Ultrasonic Study on the Hexagonal Antiferromagnet $Dy_3Ru_4Al_{12}$

I Ishii[1†], K Takezawa[1], H Goto[1], S Kamikawa[1], A V Andreev[2], D I Gorbunov[2,3], M S Henriques[2] and T Suzuki[1,4,5‡]

[1]Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
[2]Institute of Physics, Academy of Sciences, Na Slovance 2, 182 21 Prague, Czech Republic
[3]Dresden High Magnetic Field Laboratory, Helmholtz-Zentrum Dresden-Rossendorf, D-01314 Dresden, Germany
[4]Institute for Advanced Materials Research, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
[5]Cryogenics and Instrumental Analysis Division, N-BARD, Hiroshima University, Higashi-Hiroshima 739-8526, Japan

E-mail: [†]ish@hiroshima-u.ac.jp, [‡]tsuzuki@hiroshima-u.ac.jp

Abstract. In the distorted kagome lattice antiferromagnet $Dy_3Ru_4Al_{12}$ with $T_N = 7$ K, a crystal electric field (CEF) effect is expected at high temperatures. To investigate the CEF effect and the phase transition at T_N, we performed ultrasonic measurements on a single-crystalline sample. At high temperatures, both the longitudinal elastic modulus C_{11} and the transverse modulus C_{44} increase monotonically with decreasing temperature. Below 60 K a characteristic elastic softening is observed in C_{44} in contrast to C_{11} with monotonic hardening down to T_N. We analyzed C_{44} using the Curie-Weiss-type equation and obtained a negative parameter: Θ which is proportional to a quadrupole-quadrupole coupling constant under the hexagonal CEF. With further decreasing temperature, both moduli exhibit abrupt elastic hardening at T_N due to a magnetostriction.

1. Introduction

Many compounds of which magnetic ions form a kagome lattice have attracted considerable interest for their fascinating magnetic properties originating from geometrical frustration of spins [1, 2, 3, 4]. The ternary rare-earth compounds $R_3Ru_4Al_{12}$ (R: rare-earth) crystallize in the hexagonal $Gd_3Ru_4Al_{12}$-type structure (space group $P6_3/mmc$) [5, 6, 7, 8, 9]. In this hexagonal structure, constituent atoms are stacked in layers perpendicular to the c-axis with R-Al and Ru-Al layers alternately, and R atoms form a distorted kagome net.

In the Dy-based compound $Dy_3Ru_4Al_{12}$, the electrical resistivity shows a metallic behavior [8]. The specific heat exhibits a clear peak at $T_N = 7$ K indicating the phase transition. The transition is of first-order and is not accompanied by a structural transformation [9]. A cusp-type anomaly is observed at T_N in the magnetic susceptibility along the magnetically easy c-axis. Magnetization experiments manifested that there are one and two meta-magnetic phase transitions in the magnetic field along the a- and c-axes, respectively, suggesting rather complicated magnetic structure. The phase transition at T_N is reported as an antiferromagnetic ordering with two possible noncollinear magnetic structures by neutron diffraction experiments [8].

At high temperatures, the magnetic susceptibility obeys the Curie-Weiss law above 100 K along the a-axis and above 170 K along the c-axis, suggesting an almost localized character of $4f$-electrons. The estimated effective magnetic moments (10.1 μ_B for both axes) are close to the value of the free Dy^{3+} ion (10.6 μ_B) [8]. A crystal electric field (CEF) effect is expected in $Dy_3Ru_4Al_{12}$ and magnetic anisotropy can be attributed to the CEF effect. Here, the sixteen-fold multiplet of the Dy^{3+} ion splits into eight Kramers doublets in the hexagonal CEF, where the total angular momentum J is equal to 15/2. In the preset work, we carried out ultrasonic measurements on $Dy_3Ru_4Al_{12}$ in order to study the CEF effect on the elastic moduli and the phase transition at T_N.

2. Experimental

A single crystal of $Dy_3Ru_4Al_{12}$ was grown by a modified Czochralski method [8]. The elastic moduli C_{11} and C_{44} were measured as a function of the temperature T from 4.2 to 150 K using the phase comparison-type pulse echo method [10]. The modulus C_{11} is the longitudinal mode propagating along the a-axis, and C_{44} is the transverse mode propagating along the a-axis with the polarization direction along the c-axis. We used $LiNbO_3$ transducers with the fundamental resonance frequency of about 30 MHz. The modulus C_{ii} was calculated using the relation $C_{ii} = \rho v^2$ with a room-temperature mass density $\rho = 6.30$ g/cm^3, where v is the sound velocity in a sample. The absolute value of v is estimated at 4.2 K by using the sample length and a time interval between pulse echoes.

3. Results and discussion

Figure 1 shows the T dependence of the longitudinal elastic modulus C_{11} in $Dy_3Ru_4Al_{12}$. The modulus C_{11} increases monotonically with decreasing T down to T_N. An abrupt elastic hardening is detected at T_N, as shown in the inset of Fig. 1.

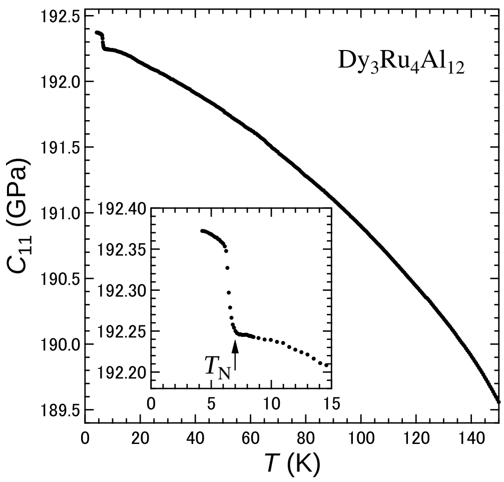

Figure 1. Temperature dependence of the longitudinal elastic modulus C_{11}. The inset represents the same data in an expanded scale below 15 K.

