Elastic constants of antiferro-quadrupole ordering system DyB_2C_2

Yuichi Nemoto [a,1], Tatsuya Yanagisawa [a], Koichiro Hyodo [a], Terutaka Goto [a], Shingo Miyata [b], Ryuta Watanuki [b], Kazuya Suzuki [b]

[a] *Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan*

[b] *Graduate School of Engineering, Yokohama National University, Yokohama 240-8501, Japan*

Abstract

Elastic constants of tetragonal system DyB_2C_2 with antiferro-quadrupole (AFQ) ordering have been measured by using ultrasonic method. Remarkable elastic softenings of the C_{44} and $(C_{11} - C_{12})/2$ have been observed towards the AFQ transition at T_Q=25 K. The present results show that a CEF ground state is a pseudo quartet consisting of two Kramers doublets of $E_{1/2}$ and $E_{3/2}$ in C_{4h} site symmetry of Dy^{3+} ion. The dominant softening of C_{44} probably leads to the order parameter of O_{yz} and O_{zx} in AFQ phase. Magnetic phase diagram applied fields along the [100] direction determined by field dependence of the C_{44} is presented.

Key words: antiferro-quadrupole ordering; elastic constant; DyB_2C_2

Recently, rare-earth compounds DyB_2C_2 and HoB_2C_2 have attracted much interest because of the first showing antiferro-quadrupole (AFQ) orderings in tetragonal lattice [1,2]. DyB_2C_2 undergoes to the AFQ phase II at T_Q=25 K and succesively to antiferromagnetic (AFM) phase III at T_N=15 K. In the case of HoB_2C_2, it has been found the AFQ phase II, AFM phase III and phase IV. The phase IV that appears between paramagnetic phase I and AFM phase III coexisting with AFQ is characterized by a considerable elastic softening and enhancement of ultrasonic attenuation of C_{44} [3]. Resonant x-ray scattering has succeeded in showing the presence of the AFQ ordering in DyB_2C_2 and HoB_2C_2 [4,5]. However, the order parameter of the AFQ phase has not been identified yet for both cases. Furthermore, energy level scheme in the tetragonal crystalline electric field (CEF) potential has not been determined by inelastic neutron scattering. In this paper, we present the results of the ultrasonic experiments on single crystals of DyB_2C_2. The AFQ order parameter and CEF ground state in DyB_2C_2 are argued.

[1] Corresponding author. E-mail: nemoto@phys.sc.niigata-u.ac.jp

Fig. 1 shows temperature dependence of the elastic constants C_{66}, C_{44} and $(C_{11} - C_{12})/2$ for transverse waves in DyB_2C_2. Clear anomalies have been observed at the AFQ transition at T_Q=25 K and AFM one at T_N=15 K. It should be noted that remarkable softening of the C_{44} and $(C_{11} - C_{12})/2$ towards T_Q that is responsible for orbital degeneracy of the CEF ground state. On the other hand, the C_{66} increases monotonously with decreasing temperature down to T_Q. Hund's rule multiplet $J = 15/2$ of Dy^{3+} splits into four $E_{1/2}$ and four $E_{3/2}$ Kramers doublets in the tetragonal C_{4h} site symmetry. Taking into account a pseudo quartet state predicted by specific heat showing release entropy of $R\ln 4$ [1], we propose a model consisting of accidentally degenerated two Kramers doublets of $E_{1/2}$ and $E_{3/2}$ that leads to the softening of C_{44} and $(C_{11} - C_{12})/2$ for the CEF ground state. The elastic softenings of C_{44} associated with the quadrupole O_{yz}, O_{zx} and $(C_{11} - C_{12})/2$ with O_2^2 are described by an equation of $C_\Gamma(T) = C_\Gamma^0(T - T_c^0)/(T - \Theta)$ deduced from the quadrupole-strain susceptibility. Furthermore, the dominant elastic softening of C_{44} probably leads to the order parameter of the AFQ phase II to be O_{yz} and O_{zx}.

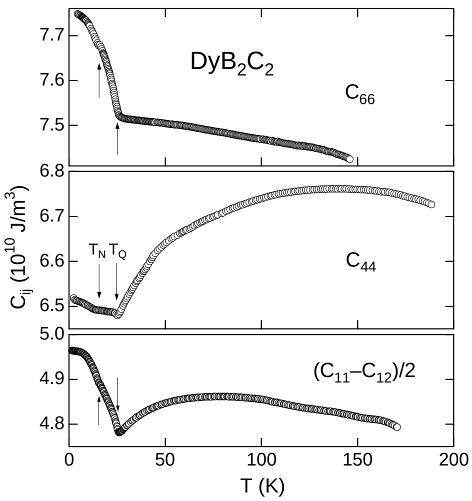

Fig. 1. Temperature dependence of the elastic constants C_{66}, C_{44} and $(C_{11} - C_{12})/2$ of DyB_2C_2.

