

Magnetic Properties of TmFe_2O_4

J. Kim and B. W. Lee*

Department of Physics, Hankuk University of Foreign Studies, Yongin, Kyunggi 449-791, Korea

(Received 26 February 2010, Received in final form 11 March 2010, Accepted 12 March 2010)

We studied the magnetic properties of TmFe_2O_4 . The magnetization measurements revealed the magnetic ordering of Fe spins at around 240 K. The difference between zero-field-cooled (ZFC) and field-cooled (FC) magnetization was close to the thermoremanent magnetization (TRM), indicating the glass behavior exhibited by this material.

Keywords : TmFe_2O_4 , magnetic transition, thermoremanent magnetization

1. Introduction

Recently, the mixed valence material LuFe_2O_4 was found to exhibit ferroelectricity associated with the charge ordering leading to Fe^{2+} and Fe^{3+} ions [1], and to display coupling between the dielectric response and the magnetic field even at room temperature [2]. LuFe_2O_4 is a member of the family of rare earth iron oxides $R\text{Fe}_2\text{O}_4$ ($R=\text{Y, Ho, Er, Tm, Yb}$ and Lu), which have a rhombohedral structure (space group $R\bar{3}m$) [3]. It consists of the alternate stacking of the Fe-O triangular lattice layers and the R-O blocks along the c -axis. The average valence of the Fe ions is $\text{Fe}^{2.5+}$, which implies that equal numbers of Fe^{2+} and Fe^{3+} ions occupy the equivalent Fe sites on the hexagonal net plane. Strong magnetic interactions between the localized Fe moments induce magnetic ordering below 240 K [4-7]. The magnetic phenomena depend on the rare-earth metal element R . The magnetic transition is observed at 230-240 K and a structural one occurs at 190-200 K for $R=\text{Y}$ or Er [4, 5]. However, $R=\text{Tm, Yb}$ and Lu compounds do not show this structural transition but rather a thermoremanent magnetization (TRM) below 220 K [6, 7]. The induction of a ferromagnetic moment by cooling the sample in an external magnetic field results in TRM, which is explained by a successive freezing process of the small ferromagnetic clusters [8]. In this paper, we experimentally investigate the TRM properties of TmFe_2O_4 by conducting dc magnetization and hysteresis experiments under warming and cooling processes.

2. Experiments

A stoichiometric polycrystalline sample of TmFe_2O_4 was prepared by solid-state reaction method. High purity powders (99.99%) of Tm_2O_3 and Fe_2O_3 were mixed, ground, and sintered at 1200°C in a stream of CO/CO₂ gas for 24 h. Powder x-ray diffraction (XRD) patterns were measured using $\text{CuK}\alpha$ radiation. Magnetization measurements were taken with a commercial vibrating sample magnetometer (Lake Shore, model 7300). The temperature dependence of magnetization $\sigma(T)$ was measured in both zero-field-cooled (ZFC) and field-cooled (FC) modes. The ZFC magnetization was measured while heating in a field after zero-field cooling to 15 K. The FC magnetization was performed while cooling in a field. Additionally, TRM was measured by field cooling the sample from room temperature down to 10 K in a constant field and reducing the field to zero.

3. Results and Discussion

Fig. 1 shows XRD pattern for polycrystalline TmFe_2O_4 . The diffraction peaks were indexed with respect to the rhombohedral structure with a space group of $R\bar{3}m$. The lattice parameters were $a=3.4743$ and $c=25.0210$ Å.

The $\sigma(T)$ was measured in various applied fields. The results measured in external fields of $H=0.1$ and 1 kOe are shown in Fig. 2. The FC magnetization data were obtained while cooling the sample in a magnetic field (FCC), and heating the sample in a field after cooling in a field (FCW). The shapes of the magnetization curves are almost identical for the two different fields. Both FC and

*Corresponding author: Tel: +82-31-330-4362
Fax: +82-31-333-1696, e-mail: bwlee@hufs.ac.kr

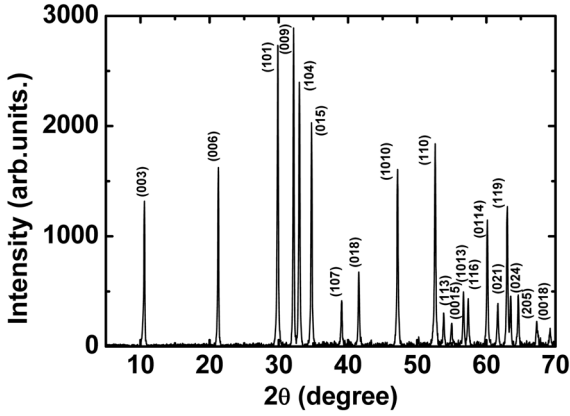


Fig. 1. Powder x-ray diffraction (XRD) pattern for polycrystalline TmFe_2O_4 (space group $R\bar{3}m$).

