Room Temperature Ferromagnetism in Hydrogenated Co-doped ZnO Bulk Samples

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We report room temperature ferromagnetism (RTFM) in hydrogenated $Zn_{0.96}Co_{0.04}O$ bulk samples synthesized via a standard solid state reaction route. In a previous work, we reported the absence of intrinsic ferromagnetism in high-quality bulk samples synthesized in oxygen atmosphere at 1400^{o}C. We concluded that the presence of Co is not a sufficient condition to induce a ferromagnetic behavior. In this work, we submitted the $Zn_{0.96}Co_{0.04}O$ samples to a hydrogenation annealing process in order to introduce defects, as an attempt to induce RTFM.

Paramagnetic $Zn_{0.96}Co_{0.04}O$ samples were annealed in a gaseous mixture of argon 95% and hydrogen 5%. The effects of hydrogenation on the structural properties were investigated by x-ray diffraction (XRD). The microstructure and composition distributions were characterized by scanning electron microscopy (SEM) and energy dispersive x-ray (EDS) measurements. Raman scattering was used to study of the incorporation of dopants and the resulting lattice disorder of the host lattice. Co K-edge x-ray absorption near-edge structure (XANES) was used to determine the valence state of Co in the ZnO lattice. Changes in the density of defects were estimated by Hall and Photoluminescence (PL) measurements. Magnetic characterizations were performed using a superconducting quantum interference device (SQUID) magnetometer.

The structural characterization confirmed the absence of secondary phases or Co clusters. Hall Effect showed an increase on the carrier concentration associated to the induction of defects due to the hydrogenation process. PL revealed the presence of an acceptor-like band associated to zinc vacancies and a second band of donor-like character, confirming the Hall assumptions. The magnetic measurements revealed a two-phase behavior, with the coexistence of paramagnetic and ferromagnetic phases at room temperature. The magnetization saturation increased from 0.006 emu/g to 0.4 emu/g with the hydrogenation process.

We discuss the origin of the RTFM characteristic observed solely for hydrogenated $Zn_{0.96}Co_{0.04}O$ samples, which we attribute to the presence of zinc vacancies and other defects at oxygen sites.

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