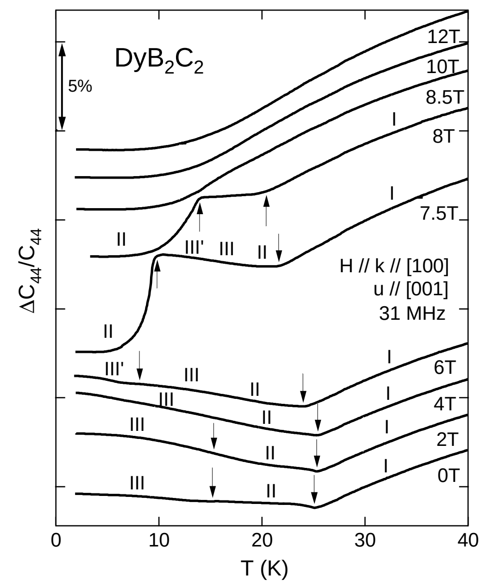

Fig. 2. Temperature dependence of the elastic constants C_{44} in various fixed magnetic fields along the [100] direction.

Results shown in Fig. 2 is the C_{44} in various fixed magnetic fields along the [100] direction. The data below 2 T show the succesive transitions from the paramagnetic phase I through AFQ phase II to AFM phase III as the tempetrature is lowered. However, the II-III phase boundary becomes obscure above 2 T up to 6 T. The results above 10 T indicate no sign of phase transition. In the middle of magnetic fields at 7.5 T and 8 T, several anomalies, that probably associated with transitions to a subphase III$'$ of the AFM III and reentrance

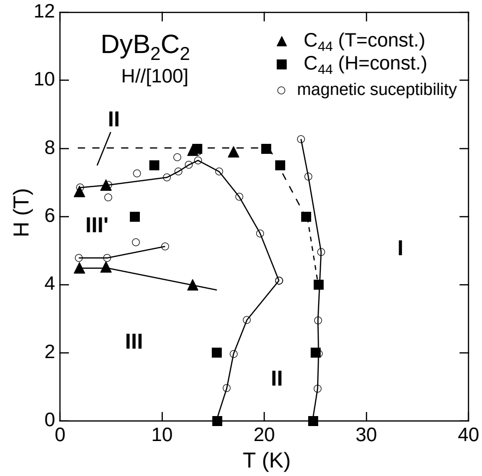

Fig. 3. Magnetic phase diagram of DyB_2C_2 under fields along the [100] direction. Solid triangles and squares are the data obtained from the present study.

to the AFQ II, have been observed. Another important finding is that the softening still remains up to 12 T at higher temperatures, which suggests the quadrupole O_{yz} and O_{zx} plays a key role even in high fields.

The magnetic phase diagram of DyB_2C_2 under fields along the [100] direction is plotted in Fig. 3. Solid traiangles and squares are determined by the present ultrasonic experiments and open circles are obtained from the magnetic susceptibility by Yamauchi et al. Plotted data obtained from the present study is roughly consistent with the previous results [1]. However, the I-II phase boundary may close around 8 T indicated by a broken line obtained from the results of the field dependence of C_{44} that shows upturn anomaly (not shown). For optimum results of anisotropy of the AFQ and AFM interaction, ultrasonic measurements for several elastic constants in magnetic fields along the three pricipal axes are now in progress.

References

[1] H. Yamauchi, H. Onodera, K. Ohoyama, T. Onimaru, M. Kosaka, M. Ohashi, Y. Yamaguchi, J. Phys. Soc. Jpn. **68** (1999) 2507.

[2] H. Onodera, H. Yamauchi, Y. Yamaguchi, J. Phys. Soc. Jpn. **68** (1999) 2526.

[3] T. Goto, T. Yanagisawa, Y. Nemoto, S. Miyata, R. Watanuki, K. Suzuki, J. Phys. Soc. Jpn. **71** (2002) Suppl. 88.

[4] K. Hirota, N. Oumi, T. Matsumura, H. Nakao, Y. Wakabayashi, Y. Murakami, Y. Endoh, Phys. Rev. Lett. **84** (2000) 2706.

[5] T. Matsumura, N. Oumi, K. Hirota, H. Nakao, Y. Wakabayashi, Y. Murakami, J. Phys. Soc. Jpn. **71** (2002) Suppl. 91.