Magnetism of Fe Ion in FeSr$_2$YC$_{2}$O$_{6+\delta}$ Superconductor

Y. Hata, T. Mochiku$^1$, T. Wuernisha$^2$ and H. Yasuoka

Department of Applied Physics, National Defense Academy, Kanagawa, 239-8686, Japan
$^1$Superconducting Materials Center, National Institute for Materials Science, Ibaraki, 305-0047, Japan
$^2$International Center for Materials Nanoarchitechtonics, National Institute for Materials Science, Ibaraki, 305-0047, Japan

E-mail: hata@nda.ac.jp

Abstract. FeSr$_2$YC$_{2}$O$_{6+\delta}$ compounds with various oxygen contents, $6 + \delta = 7.02 \sim 7.55$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compound were synthesized and studied by the magnetization measurement. FeSr$_2$YC$_{2}$O$_{7.55}$ compound shows superconductivity below $T_c = 56$ K and a broad peak at 20 K, which corresponds to the magnetic ordering of Fe ion, in the magnetization measurement. The Curie constants and the asymptotic Curie temperature strongly depend on the oxygen content. Increasing the oxygen contents, the Curie constants and the absolute values of the asymptotic Curie temperature are increased. Since the asymptotic Curie temperature is negative value, the interaction between Fe ions is mainly antiferromagnetic. A hysteresis is observed in the magnetization curve of non-superconducting FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compound. It implies that the magnetism of Fe ion is not simple antiferromagnetism.

1. Introduction

Coexistence of superconductivity and long-range magnetic order is a problem of widespread interest. The substitution for the Cu site in YBa$_2$Cu$_3$O$_{6+\delta}$ compounds with a transition metal element has a strong effect on superconductivity, driving $T_c$ down at various rates[1]. The substitution of the magnetic ions for Cu site is a subject of interest to study the interplay between superconductivity and magnetism, especially ferromagnetism.

The substitution of Fe ion for Cu ion in YSr$_2$Cu$_3$O$_{6+\delta}$ is interesting, however it was also reported that it caused poor superconductivity[2]. To improve the superconducting property of FeSr$_2$YC$_{2}$O$_{6+\delta}$, it is reported that the reoxidization annealing and the high pressure O$_2$ annealing are effective[3, 4, 5]. It is considered that the reoxidization annealing modifies the Fe ion distribution and the high pressure O$_2$ annealing supplies an adequate holes to CuO$_2$ plane.

Coexistence of ferromagnetism and superconductivity was discovered in the 1212-type ruthenocuprates RuSr$_2$RCu$_2$O$_8$ (R=Eu and Gd). RuSr$_2$GdCu$_2$O$_8$ exhibit a Curie transition at 132K and bulk superconductivity below 46 K [6]. In comparison with these compounds, the study of the magnetism of FeSr$_2$YCu$_2$O$_{6+\delta}$ compounds is interesting.

In this paper, we focus on the magnetism of Fe ion in FeSr$_2$YC$_{2}$O$_{6+\delta}$ compounds. To study the magnetism of Fe ion, FeSr$_2$YCu$_2$O$_{6+\delta}$ compounds with various oxygen contents were synthesized and studied by magnetization measurement. To suppress the superconductivity, a part of Cu ion in CuO$_2$ plane was substituted by Zn ion.
2. Experiment

The FeSr$_2$YCu$_2$O$_{6+\delta}$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{6+\delta}$ compounds were synthesized by the solid-state reaction of mixture of Y$_2$O$_3$, SrCO$_3$, CuO, Fe$_2$O$_3$ and ZnO powders. The mixture was calcined at 900°C for 24 h in air, ground, and pressed into pellets. The pellets were sintered at 1000°C for 24 h in air. The pellets were subsequently annealed at 800 °C for 24 h in N$_2$ flow, at 300 °C for 24 h in the mixed O$_2$ and N$_2$ flow. The sample with the oxygen content $6 + \delta = 7.55$ was annealed at 350 °C in high O$_2$ pressure of 200 atm. The oxygen content, $6 + \delta$ was determined by the change of weight and the result of the powder neutron diffraction study[4].

The resistivities were measured by the conventional four-probe method. Magnetization and magnetic susceptibility were measured using a SQUID magnetometer (MPMS-XL, Quantum Design).

3. Result and discussion

Figure 1 shows the temperature dependence of the magnetization for FeSr$_2$YCu$_2$O$_{7.55}$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds under the magnetic field of 10 Oe. Diamagnetism is observed below $T_c = 56$ K in FeSr$_2$YCu$_2$O$_{7.55}$ compound. While FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds shows the hysteresis of ZFC and FC data below 20 K, diamagnetism does not observed. In the temperature dependence of resistivity measurement, it shows semiconducting like behavior and any sign of superconductivity was not observed.

Figure 2 shows the temperature dependence of the magnetization for FeSr$_2$YCu$_2$O$_{6+\delta}$ under the magnetic field of 10 kOe. The peaks around 20 K are due to the magnetism of Fe ion, since this temperature is not correspond to the superconducting transition temperature shown in Fig. 1. The susceptibility above 100 K were refined by the Curie-Weiss function,

$$\chi = \frac{C}{T - \theta_p} + \chi_0$$

(1)

where $C$ is the Curie constant and $\theta_p$ is the asymptotic Curie temperature and $\chi_0$ is the component of the susceptibility which is independent of the temperature. The oxygen content dependence of these parameters are shown in Fig. 3. Increasing the oxygen content, the Curie constant and the absolute value of the asymptotic Curie temperature are increased. This fact indicates that the magnetism of Fe ion is strongly affected by the oxygen content. Decreasing the oxygen content, the Fe ion seems to become non magnetic. The magnetic interaction between Fe ions is mainly antiferromagnetic since the asymptotic Curie temperatures are negative value.

