Successive magnetic phase transitions of DyB4

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A tetragonal rare-earth compound DyB4 has been attracting interest as a system where the quadrupolar moment of Dy might be fluctuating because of the geometrical frustration, which is geometrically equivalent to the Shastry-Sutherland model. The Dy lattice can be regarded as a combination of square and triangular connections because the nearest neighbor and the next nearest neighbor connections have almost the same distance.

DyB4 interestingly exhibits two phase transitons at TN1=20.3 K and at TN2=12.7 K. It is already established by neutron powder diffraction that an antiferromagnetic order occurs at TN1 with magnetic moments align along the c-axis and propagate along the [100] direction. What is intriguing is that the entropy of Rln2 still remain and that the elastic softening and absorption are enhanced in the temperature region TN2 < T < TN1. These results indicate that the quadrupolar degeneracy still remains even below TN1. We performed resonant x-ray scattering experiments and have revealed that the magnetic components within the ab-plane are short range ordered in this intermediate temperature region [1]. Below TN2, the in-plane moments order with the corresponding quadrupolar moment <Ozx>, resulting in a uniform structural distortion from tetragonal to monoclinic.

Observation of the short range ordering by resonant x-ray scattering and the elastic softening suggests that the in-plane moments are fluctuating in a relatively long time scale; it looks ordered by x-ray but paramagnetic by ultrasonic measurement. By investigating this phenomenon by neutron diffraction, with intermediate time scale ~THz, we expect that we could clarify if the in-plane moments are really fluctuating and its fluctuation rate.

We have performed neutron diffraction experiment on a single crystalline DyB4. The sample was grown by the floating-zone method using 11-Boron isotope to avoid absorption by 10-Boron in natural boron. The experiment was performed using a triple-axis spectrometer TOPAN, installed at the 6G port of JRR-3 in JAEA. The collimators of 15′-30′-30′-Blank was used with PG filter at neutron energy of 30.5 meV. Since the absorption by natural Dy is extremely large, the diffraction in this sample occurs in a very thin region at the surface less than 0.5 mm. Therefore the comparison of the scattering intensity among different diffraction peaks are impossible. Only the relative variation with temperature are analyzed.

Calculated magnetic structure factor from the magnetic structure model shows that the relative contribution to the intensity by the in-plane moment becomes large with increasing the c-axis component of the scattering vector. The (200) reflection involves no contribution from the in-plane component and reflects only the c-axis component. Then, we have measured the temperature dependences of (100), (101), (102), (103), and (200) reflections and analyzed the results.

Figure shows the temperature dependences of the (200) and (103) reflections. The (200) magnetic reflection arises only from the c-axis component, while the 94% of the structure factor of the (103) magnetic reflection is due to the ab-plane component. The lines in the figure are the calculated temperature dependences for each reflections, assuming the c-axis and ab-plane component illustrated in the bottom figure. The experimental results are well reproduced. This result shows that the in-plane component is indeed very weak in the intermediate temperature region, and it is or-

dered below TN2. Unfortunatelly, because the structure factor of the (103) reflection contains contribution from c-axis component by 6%, we could not tell if the in-plane component in TN2 < T < TN1 is zero or not although it is certainly less than 10%.

[1] D. Okuyama, T. Matsumura, H. Nakao and Y. Murakami, J. Phys. Soc. Jpn., 74, 2434, (2005).

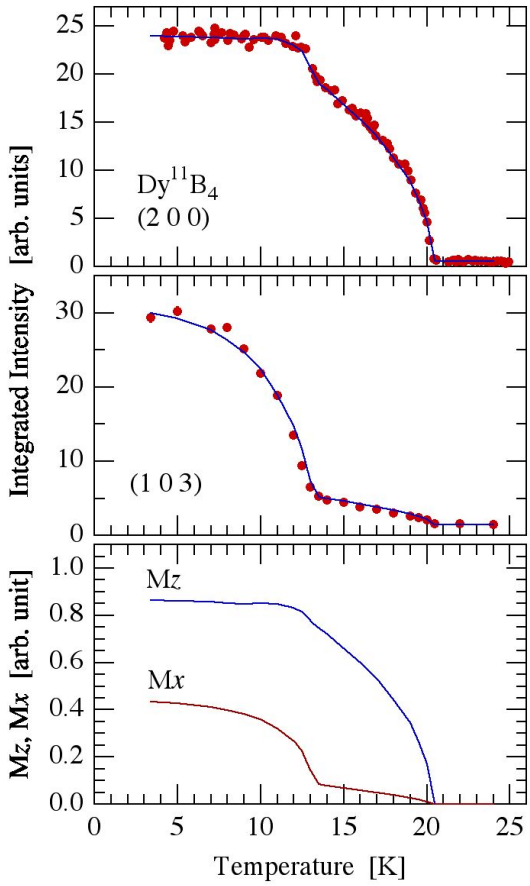

Fig. 1. Temperature dependence of (200) and (103) reflections of DyB4. Lines are calculated temperature dependences. Bottom shows the c-axis (z) and ab-plane (x) components of the magnetic moment assumed for the calculations.