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Transport properties in iron–iron oxide film near percolation threshold

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Abstract

Iron–iron oxide granular films are fabricated using dc magnetron sputtering. Their structural, magnetic and transport properties are studied in the vicinity of percolation threshold. X-ray photoelectron spectroscopy and transmission electron microscopy confirm the coexistence of iron and FeO_x. Magnetic measurement indicates the exchange coupling between the iron and the FeO_x matrix. Metal–insulator transition is observed. The non-monotonic temperature dependencies of resistivity occur near the percolation threshold and the mechanism is investigated with the longitudinal and transverse magnetic transport measurement in terms of weak localization.

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Keywords: Magnetic films and multilayers; Electronic transport; Magnetic measurements

1. Introduction

During the last several years, iron oxide thin films have been extensively investigated for their unusual magnetic properties [1–3]. In theses studies, iron oxide systems were prepared by various methods, including reactive rf sputtering from iron targets in the mixed oxygen and argon gas and cold compacting [4] or by chemical method [5]. The structural and magnetic properties of these systems were found to strongly depend on the fabrication process in detail. For the mechanical method, the volume component is easy to control but it can only form bulk material and the chemical method is convenient to fabricate discontinuous nanoparticle collection. The high vacuum sputtering method provides good choice to get the thin film structure. Though the precise volume component is hard to determine, it is convenient to get various percolation structure by modulating either the sputtering source power or ratio of argon and oxygen flow.

Magnetic studies on iron–iron oxide fine particles show both coercivity enhancement and exchange bias effect at low temperatures [6]. However, very few attentions have been paid on the magnetic transport property of such systems, such as Hall and magnetoresistance effects. In the vicinity of the percolation threshold, the geometry configuration of system changes from a metallic network to a structure with separated particles. In this region the physical property may differ much from that either in the metal or the insulator side when near the critical point. Indeed the giant Hall resistivity has been observed in this region in the Ni–SiO_2 or Cu–SiO_2 systems [7,8]. It is instructive to investigate the transport property near percolation in Fe/Fe-oxide system so as to get a better understanding of physical process in this region.

In this paper we fabricate granular iron–iron oxide films near percolation threshold by co-sputtering and measured their electronic and magnetic properties. The mechanism of the transport process is discussed along with magnetic property.

2. Experiments and measurements

The granular iron–iron oxide films are fabricated by reactive dc sputtering with a pure Fe target at room temperature. The base

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pressure of the chamber is better than 1×10^{-7} Torr and the pressure of Ar for sputtering is maintained at about 4×10^{-3} Torr during deposition. The flow rate of oxygen is varied, and the ratio of oxygen and argon are 1:400, 1:100 and 1:40 for sample S01, S02, S03, respectively. The deposition rates keep in the range of 6 Å/s. Both glass and Kapton substrates are used so that the transport and magnetic properties can be separately measured. Pure iron film is also prepared to make comparison. The average thickness of film measured by a Dektak 3 surface profiler is about 400 nm.

The structure of films is studied by X-ray diffraction (XRD) and transmission electron microscopy (TEM). X-ray photoelectron spectroscopy (XPS) is used to analyze the composition of the films. The magnetic measurements are carried out by a commercial superconducting quantum interference device (SQUID) magnetometer. Electronic transport measurements are performed in physical properties measurement systems (PPMS). The standard four-probe method is used to measure the longitudinal and the Hall resistivity.

3. Results and discussion

X-ray diffraction spectrum is employed to determine the crystal structure, where the diffraction patterns of three samples with different identify the existence of iron phases whereas signal is very weak. The peak at $2\theta = 44.8^\circ$ is consistent with the (1 1 0) orientation of bulk cubic center (bcc) iron, exhibiting the texture structure. No distinguishable peak of iron oxide is found, which is probably for the poorly crystallized at the high deposition rate. TEM morphology is taken to further investigate the microstructure of the film. Form the selected area electron diffraction patterns, both bcc α-Fe and FeO, Fe_2O_3 are recognized, as shown in Fig. 1. The granular type structure with nano-scaled Fe particles embedded the iron oxide matrix can be identified. In order to confirm the valence state of iron in our samples, we utilized XPS technique which is sensitive to

