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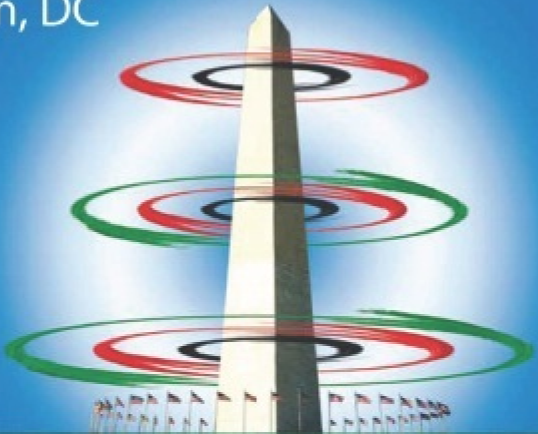
DIGESTS



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The magnetic phase changing for titanium oxide with proton irradiation.

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Introduction

Diluted magnetic semiconductors (DMS) have been increasing scientific interest as promising candidates for the spintronic devices. Recent research indicate ferromagnetism in graphite by proton irradiation [1]. Coey et al. proposed the F-center exchange model where ferromagnetic coupling is promoted by an electron trapped in an oxygen vacancy [2]. Griffin et al. observed the ferromagnetic properties in insulating Co-doped TiO_2 annealed in ultrahigh vacuum, suggesting that free charge carriers are not required for ferromagnetic ordering [3]. P. Esquinazi et al. provide that proton irradiation on graphite samples triggers ferro- or ferrimagnetism [4]. In this work, we have investigated the magnetic properties of anatase Fe-doped TiO_2 by proton irradiation.

Experiments

Anatase $Ti_{0.99}{}^{57}Fe_{0.01}O_2$ compounds were fabricated by a sol-gel process. The solution, which was dissolved in mixed solvents [acetic acid : 2-methoxyethanol = 1 : 3] was refluxed at 80 °C for 12 h to allow the gel formation and then dried at 120 °C in a dry oven for 24 h. The dried mixtures were ground and annealed at 550 °C for 2 h in air. The ^{57}Fe doped TiO_2 of a diameter 5 mm pellets were irradiated with proton beam (6.66 MeV, 26 nA). Two irradiations were consecutively applied to sample, stage NO. 1: homogeneous irradiation, NO. 2: dose: 1 pC/μm^2 (1 pC), No.3: dose: 10 pC/μm^2 (10 pC). To estimate the defect density created by the proton beam, Monte Carlo simulations (SRIM 2004) were performed. The crystal structures of the samples were examined by x-ray diffraction with Cu Kα radiation (λ = 1.5406 Å). Magnetic properties were characterized by superconducting quantum interference device magnetometer (SQUID). The Mössbauer spectra were recorded using conventional and electromechanical spectrometer with a ^{57}Co source in a rhodium matrix.

Results and Discussion

X-ray diffraction patterns of $Ti_{0.99}{}^{57}Fe_{0.01}O_2$ with proton irradiation showed a pure anatase single phase and the crystal structure was determined to be a tetragonal structure with a space group $I4_1/amd$. In both patterns above, one could not find any different peak positions of Fe or Fe-O systems other than anatase dioxide within the instrumental resolution limit. The lattice constants a_0 and c_0, and the resultant unit-cell volume of the present anatase samples are found to be close to the ones of TiO_2 with its lattice constants a_0= 3.786 Å and c_0= 9.520 Å obtained by the same fabrication process.

The magnetization curves of 1 and 10 pC proton irradiated $Ti_{0.99}{}^{57}Fe_{0.01}O_2$ compound were measured as a function of magnetic field using SQUID. Figure 1 exhibits magnetic hysteresis loops at room temperature (RT) for the anatase samples measured up to the field of 6 T. 1 pC sample shows a small magnetic moment with the value of 0.08 emu/g at RT. On the contrary, the magnetization curves of 10 pC sample show a strongly enhanced ferromagnetic behavior.

Figure 3 exhibits Mössbauer spectra of 1 and 10 pC samples at RT. The spectra of 1 pC sample consist of the magnetically ordered sextet and the paramagnetic doublet. The isomer shift values at RT for the sextet and the doublet of 1 pC sample are found to be 0.42 and 0.21 mm/s relative to the Fe metal, respectively, which are consistent with the high spin Fe^{3+} charge state. The electric quadrupole splitting (ΔE_Q) of central doublet at RT was 1.12 mm/s. The 10 pC spectra were substantially different from the spectra of 1 pC sample. The new doublet for 10 pC sample is attributable to the Fe^{2+} state with isomer shift value of 1.00 mm/s and large ΔE_Q= 2.08 mm/s at RT. It could explain that some of Fe^{3+} ions of 10 pC sample are converted to Fe^{2+}, which enhanced the ferromagnetic properties. Therefore, it is suggested that the created Fe^{2+} ions as a result of proton irradiation are responsible for the observed ferromagnetic enhancement in this system.

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[3] K. A. Griffin *et. al*, Phys. Rev. Lett. 94, 157204 (2005).
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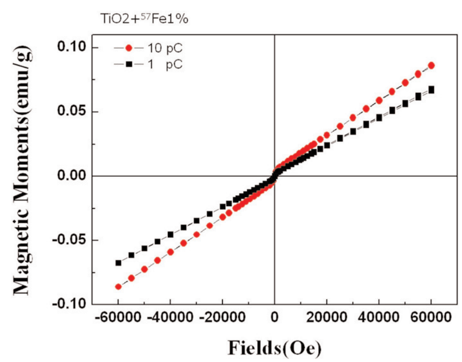

Fig. 1 Magnetic hysteresis loops of $Ti_{0.99}{}^{57}Fe_{0.01}O_2$ with proton irradiation at RT.

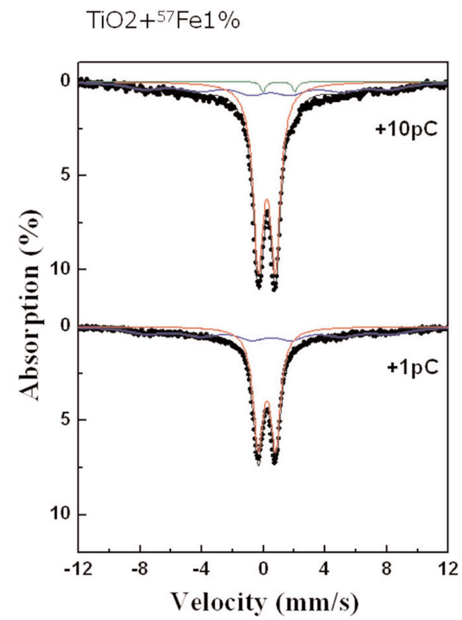

Fig. 2 Mössbauer spectra of Fe doped TiO_2 samples with proton irradiation at RT.