification. The coincidence of the ZFC and FC data suggests that no spontaneous magnetization is present in DyRhIn\(_5\). The susceptibility for \( B || [001] \) shows a clear maximum at \( T_N = 28.1 \) K. In contrast, those for \( B || [100] \) and \( B || [110] \) show no clear anomaly around \( T_N \) and increase even below \( T_N \). These results are mostly consistent with Ref. [4], but it is slightly different in that the susceptibility for \( B || [100] \) and \( B || [110] \) continue to increase down to 1.8 K whereas the one for \( B || [100] \) saturated toward the lowest temperature in previous report [4].

The magnetic susceptibility above 150 K follows the Curie-Weiss law (shown in Fig. 1(b)) with the effective paramagnetic moments \( \mu_{\text{eff}} \) and the Weiss temperatures \( \theta_p \): \( \mu_{\text{eff}} = 10.39 \mu_B \) and \( \theta_p = 9.6 \) K for \( B || [001] \), \( \mu_{\text{eff}} = 10.59 \mu_B \) and \( \theta_p = -51.2 \) K for \( B || [100] \) and \( \mu_{\text{eff}} = 10.44 \mu_B \) and \( \theta_p = -47.1 \) K for \( B || [110] \), respectively. The effective paramagnetic moments are close to 10.63 \( \mu_B \) of the free ion value of Dy\(^{3+}\).

Figure 2 shows the temperature dependence of the relative change in the elastic constants \( \Delta C / C \) of DyRhIn\(_5\). The transverse mode \( \Delta C_{44} / C_{44} \) related with elastic strain \( \varepsilon_{xy} \) or \( \varepsilon_{xz} \) shows
a softening of 0.5% from \( \sim 13 \text{ K} \) down to 1.8 K and the slight anomaly at approximately \( T_N \). Meanwhile the longitudinal mode \( \Delta C_{33}/C_{33} \) and the transverse mode \( \Delta C_{66}/C_{66} \) show a smaller softening and the longitudinal mode \( \Delta C_{11}/C_{11} \) shows the monotonic hardening with decreasing temperature down to 1.8 K. \( \Delta C_{33}/C_{33} \) shows a nonmonotonic temperature dependence below \( T_N \). However, a reason of the nonmonotonic behavior of \( \Delta C_{33}/C_{33} \) is still unknown. The elastic softenings below \( T_N \) suggest that the degeneracy of the quadrupolar degrees of freedom still exist in magnetically ordered phase. Moreover the quadrupole moments related with \( C_{44} \) are hardly influenced by the magnetic ordering in the [001] direction.

Table 1. The symmetry of quadrupole moments, elastic constants and dipole moments in a tetragonal system with \( D_{4h} \)

| Symmetry | Quadrupole Moments | Elastic constants | Dipole moments |
|----------|--------------------|------------------|---------------|
| \( \Gamma_1 \) | \( O_2^\| = \frac{1}{2}(2J^2_z - J^2_x - J^2_y) \) | \( C_u = \frac{1}{8}(C_{11} + C_{12} - 4C_{13} + 2C_{33}) \) | \( J_z \) |
| \( \Gamma_3 \) | \( O_2^\| = \frac{\sqrt{3}}{2}(J^2_x - J^2_y) \) | \( \frac{1}{2}(C_{11} - C_{12}) \) | |
| \( \Gamma_4 \) | \( O_{xy} = \frac{\sqrt{3}}{2}(J_xJ_y + J_yJ_x) \) | \( C_{66} \) | |
| \( \Gamma_5 \) | \( O_{yz} = \frac{\sqrt{3}}{2}(J_yJ_z + J_zJ_y) \) | \( C_{44} \) | \( J_x, J_y \) |
| | \( O_{zx} = \frac{\sqrt{3}}{2}(J_xJ_z + J_zJ_x) \) | | |

Here, we show the symmetry classification of the quadrupole moments, elastic constants and dipole moments in a tetragonal system with \( D_{4h} \) of HoCoGa\(_5\)-type structure in Table 1. The behaviors of the magnetic susceptibility and the elastic constant \( C_{44} \) of DyRhIn\(_5\) are quite similar
to those in the intermediate phase of TbCoGa₅ [2]. The $ab$-component of the magnetic moments seems to be influenced by the quadrupole moment system, but not the $c$-component in DyRhIn₅ and TbCoGa₅. The largest softening of $C_{44}$ in the measured $C_\Gamma$ modes indicates that the dominant components of the quadrupole moments are $O_{yz} = J_y J_z + J_z J_y$ and $O_{zx} = J_z J_x + J_x J_z$ with $\Gamma_5$ symmetry. These results strongly suggest the relation between the magnetic ordering of the $c$-component of the magnetic moments with the paramagnetic behavior along the $ab$-plane and the largest softening of the elastic constant $C_{44}$.

It is known that rare-earth compounds may have degenerate crystalline electric field (CEF) ground state where orbital degrees of freedom remain. In general, if quadrupolar degrees of freedom are added in a magnetic system, the magnetic transition temperature may be suppressed from the original one due to the increase in entropy change during the magnetic transition. In DyRhIn₅ and TbCoGa₅, the dominant components of the quadrupole moments $O_{yz}$ and $O_{zx}$ related with the elastic constant $C_{44}$ are coupled with the $ab$-component of the magnetic dipole moment ($J_x, J_y$) in $\Gamma_5$ symmetry. Therefore, the transition temperature with the magnetic ordering of the $ab$-component can be strongly suppressed, but the ordering of the $c$-component ($J_z$) is not affected. Consequently, the magnetic ordering only the $c$-component occurs whereas the $ab$-components are paramagnetic.

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