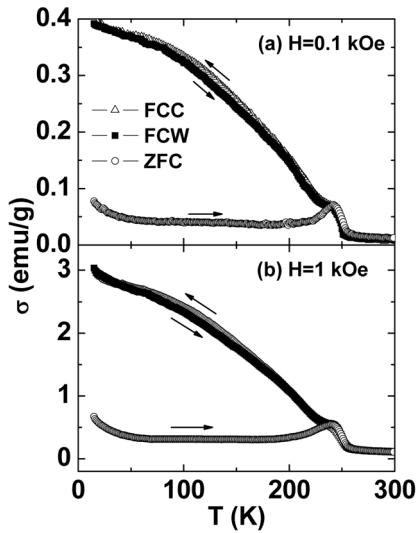


Fig. 2. Temperature dependence of magnetization $\sigma(T)$ for TmFe_2O_4 , measured with external fields of $H=0.1$ and 1 kOe. FCC, FCW and ZFC stand for field-cooled-cooling, field-cooled-warming and zero-field-cooled magnetization, respectively.

ZFC curves exhibit a magnetic transition at around 240 K, as reported for other $R\text{Fe}_2\text{O}_4$ oxides [4-7]. The ZFC curve has a broad maximum at around 240 K, and the appearance of irreversibility between the ZFC and FC curves starts at a temperature of just above 240 K. The field-cooling effect is observed in the FC magnetization below 240 K, where the magnetization curve has a peak. The cooling of the sample under a magnetic field grows magnetic domains along the field direction, leading to separation between the FC and ZFC magnetization (i.e., $\sigma_{\text{FC}} > \sigma_{\text{ZFC}}$). Unlike the ZFC magnetization case, the FC magnetization rises continuously with decreasing temperature. Such behavior has been observed in cluster glass materials [9] where magnetic interactions within the cluster

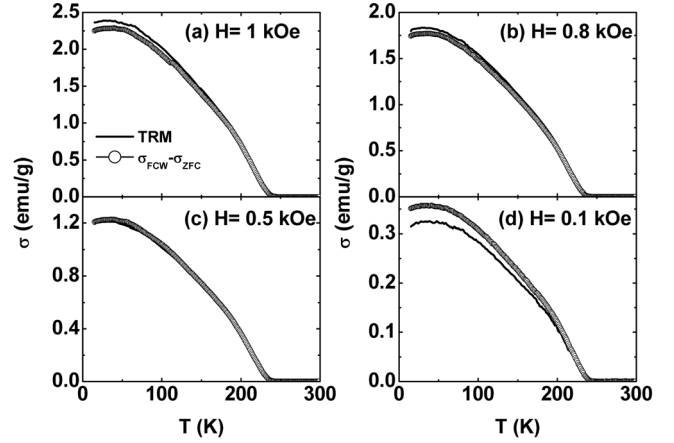


Fig. 3. Temperature dependence of TRM and the difference between FC and ZFC magnetization ($\sigma_{\text{FC}} - \sigma_{\text{ZFC}}$) for TmFe_2O_4 with various magnetic fields.

tend to align the moments in the direction of the applied field, thereby increasing the FC magnetization. The figure also shows the absence of thermal hysteresis between the FCC and FCW curves throughout the temperature range. However, thermal hysteresis appears between FC curves below 200 K in LuFe_2O_4 [10].

The sample was cooled in various external magnetic fields down to 10 K, and TRM was measured while heating the sample after reducing the field to zero. The temperature dependence of TRM and the difference between the FC and ZFC curves of magnetization ($\sigma_{\text{FC}} - \sigma_{\text{ZFC}}$), obtained by subtracting the corresponding experimental data, are shown in Fig. 3. Since the thermoremanent state is obtained by cooling a sample in a magnetic field from a temperature at which all moments fluctuate, to one at which some of the fluctuations are quenched, the temperature variation of TRM is explained by a successive freezing process of the small ferromagnetic clusters [8]. From TRM data, the magnetic phase

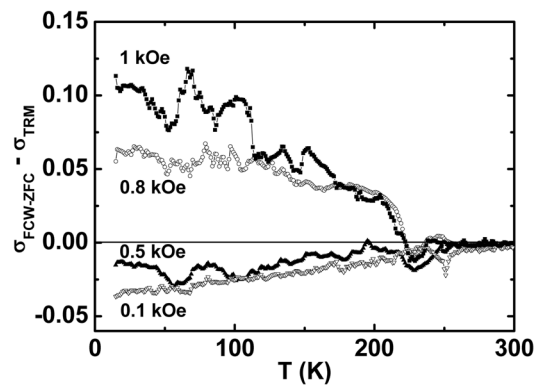


Fig. 4. Difference between $\sigma_{\text{FCW-ZFC}}$ and σ_{TRM} of TmFe_2O_4 in various magnetic fields.

