実験報告書様式（一般利用課題・成果公開利用）

MLF Experimental Report	提出日 Date of Report May 12, 2013
課題番号 Project No. 2013A0023 実験課題名 Title of experiment Successive Magnetic Phase Transition Accompanying Drastic Variation in Magnetic Anisotropy of $DyFe_2Zn_{20}$ 実験責任者名 Name of principal investigator Kazuaki Iwasa 所属 Affiliation Department of Physics, Tohoku University	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/(BL No.) AMATERAS (BL14) 実施日 Date of Experiment April 28, 2013 – May 5, 2013

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
Please report your samples, experimental method and results, discussion and conclusions.　Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Co-aligned single crystals of $DyFe_2Zn_{20}$ (6.4 g)

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results.　If you failed to conduct experiment as planned, please describe reasons.

Combinations between rare earth and transition metal elements provide various physical properties in strongly correlated electron systems and cross correlation materials: for example, multiferroic materials and pyrochlore oxides. We usually try to understand material properties by taking a dominant electrons state, *f* or *d*, into account. However, it is interesting to search for cooperative phenomena involving the both electronic states. Here, we focus our attention on magnetic properties in $DyFe_2Zn_{20}$.

$DyFe_2Zn_{20}$ crystallizes in *Fd-3m* space group, in which Dy ions form a diamond lattice. They are surrounded by Zn cages, and Fe atoms are inserted between them. The most interesting magnetic character is that ordering without any anisotropy of magnetization sets in below 50 K, while strong anisotropy appears at the second transition at 25 K. In the lowest temperature phase, a metamagnetic magnetization is observed under a magnetic field of approximately 1.5 T applied along the [001] axis (Y. Isikawa *et al.*: ICM2012). On the other hand, an isostructural compound $DyRu_2Zn_{20}$ does not show such a high-temperature successive magnetic phase transition, and shows only single transition at 4 K. The phase transitions in $DyFe_2Zn_{20}$ can be understood as a result of cooperative interaction between 4*f* electrons located at Dy ions and 3*d* itinerant electrons originating from Fe. The aim of this study is to clarify magnetic excitations composed of both Dy 4*f* electrons and Fe 3*d* electrons, and to discuss how the successive magnetic phase transitions accompanying drastic anisotropy change appears.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

Inelastic scattering measurements were carried out by using the cold-neutron disk-chopper spectrometer AMATERAS (BL14). Multi incident energy mode providing 2.77, 6.40, and 11.67 meV was chosen. Sample temperature between 10 and 50 K was controlled by the GM refrigerator, which was rotated horizontally on the spectrometer to scan within the reciprocal lattice for revealing dispersion relations of magnetic excitations.

Figure 1(a) shows a measured intensity contour map at 32 K in the space of the excitation energy E and the scattering vector $\mathbf{Q} = (Q_a, 2, 2)$. The incident energy is 11.67 meV. Quasi-elastic scattering is observed below approximately 3 meV. This intensity is large near the nuclear Bragg peak positions of $\mathbf{Q} = (0, 2, 2)$ and (4, 2, 2) in the measurements with lower incident energy (not shown here). No other significant signals were detected. Figure 1(b) shows the contour map measured at 10 K. Clear inelastic signals emerges below 2.3 meV. It is notable that distinct dispersion curve arises in the magnetically anisotropic phase below 25 K, in contrast to the magnetic excitation similar to a paramagnetic response in the isotropic ferromagnetic phase between 25 and 50 K (Fig. 1(a)). The maximum excitation energy of 2.3 meV in the dispersion curve appears at the Bragg peak positions $\mathbf{Q} = (2, 2, 2)$ and $\mathbf{Q} = (6, 2, 2)$. These two $\mathbf{Q}$ positions correspond to no contribution of the Dy nuclear diffraction, so that we expect that the observed magnetic excitations originate from motion of the Dy magnetic moments showing the ferromagnetic correlation. However, the steep slope of dispersion curve at $\mathbf{Q} = (0, 2, 2)$, (4, 2, 2) and (8, 2, 2), at which the excitation energy reaches zero, is not consistent with the crystal-field excitations of $4f$ electrons of Dy ions because they usually appear as discrete energy levels corresponding to magnetic anisotropy. Thus, we expect contribution of the magnetic moments of itinerant electron originating from Fe ions to the dispersive magnetic excitations. There is no magnetic excitation observed even at 10 K, except that with clear dispersion relation shown in Fig. 1(b). It means that the magnetic fluctuation concentrates in the lower-energy region which is in the same order of the characteristic temperature of magnetic anisotropy evolution.

We will also perform experiments in order to determine magnetically ordered structure by using a neutron diffractometer. Combination between the present inelastic data and the diffraction information will unveil the f–d correlation phenomena in DyFe$_2$Zn$_{20}$.

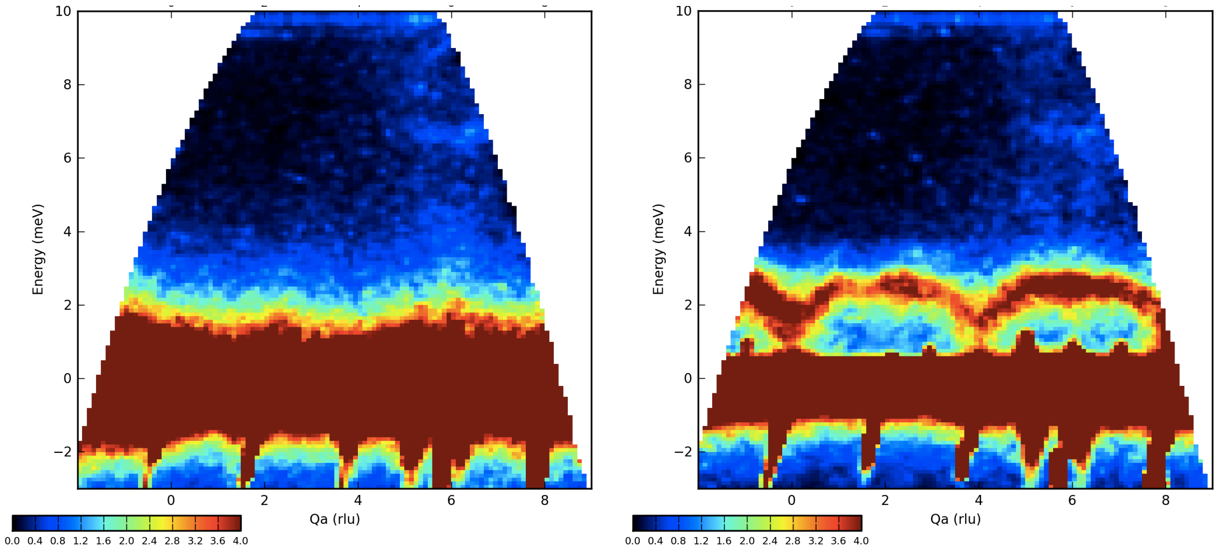

Fig. 1. Q_a–E map of inelastic neutron intensities from DyFe$_2$Zn$_{20}$ (E_i = 11.67 meV) (a) at 32 K, and (b) at 10 K. The scattering vector is $\mathbf{Q} = (Q_a, 2, 2)$. Intensities above 3 meV located at Q_a = 6 originate from scattering of an aluminum sample container.