The T dependence of the transverse modulus C_{44} is shown in Fig. 2. At high temperatures, monotonic hardening is observed above 60 K. The modulus C_{44} turns into soften below 60 K. The elastic softening stops at T_N, and then C_{44} exhibits abrupt hardening at T_N, as shown in the inset of Fig. 2. Further elastic softening appears below 6 K.

The softening between T_N and 60 K in C_{44} is a characteristic behavior originating from a quadrupole interaction under the CEF. To simplify the analysis, we performed the theoretical

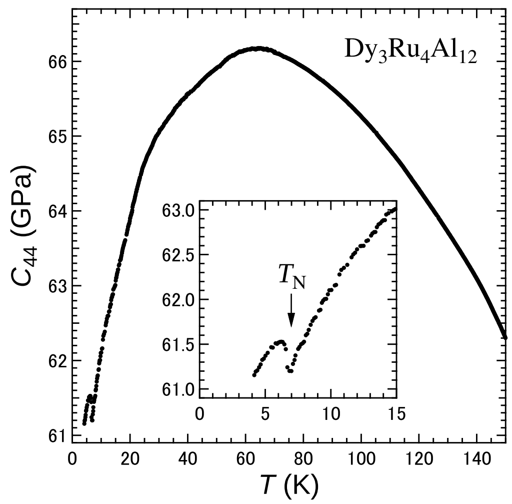

Figure 2. Temperature dependence of the transverse elastic modulus C_{44}. The inset represents the same data in an expanded scale below 15 K.

fitting based on the Curie-Weiss-type formula for the co-operative Jahn-Teller effect [11].

$$C(T) = C_0 \frac{T - T_c}{T - \Theta}, \tag{1}$$

where Θ is proportional to a quadrupole-quadrupole coupling constant and $T_c - \Theta$ is a measure of Jahn-Teller energy including a strain-quadrupole coupling constant. We assumed the back ground stiffness C_0 as

$$C_0 = a + bT^2 + cT^4, \tag{2}$$

where a, b, and c are constants.

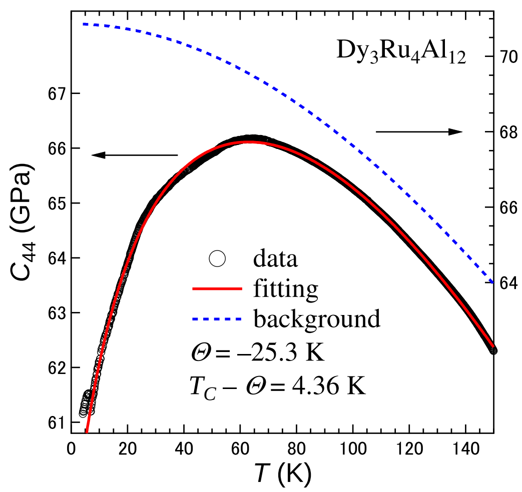

Figure 3. Temperature dependence of C_{44}. The red solid and blue broken curves demonstrate the fitting result and the background stiffness, respectively.

The softening of C_{44} is well reproduced above T_N with fitting parameters listed in Table 1, as shown in Fig. 3. This analysis reveals that the softening of C_{44} arises from the quadrupole interaction. Furthermore, the negative value of Θ suggests that the interaction is of antiferroquadrupolar-type. Under the hexagonal CEF with the Dy^{3+} ion ($J = 15/2$), no elastic

Table 1. Fitting parameters of C_{44}: Θ (K), $T_c - \Theta$ (K), a (GPa), b ($\times 10^{-4}$ GPa/K^2), and c ($\times 10^{-9}$ GPa/K^4).

Θ	$T_c - \Theta$	a	b	c
-25.3	4.36	70.9	-3.37	1.32

softening is expected by only the ground doublet, since the Kramers doublet has no quadrupole degeneracy. The softening is caused by an indirect quadrupole interaction between the ground doublet and the excited doublets, such as our earlier results of YbIrGe and YbPtGe [12, 13]. Consequently, we obtained information of the quadrupole interaction due to $4f$-electronic states of the Dy^{3+} ion under the hexagonal CEF from the T dependence of C_{44}. In the future works, we are planning to measure the elastic moduli of other modes (C_{33} and C_{66}) and perform the theoretical fitting using the hexagonal CEF to reproduce the elastic moduli and the magnetic susceptibility.

As for the phase transition at T_N, antiferromagnetic ordering is reported by neutron diffraction experiments [8]. In thermal expansion measurements, anisotropic step-wise lattice contraction due to a magneto-elastic coupling is detected at T_N [9]. Both moduli C_{11} and C_{44} show the abrupt elastic hardening at T_N. Our results also indicate that a strain strongly couples to a magnetic order parameter. These hardening might originate from the magnetostriction and can be explained by the thermodynamic theory of elastic modulus with the Landau theory [14].

4. Conclusion

The elastic moduli C_{11} and C_{44} were measured in the distorted kagome lattice antiferromagnet $Dy_3Ru_4Al_{12}$. We found characteristic elastic softening due to the quadrupole interaction below 60 K in the transverse modulus C_{44} in contrast to the longitudinal modulus C_{11} without the softening. The negative Θ obtained by the Curie-Weiss-type fitting suggests that there is the antiferroquadrupolar interaction. The indirect quadrupole interaction between the ground doublet and the excited doublets under the hexagonal CEF plays a central role for the softening. The abrupt elastic hardening, owing to the strong coupling between the strain and the magnetic order parameter, is observed at T_N in both modes.

5. Acknowledgments

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