Magneto Caloric Effect in $(Dy_xGd_{1-x})_3Ga_5O_{12}$ for Adiabatic Demagnetization Refrigeration

Takenori Numazawa [a,1], Koji Kamiya [a], Takahiro Okano [b], Koichi Matsumoto [b]

[a] *Tsukuba Magnet Laboratory, National Institute for Materials Science, Tsukuba, Ibaraki 305-0003, Japan*
[b] *Department of Physics, Kanazawa University, Kanazawa, Ishikawa 920-1192, Japan*

Abstract

Gadolinium and dysprosium gallium garnet single crystals $(Dy_xGd_{1-x})_3Ga_5O_{12}$ ($x = 0, 0.5$ and 1) have been investigated for adiabatic demagnetization refrigeration used as magnetic materials between 0.5 K and 5 K. Specific heat measurement of $(Dy_{0.5}Gd_{0.5})_3Ga_5O_{12}$ showed a large and broad peak similar to that of $Gd_3Ga_5O_{12}$ and it suggests that the geometrical frustration in $Gd_3Ga_5O_{12}$ still remains in$(Dy_{0.5}Gd_{0.5})_3Ga_5O_{12}$. Magneto caloric effect of $(Dy_{0.5}Gd_{0.5})_3Ga_5O_{12}$ was about four times larger than that of $Gd_3Ga_5O_{12}$ for the magnetic field of 1 T between 0.5 K and 5 K. Therefore, the magnetic entropy change of $Gd_3Ga_5O_{12}$ by the external magnetic fields could be enhanced by substituting Dy^{3+} ion for Gd^{3+} ion for the magnetic fields of < 2 T between 0.5 K and 5 K.

Key words: magnetic refrigeration; magneto-caloric effect; magnetic material

1. Introduction

Adiabatic demagnetization refrigeration (ADR) is one of typical cooling methods for low temperatures below 1 K. In particular, ADR does not use working fluids such as ^{4}He or ^{3}He and this is a dominant advantage for space applications under the weak gravity condition. NASA/Goddard Space Flight Center is developing an ADR system for the sensor cooling in space between 0.05 K and 6 K [1].

For the lowest temperature stage of ADR at 0.05 K, ferric ammonium alum (FAA) and chrome potassium alum (CPA) salt pills are used to provide a cooling capacity of $10\mu W$@0.05K with the magnetic field of < 2 T, but for the higher temperature ranges of > 1 K, these salt pills can not provide sufficient cooling capacities unless strong magnetic fields of > 6 T are applied.

On the other hand, several rare-earth garnet crystals show the large magneto caloric effect (MCE) which is caused by the entropy change due to the external magnetic field. This is because the rare-earth elements have large magnetic moments and the large unit cell in the garnet structure gives weak magnetic interactions which provide the lower Neel temperatures.

In particular, $Gd_3Ga_5O_{12}$ (GGG) shows an unique specific heat characteristics which has a broad and large peak around 0.85 K. This corresponds to the magnetic short range order in Gd^{3+} ions based on the magnetic frustration caused by the unusual structure of garnet lattice [2]. The frustration will provide a large entropy and therefore, we have interest in the mixture system $(Dy_xGd_{1-x})_3Ga_5O_{12}$ where the Dy^{3+} ion may provide a strong anisotropic magnetic interaction into the frustration system [3].

In the present paper, we chose three Dy-Gd garnets of GGG (x=0), DGGG (x=0.5) and DGG (x=1) for the space application. Heat capacity and magnetization were measured to estimate the enhancement on MCE of DGGG between 0.5 K and 5 K with the magnetic field up to 5 T.

[1] E-mail:numazawa.takenori@nims.go.jp

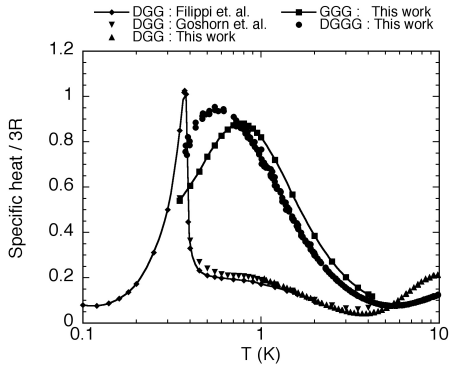

Fig. 1. Specific heat of GGG (x=0), DGGG (x=0.5) and DGG (x=1) for zero magnetic field. The data of DGG below 1 K are taken after Filippi et. al.[5] and Goshorn et. al.[6].

