Investigation of a quadrupolar interaction in DyCu by elastic constants measurement

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Abstract. We have investigated a quadrupolar interaction by measuring the temperature dependence for elastic constants of $C_B = (C_{11} + 2C_{12})/3$ ($\Gamma_1$-symmetry), $C' = (C_{11} - C_{12})/2$ ($\Gamma_3$-symmetry) and $C_{44}$ ($\Gamma_5$-symmetry) in DyCu. We have evaluated the quadrupolar interaction coefficient $K(0)$ by analyzing critical fields of metamagnetic transitions. The following results are obtained; (i) $C_{44}$ in the paramagnetic phase exhibits softening at temperatures near $T_N$ while $C_B$ and $C'$ do not. This indicates that the dominant components of the quadrupolar moment are $Oyz$, $Ozx$ and $Oxy$ with $\Gamma_5$-symmetry. (ii) From an analysis of critical fields of metamagnetic transitions based on a mean-field approximation we have determined the quadrupolar interaction coefficient to be $K(0)|Q|^2/k_B = -16.8$ K. The negative value of $K(0)$ indicates that the quadrupolar interaction has an antiferro-type of $\Gamma_5$-symmetry.

1. Introduction

The rare earth intermetallic compound DyCu that has a CsCl-type cubic crystal structure shows a magnetic transition from an antiferromagnetic (AFM) phase to a paramagnetic (PM) phase at $T_N = 63.0$ K. The AFM phase has a triple-$q$ structure (propagation vector: $q = <1/2 1/2 0>$2$\pi/a$, where $a$ is the crystal lattice parameter) with magnetic moments of Dy$^{3+}$ being parallel to the four equivalent $<1 1 1>$ directions [1,2]. When a magnetic field is applied to the AFM phase, multi-step metamagnetic transitions occur [2-5]. These transitions have a field-direction dependence, that is, magnetization curves in the [0 0 1], [1 1 0] and [1 1 1] directions exhibit two-step, three-step and four-step transitions, respectively. The origin of the complicated magnetic structure and metamagnetic transitions are reported to be due to the coexistence of exchange and quadrupolar interactions [1-6]. However, there have been a few studies on the quadrupolar interaction type ($\Gamma_3$ or $\Gamma_5$-symmetry) and on the quantitative analysis for quadrupolar interaction coefficients.

In this study, we have measured the temperature dependence of elastic constants for DyCu to investigate the quadrupolar interaction [5,7]. We have also evaluated the exchange interaction coefficient and the quadrupolar interaction coefficient by an analysis of critical fields of metamagnetic transitions.

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2. Experimental procedure
An ingot of DyCu was prepared by arc-melting using Dy (99.9 %) and Cu (99.99 %) as starting materials. A single crystal rod of DyCu was grown by a floating zone method at a growth rate of 5.0 mm h⁻¹. The specimen used for elastic constants measurement was cut into 3.17 × 3.29 × 3.45 mm³ and it weighted 318.53 mg. Its surfaces were parallel to the {1 0 0} plane. The temperature dependence of the elastic constants were measured by using a rectangular parallelepiped resonance method, which enables the determination of all elastic constants simultaneously [7,8]. This was done in the temperature range of 2 - 300 K. Resonance spectra were obtained in the range of 200 - 800 kHz with a step of 0.1 kHz and the number of resonance peaks used for the analysis was about 50 - 55 at all temperatures.

3. Results
Figure 1 shows the temperature dependence of elastic constants $C_B = (C_{11} + 2C_{12})/3$, $C' = (C_{11} - C_{12})/2$ and $C_{44}$ of DyCu.

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3. Results
Figure 1 shows the temperature dependence of elastic constants of $C_B = (C_{11} + 2C_{12})/3$ ($\Gamma_1$-symmetry), $C' = (C_{11} - C_{12})/2$ ($\Gamma_3$-symmetry) and $C_{44}$ ($\Gamma_5$-symmetry). It should be noted that $C_{44}$ in the PM phase shows a small softening at temperatures near $T_N = 62.0$ K, while $C_B$ and $C'$ do not show any softening. Considering that $C_{44}$ in the present system has $\Gamma_5$-symmetry, we can say that the quadrupolar interaction is $\Gamma_5$-symmetry type and the dominant components of the quadrupolar moment are $O_{yz}$, $O_{zx}$ and $O_{xy}$. Using this interaction symmetry type, we propose quadrupolar structures for DyCu and discuss the quadrupolar interaction coefficients in the next section.

