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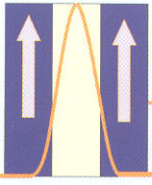

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Magneto-electric coupling changed by proton irradiation in the $CoCr_2O_4$

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Introduction

In multiferroic materials, ferromagnetism and ferroelectricity coexist. Especially, when polarization occurs due to certain types of ordering in the system, it is known as improper ferroelectricity[1]. $CoCr_2O_4$ materials have Co2+ ions on the A sites and Cr3+ on the B sites of the spinel structure. Especially, ferroelectric transition has been observed in a $CoCr_2O_4$ upon the transition to the spiral spin behavior about 28 K. P. Esquinazi et al. show that proton irradiation on graphite samples controls ferro- or ferrimagnetism [2]. Proton irradiation measurements induce lattice transformation or various distortion effects. In this article, we have investigated the crystallographic and magnetic structure of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ by proton irradiation. We present crystallographic and magnetic properties of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ using the x-ray diffractometer (XRD) with particular emphasis on Mössbauer spectroscopy for $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$.

Experiment and results

Magnetic properties of proton-irradiated $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ compounds has been studied with x-ray diffraction (XRD), magnetization, Mössbauer spectroscopy measurements and compared with non-irradiated $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$. The crystal structure of the proton irradiated sample is determined to be a cubic structure of Fd-3m with lattice constant a_0 = 8.339 Å. As shown figure 1, for $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2, the ZFC (zero field cooled) data with an applied field of 100 Oe shows a completely different behavior in magnetization curve when compared with non-irradiated sample. It shows negative magnetization and enhancement of the gap in step-like phase transition at 13 and 27 K, up to 1 kOe. Furthermore, the maximum value of magnetization with applied field of 1 T for proton irradiated $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ with 10 pC/μm2 has been increased from 5.42 emu/g to 5.78 emu/g. Mössbauer spectra of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2 were taken at various temperatures ranging from 4.2 to 100 K as shown in Fig. 2. Below 26 K, the Mössbauer spectra of the $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2 were least-square fitted to two sets of six Lorentzian lines. While temperature increases up to 26 K, the six-line patterns of Mössbauer spectra still persists, but the hyperfine field decreases gradually. From the Mössbauer spectra of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2, we carefully suggest that two different types of iron ion exist below 26 K. This agrees with x-ray refinement results that the octahedral structure has two magnetic behaviors of the B1 site (outer sextet) and B2 site (inner sextet) in Cr3+ ions. The inner sextet of Mössbauer spectra below 26 K was observed to be broader than the outer-sextet was, because different electric charge distribution induced by proton irradiation can affect the orbital contribution in magnetic hyperfine field and electric quadrupole shift. Since the enhancement of orbital contribution induces spin-orbital coupling, a reduction of quadrupole shift in B2 sites and increase in saturation magnetization in the $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2 are expected. While there is only one type of iron ion corresponding to one sextet of spectra above 30 K, this anomaly point corresponds to the temperature of the induced multiferroism in $CoCr_2O_4$.

In summary, we suggest that the change in spin-orbital coupling and exchange interaction due to proton irradiation bring forth an changed ferroelectricity and magnetism.

[1] S-W Cheong and Maxim Mostovoy, Nature materials, 6, 13 (2007).

[2] P. Esquinazi, D. Spemann, R. Hohne, A. Setzer, K.-H. Han, and T. Butz, Phys. Rev. Lett., 91, 227201 (2003).

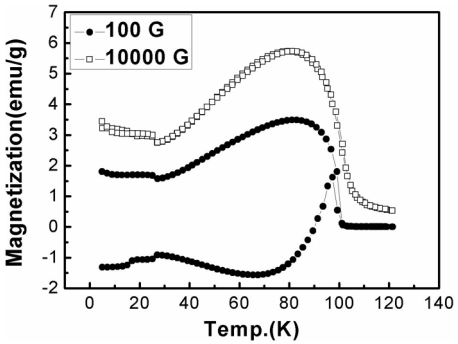

Fig. 1 Magnetizaiton data of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$ irradiated by proton with 10 pC/μm2 at various temperatures under applied fields of 100 and 10000 Oe.

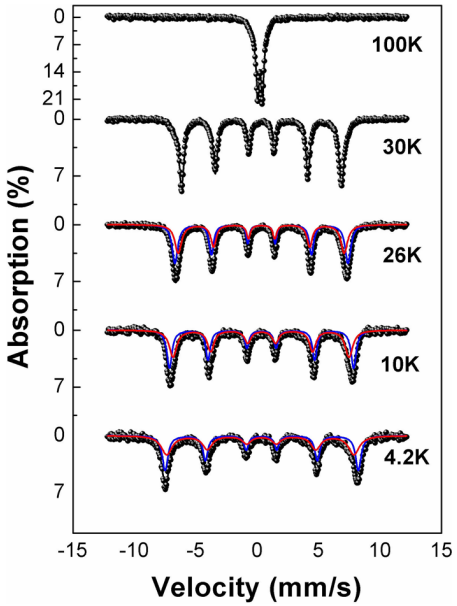

Fig. 2 Mössbauer specta of $CoCr_{1.98}{}^{57}Fe_{0.02}O_4$irradiated by proton with 10 pC/μm2 at various temperatures.