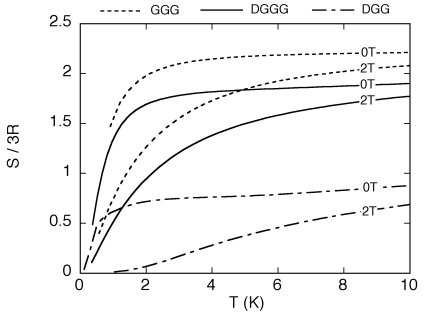

Fig. 2. Entropy diagram of GGG, DGGG and DGG as functions of temperature and magnetic field.

2. Results and Discussion

The crystals were grown with [111] direction by CZ method. X-ray analysis showed that both Dy^{3+} and Gd^{3+} ions are distributed randomly and homogeneously in the $(Dy_xGd_{1-x})_3Ga_5O_{12}$ crystals for [16a] and {24*c*} sites [4]. A relaxation heat flow method was used for heat capacity measurements from 0.4 K to 20 K with magnetic fields up to 5 T. For magnetization measurements, a SQUID magneto meter was used. The direction of applied magnetic field was perpendicular to [111] of the sample.

Figure 1 shows the heat capacity C of GGG, DGGG and DGG. DGGG keeps a large and broad peak similar to GGG and it is apparently different from the lambda type transition appeared in DGG. Since we are interested in only the electronic contribution, nuclear one is neglected here. The entropy S can be calculated by integrating the function C/T, but there is the error for estimating the S nearby $T = 0$. Thus we also calculated the entropy change by the magnetization data for the higher temperatures [7], then the entropy data were decided by comparison with both results.

Figure 2 shows the calculated results for the entropy diagram. Since only Kramer's doublet remains for DGG in the temperatures, the saturation entropy at 0 T of DGG is $\sim$ln(2), while GGG has the larger value of $\sim$ln(8). The saturation value of DGGG is larger than [ln(8) + ln(2)] / 2 = 1.4, this suggests that the geometrical frustration in GGG still remains in DGGG sample.

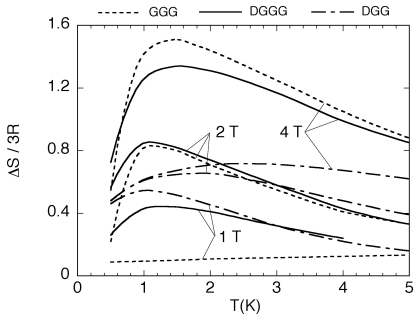

Fig. 3. Entropy change of GGG, DGGG and DGG as functions of temperature and magnetic field.

Figure 3 shows the entropy change ΔS as a function of temperature calculated from Fig.2 for the magnetic fields of 1, 2 and 4 T. DGG has large ΔS for 1 T, but the ΔS becomes to be saturated for > 2 T. This is because the zero magnetic field entropy of DGG is only $\sim$ln(2) and it makes the limit of the maximum entropy change small. On the other hand, GGG and DGGG have the higher entropy values at 0 T and ΔS can be increased with the magnetic field of 4 T. It is noticeable that ΔS of DGGG is considerably larger than that of GGG for 1 T. This enhancement on MCE will come from the substitution of Dy^{3+} ion to Gd^{3+} ion. Such properties of DGGG is suitable for space ADR between 0.5 K and 5 K in low magnetic field operation.

References

[1] P. Shirron, E. Canavan, M. DiPirro, J. Tuttle, C. Yeager, Adv. Cryo. Eng. **45B** (2000) 1629.

[2] P. Schiffer, A.P. Ramirez, D.A. Huse, A.J. Valentino, Phys. Rev. Lett. **73** (1994) 2500.

[3] T. Numazawa, H. Kimura, M. Sato, H. Maeda, K. Shimamura, T. Fukuda, Adv. Cryo. Eng. **42** (1996) 459.

[4] H. Kimura, T. Numazawa, M. Sato, H. Maeda, J. Matt. Sci. Lett. **13** (1994) 1164.

[5] J. Filippi, J.C. Lasjaunias, A. Ravex, F. Tcheou, J. Rossat-Mignold, Solid State Commun. **23** (1997) 613.

[6] D.P. Goshorn, D.G. Onn, J.P. Remeika, Phys. Rev. **B15** (1997) 3527.

[7] K. Matsumoto, T. Okano, A. Matsuzaki, K. Kamiya, T. Numazawa, subimitted to LT23 proceedings.