4. Discussion
First of all, we propose quadrupolar structures for the AFM phase and the first field-induced metamagnetic phase in the [0 0 1] and [1 1 1] directions (FM-I₀₀₁ and FM-I₁₁₁) considering the
Table 1. Magnetic structures and quadrupolar structures of AFM, FM-I001 and FM-I111 phases.

| Phase  | Magnetic structure, $S_{\text{hkl}} = [S_x, S_y, S_z]$ | Quadrupolar structure, $Q_{\text{hkl}} = [O_{yz}, O_{zx}, O_{xy}]$ |
|--------|--------------------------------------------------|--------------------------------------------------|
| AFM    | $S_{000} = (1/\sqrt{3}) [1 1 1]$                | $Q_{000} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{011} = (1/\sqrt{3}) [1 1 1]$                | $Q_{011} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{100} = (1/\sqrt{3}) [1 1 1]$                | $Q_{100} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{110} = (1/\sqrt{3}) [1 1 1]$                | $Q_{110} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{001} = (1/\sqrt{3}) [1 1 1]$                | $Q_{001} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{101} = (1/\sqrt{3}) [1 1 1]$                | $Q_{101} = (1/\sqrt{3}) [1 1 1]$                |
|        | $S_{111} = (1/\sqrt{3}) [1 1 1]$                | $Q_{111} = (1/\sqrt{3}) [1 1 1]$                |

| FM-I001 | $S_{000} = (1/\sqrt{3}) [1 1 1]$                | $Q_{000} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{011} = (1/\sqrt{3}) [1 1 1]$                | $Q_{011} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{100} = (1/\sqrt{3}) [1 1 1]$                | $Q_{100} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{110} = (1/\sqrt{3}) [1 1 1]$                | $Q_{110} = (1/\sqrt{3}) [1 1 1]$                |
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|         | $S_{111} = (1/\sqrt{3}) [1 1 1]$                | $Q_{111} = (1/\sqrt{3}) [1 1 1]$                |

| FM-I111 | $S_{000} = (1/\sqrt{3}) [1 1 1]$                | $Q_{000} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{011} = (1/\sqrt{3}) [1 1 1]$                | $Q_{011} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{100} = (1/\sqrt{3}) [1 1 1]$                | $Q_{100} = (1/\sqrt{3}) [1 1 1]$                |
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|         | $S_{101} = (1/\sqrt{3}) [1 1 1]$                | $Q_{101} = (1/\sqrt{3}) [1 1 1]$                |
|         | $S_{111} = (1/\sqrt{3}) [1 1 1]$                | $Q_{111} = (1/\sqrt{3}) [1 1 1]$                |

following two assumptions.

- Only the quadrupolar components of $O_{yz}, O_{zx}$ and $O_{xy}$ are considered.
- The most symmetrical structure with respect to the magnetic field direction is considered.

Proposed quadrupolar structures are shown in table 1 and the magnetic structures as determined by neutron diffractions are also shown [1-3,10].

By considering the magnetic and quadrupolar structures in table 1, we quantitatively evaluate the exchange and quadrupolar interaction coefficients using measured critical fields based on fitting the following total Hamiltonian $\hat{H}_{\text{tot}}$, consisting of the exchange interaction term $\hat{H}_{\text{exch}}$, the quadrupolar interaction term $\hat{H}_{\text{quad}}$ and the Zeeman term $\hat{H}_{\text{Zeeman}}$:

$$\hat{H}_{\text{tot}} = \hat{H}_{\text{exch}} + \hat{H}_{\text{quad}} + \hat{H}_{\text{Zeeman}}.$$  \hspace{1cm} (1)