Figure 4 shows the magnetization curves of FeSr$_2$YCu$_2$O$_{7.55}$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds at 2 K. The magnetization curve of FeSr$_2$YCu$_2$O$_{7.55}$ compound consists of a paramagnetic like magnetization and a typical hysteresis loop of the mixed state in type-II superconductor. The former is considered to be due to the magnetism of Fe ion. The large diamagnetism of superconductivity makes it difficult to study the magnetism of Fe ion in FeSr$_2$YCu$_2$O$_{7.55}$ compounds. A hysteresis is observed in non superconducting FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds. Its coercive force is about 2 kOe and it implies that the magnetism of Fe ion is not simple antiferromagnetism.

In the case of RuSr$_2$RCu$_2$O$_8$ compound, Jorgensen et al. concluded that the magnetism of Ru is an antiferromagnetic order by powder neutron diffraction study[7]. From the magnetization measurement, they also concluded that RuSr$_2$RCu$_2$O$_8$ compound are weak ferromagnets. Around Fe ion, oxygen defects exist in FeSr$_2$YCu$_2$O$_{7.55}$ compound. The defects distribute at random[8]. In the case of FeSr$_2$Cu$_2$O$_{6+\delta}$ compounds, the oxygen defects affect the magnetism of Fe ion and it is considered that the hysteresis in magnetization curve of FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compound is originated from the lowering the symmetry of the Fe site caused by the oxygen defects. To determine the magnetism of Fe ion in detail, powder neutron diffraction study or $^{57}$Fe Mössbauer spectroscopy study at low temperature are needed.
4. Conclusion

Fe$_{2}$YCu$_{2}$O$_{6+\delta}$ compounds with various oxygen contents, 6 + $\delta$ were synthesized and studied by the magnetization measurement. The Curie constant and the asymptotic Curie temperature strongly depend on the oxygen content. Increasing the oxygen content, the Curie constant and the absolute value of the asymptotic Curie temperature are increased. Since the asymptotic Curie temperature is negative value, the interaction between Fe ions is mainly antiferromagnetic. A hysteresis is observed in the magnetization curve of FeSr$_{2}$Y(Cu$_{0.95}$Zn$_{0.05}$)$_{2}$O$_{7.55}$ compound. It implies that the magnetism of Fe ion is not simple antiferromagnetism.

![Figure 1](image1)

Figure 1. Temperature dependences of the magnetization of FeSr$_{2}$YC$_{2}$O$_{6+\delta}$ and FeSr$_{2}$Y(Cu$_{0.95}$Zn$_{0.05}$)$_{2}$O$_{7.55}$ compounds under the magnetic field of 10 Oe. The data of FeSr$_{2}$Y(Cu$_{0.95}$Zn$_{0.05}$)$_{2}$O$_{7.55}$ were enlarged 10 times.

![Figure 2](image2)

Figure 2. Temperature dependences of the magnetization of FeSr$_{2}$YC$_{2}$O$_{6+\delta}$ compounds under the magnetic field of 10 kOe. The bold lines indicate the fitted lines by Eq. 1.

References
[1] X. Z. Zhou, M. Raudsep, Q. A. Pankhurst, A. H. Morrish, Y. L. Luo and I. Maartense, Phys. Rev. B 36 (1987) 7230.
[2] T. Terziev, R. Suryanarayanan, Mamidanna S. R. Rao, L. Ouhammou, O. Gorochov and J. L. Dormann, Phys. Rev. B 48 (1993) 13037.
[3] J. Shimoyama, K. Otzuchi, T. Hinouchi and K. Kishio, Physica C 341-348 (2000) 563-564.
[4] T. Mochiku, Y. Mihara, Y. Hata, S. Kamisawa, M. Furuyama, J. Suzuki, K. Kadowaki, N. Metoki, H. Fujii and K. Hirata, J. Phys. Soc. Jpn. 71 (2002) 790.
[5] Y. Hata, Y. Mihara, J. Suzuki, I. Kakeya, K. Kadowaki, E. Kita and H. Yasuoka, Physica C 417 (2004) 17.
[6] C. Bernhard, J. L. Tallon, Ch. Neidermayer, Th. Blasius, A. Golnik, E. Brucher, R. K. Kremer, D. R. Noakes and C. E. Stronach, Phys. Rev. B 59 (1999) 14099.
[7] J. D. Jorgensen, O. Chmaissem, H. Shaked, S. Short, P. W. Klamut, B. Dabrowski and J. L. Tallon, Phys. Rev. B 63 (2001) 054440.
Figure 3. Oxygen content dependence of the Curie Constants $C$, the asymptotical Cure temperatures $\theta_P$ and $\chi_0$. Open circles and filled circle indicate FeSr$_2$YCu$_2$O$_{6+\delta}$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds, respectively.

Figure 4. Magnetization curves of FeSr$_2$YCu$_2$O$_{7.55}$ and FeSr$_2$Y(Cu$_{0.95}$Zn$_{0.05}$)$_2$O$_{7.55}$ compounds at 2 K.

[8] H. Fujii, Y. Mihara, T. Mochiku, Y. Hata and K. Kadowaki, Physica C 415 (2004) 85.