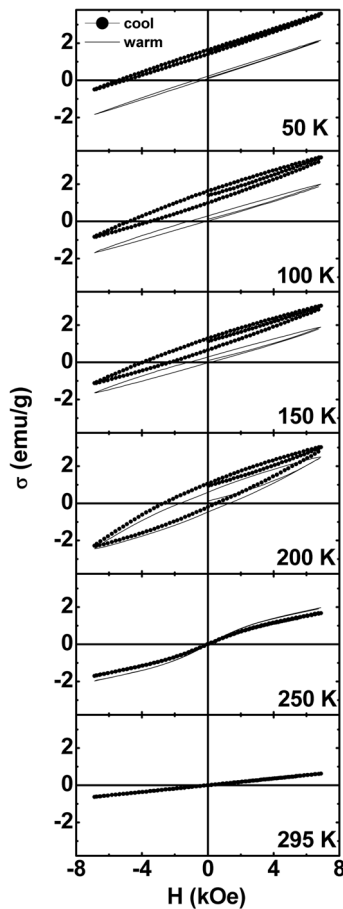


Fig. 5. Magnetic hysteresis of TmFe_2O_4 measured at various temperatures during cooling and warming processes.

transition temperature was clearly estimated to be 240 K. Even though TRM depends on the external field, the shape of the TRM curves is not affected by the external field. The temperature dependence of TRM is close to that of $\sigma_{\text{FC}} - \sigma_{\text{ZFC}}$.

The difference between $\sigma_{\text{FCW-ZFC}}$ and σ_{TRM} of TmFe_2O_4 in various magnetic fields is shown in Fig. 4. The slope of the difference depends on the external fields. Although a coincidence of functions $\sigma_{\text{FCW-ZFC}}$ and σ_{TRM} may be expected in spin-glass, in cluster-glass a deviation from this behavior may arise due to anisotropy of the clusters, possibly associated with their shape and orientation [11].

Fig. 5 shows the magnetic hysteresis measured at various temperatures during cooling from room temperature to 10 K and warming from 10 K to room temperature. The shift of the magnetic hysteresis towards positive magnetization can be observed below 240 K in the cooling process, but not in the warming process. This shift means that the spin may be frozen in directions energetically favored by their local anisotropy or by the external field if the sample is cooled down in zero or nonzero

field, respectively, leading to a difference between ZFC and FC magnetization [12].

4. Conclusion

We investigated the magnetic properties of TmFe_2O_4 . The magnetization measurements revealed the magnetic ordering of Fe spins and the appearance of irreversibility between FC and ZFC magnetization at around 240 K. The difference between FC and ZFC magnetization was close to TRM. The difference between $\sigma_{\text{FCW-ZFC}}$ and σ_{TRM} depended on the external magnetic fields, indicating the glass behavior of this compound. The shift of the magnetic hysteresis towards positive magnetization was observed below 240 K in the cooling process.

Acknowledgments

This research was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea Government (MEST) (No. 20090075966). This work was supported by Hankuk University of Foreign Studies Research Fund of 2009.

References

- [1] N. Ikeda, H. Ohsumi, K. Ohwada, K. Ishii, T. Inami, K. Kakurai, Y. Murakami, K. Yoshii, S. Mori, Y. Horibe, and H. Kito, *Nature* **436**, 1136 (2005).
- [2] M. A. Subramanian, T. He, J. Chen, N. S. Rogado, T. G. Calvarese, and A. W. Sleight, *Adv. Mater.* **18**, 1737 (2006).
- [3] N. Kimizuka, A. Takenaka, Y. Sasada, and T. Katsura, *Solid State Commun.* **15**, 1321 (1974).
- [4] Y. Nakagawa, M. Inazumi, N. Kimizuka, and K. Siratori, *J. Phys. Soc. Jpn.* **47**, 1369 (1979).
- [5] J. Iida, M. Tanaka, H. Kito, and J. Akimitsu, *J. Phys. Soc. Jpn.* **59**, 4190 (1990).
- [6] K. Yoshii, N. Ikeda, and A. Nakamura, *Physica B* **378**, 585 (2006).
- [7] K. Yoshii, N. Ikeda, Y. Matsuo, Y. Horibe, and S. Mori, *Phys. Rev. B* **76**, 024423 (2007).
- [8] J. Iida, M. Tanaka, Y. Nakagawa, S. Funahashi, N. Kimizuka, and S. Takekawa, *J. Phys. Soc. Jpn.* **62**, 1723 (1993).
- [9] S. Mukherjee, R. Ranganathan, P. S. Anilkumar, and P. A. Joy, *Phys. Rev. B* **54**, 9267 (1996).
- [10] J. Iida, Y. Nakagawa, and N. Kimizuka, *J. Phys. Soc. Jpn.* **55**, 1434 (1986).
- [11] N. A. Belous, I. A. Zorin, N. V. Kulich, I. V. Lezhnenko, and A. I. Tovstolytkin, *Sov. Phys. Solid State* **32**, 887 (1990).
- [12] R. Laiho, K. G. Lisunov, E. Lähderanta, P. Petrenko, J. Salminen, V. N. Stamov, and V. S. Zakhvalinskii, *J. Phys. Condens. Matter* **12**, 5751 (2000).