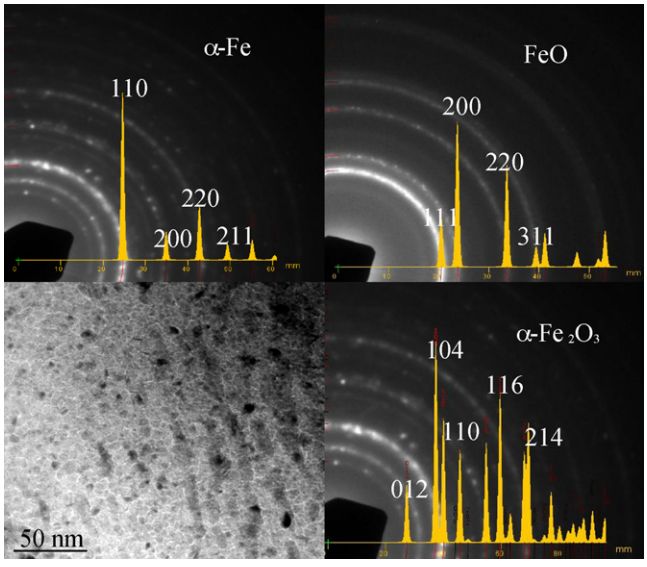

Fig. 1. Bright field TEM image and selected area electron diffraction patterns of S02.

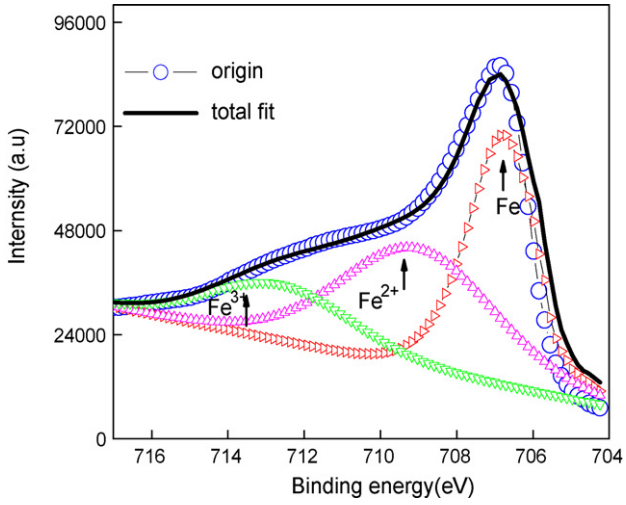

Fig. 2. XPS spectrum of Fe 2p3/2 with computer fitting curve of S02.

detect different chemical species in the sample. On account of the XPS results are also sensitive to the sample surface, the top of film about 5 nm thick has been etched before the measurement. Therefore the complexity of surface can be obviated. For sample S02 the asymmetric broadening of the higher energy edge peak can be decomposed to three separate characteristic peaks of the Fe, Fe^{2+} and Fe^{3+} at 706.6, 709 and 711.3 eV, as shown in Fig. 2. By fitting the XPS data we estimated the atom ratio of Fe, Fe^{2+} and Fe^{3+} is 43.7%, 30.4% and 25.9%, respectively, exhibiting a structure near percolation threshold.

3.1. Magnetic property

Magnetic properties are performed in SQUID with the film deposited on the Kapton. First the samples are cooled from 300 to 10 K with a magnetic field at 20,000 Oe applied parallel to the film plane. Then M–H loops are measured at different temperatures. Fig. 3 is the M–H loops of sample S02 at 10 K. The loop becomes asymmetrical and shifts to negative field axis. The exchange bias field H_E is defined as $(H_{CR} - H_{CL})/2$, where H_{CR} and H_{CL} are the coercivities at right and left

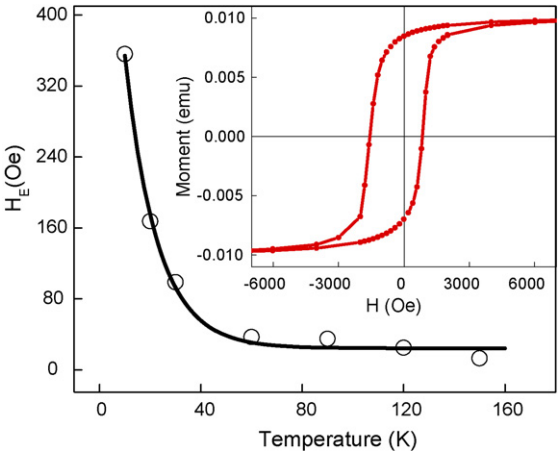

Fig. 3. The H_E dependence on temperature, the fitted line is just for convenience to see. Inset is the hysteresis loop of S02 after cooled with 20,000 Oe.