The exchange interaction term $\hat{H}_{\text{exch}}$ is given by:

$$\hat{H}_{\text{exch}} = \sum_{\text{n.n.r}} -2J_1(S_{x,i}S_{x,j} + S_{y,i}S_{y,j} + S_{z,i}S_{z,j}) + \sum_{\text{n.n.n}} -2J_2(S_{x,i}S_{y,j} + S_{y,i}S_{z,j} + S_{z,i}S_{x,j})$$  \hspace{1cm} (2)

where $J_1$ and $J_2$ are the exchange interaction coefficients of the first and second nearest-neighbours, respectively, $S_{x,i}$, $S_{y,i}$ and $S_{z,i}$ are the components of magnetic moment for the i-site and $J(0)$ is the Fourier transform of $J_1$ and $J_2$. The quadrupolar interaction term $\hat{H}_{\text{quad}}$ is written as:

$$\hat{H}_{\text{quad}} = \sum_{\text{n.n.r}} -K_1(O_{yz,i}O_{yz,j} + O_{zx,i}O_{zx,j} + O_{xy,i}O_{xy,j}) + \sum_{\text{n.n.n}} -K_2(O_{yz,i}O_{yz,j} + O_{zx,i}O_{zx,j} + O_{xy,i}O_{xy,j})$$  \hspace{1cm} (3)
where $K_1$ and $K_2$ are the quadrupolar interaction coefficients of the first and second nearest-neighbours, respectively. $O_{yz,i}, O_{zx,i}$ and $O_{xy,i}$ are the components of the quadrupolar moment for the $i$-site and $K(0)$ is the Fourier transform of $K_1$ and $K_2$. The Zeeman term is given by:

$$\hat{H}_{\text{Zeem}} = g_1 \mu_B \mu_0 (S_x H_x + S_y H_y + S_z H_z),$$

(4)

where $g_1$ is the Landé $g$-factor, $\mu_B$ is the Bohr magneton, $\mu_0$ is the magnetic permeability in vacuum and $H_x, H_y$ and $H_z$ are components of the applied magnetic field. We then determine the exchange and quadrupolar interaction coefficients $J(0)$ and $K(0)$ using the critical fields of metamagnetic transitions from the AFM phase to the FM-I001 phase ($\mu_0 H_{C1,001} = 6.6$ T at $T = 4.2$ K) and from the AFM phase to the FM-I111 phase ($\mu_0 H_{C1,111} = 8.0$ T at $T = 4.2$ K). This was based on $\hat{H}_{\text{tot}}$ between the AFM and FM-I001 phases and between the AFM and FM-I111 phase which are equal at the critical fields when a mean-field approximation is used. As a result, we obtain the following values:

$$J(0) \mid S \mid^2 / k_B = -53.7 \text{ K} \quad \text{and} \quad K(0) \mid Q \mid^2 / k_B = -16.8 \text{ K},$$

(5)

where $S$ and $Q$ are the absolute values of magnetic moment and quadrupolar moment and $k_B$ is the Boltzmann constant. The negative value of $K(0)$ indicates that the quadrupolar interaction has an antiferro-type of $\Gamma_5$-symmetry. This result agrees with the experimental result reported by Aléonard et al. where the magneto-striction was isotropic in the transition from the PM phase to the AFM phase [2].

5. Conclusions
We have investigated the temperature dependence of elastic constants for DyCu and determined that the quadrupolar interaction has a $\Gamma_5$-symmetry type. Considering the quadrupolar interaction symmetry we have proposed quadrupolar structures and evaluated the quadrupolar interaction coefficient, $K(0)\mid Q \mid^2 / k_B = -16.8 \text{ K}$, by an analysis of critical fields of metamagnetic transitions. The negative value of $K(0)$ indicates that the quadrupolar interaction has an antiferro-type of $\Gamma_5$-symmetry.

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References
[1] Wintemberger M, Belakhovsky M and Pierre J 1971 Phys. Status Solidi B 48 705
[2] Aléonard R, Morin P and Rouehy J 1984 J. Magn. Magn. Mater. 46 233
[3] Amara M, Morin P and Bourdarot F 1997 J. Phys. Condens. Matter. 9 7441
[4] Amara M and Morin P 1998 J. Alloys Compd. 294 275
[5] Morin P and Schmitt D Ferromagnetic Materials vol 5 (Elsevier, Amsterdam, 1990)
[6] Kakeya I, Kakeshita T, Kindo K, Yamamoto Y and Saburi T 1999 J. Phys. Soc. Jpn. 68 1025
[7] Levy P M 1973 J. Phys. C 6 3545
[8] Demarest Jr H H 1971 J. Acous. Soc. Am. 49 768
[9] Ohno I 1976 J. Phys. Earth 24 355
[10] Yasui M, Terai T, Kakeshita T, Matsuda M, Metoki N and Nojiri H 2008 J. Appl. Phys. 103 07B710