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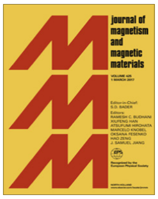

Research articles

Electrical transport properties in Fe-Cr nanocluster-assembled granular films

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ABSTRACT

The $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films with Cr atomic fraction (x) ranging from 0 to 100 were fabricated by using a plasma-gas-condensation cluster deposition system. The TEM characterization revealed that the uniform Fe clusters were coated with a Cr layer to form a Fe-Cr core-shell structure. Then, the as-prepared $Fe_{100\text{-}x}Cr_x$ nanoclusters were randomly assembled into a granular film in vacuum environments with increasing the deposition time. Because of the competition between interfacial resistance and shunting effect of Cr layer, the room temperature resistivity of the $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films first increased and then decreased with increasing the Cr atomic fraction (x), and revealed a maximum of 2×10^4 μΩ cm at $x = 26$ *at.%*. The temperature-dependent longitudinal resistivity (ρ_{xx}), magnetoresistance (MR) effect and anomalous Hall effect (AHE) of these $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films were also studied systematically. As the x increased from 0 to 100, the ρ_{xx} of all samples firstly decreased and then increased with increasing the measuring temperature. The dependence of ρ_{xx} on temperature could be well addressed by a mechanism incorporated for the fluctuation-induced-tunneling (FIT) conduction process and temperature-dependent scattering effect. It was found that the anomalous Hall effect (AHE) had no legible scaling relation in $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films. However, after deducting the contribution of tunneling effect, the scaling relation was unambiguous. Additionally, the $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films revealed a small negative magnetoresistance (MR), which decreased with the increase of x. The detailed physical mechanism of the electrical transport properties in these $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films was also studied.

1. Introduction

The electrical transport properties in magnetic materials, which involve the influence of temperature or magnetic field on the transport of electrons, were always the key issue in the field of condensed matter physics [1–3]. According to the direction relationship between the voltage and current, the electrical transport study can be simply divided into two main issues (longitudinal and transversal). For longitudinal electrical transport issue (measuring the voltage parallel to current direction), the research focused on the temperature-dependence of resistivity, giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR), etc [4–7]. While for the transversal part (measuring the voltage perpendicular to current direction), the researches always included the anomalous Hall effect (AHE), planar Hall effect (PHE), giant Hall effect (GHE), etc [8–10]. The GMR and AHE were the most significant features of the electrical transport properties in the magnetic materials. In the field of GMR, the study of magnetoresistance (MR) can be traced back to the work of Thomson et al., who found a very small positive MR in the iron resulted from the spin-orbit coupling effect of conduction electrons and named as anisotropic magnetoresistance (AMR) [11]. Although AMR had been put into practical application, such as magnetic field sensors, the low magnetic sensitivity and small MR value limited its further performance. Subsequently, A. Fert [12] and P. Grünberg [13] independently found a large negative magnetoresistance in magnetic Fe/Cr multilayer films. The saturated MR was approximately -50% at 4.2 K, which could be ascribed to the spin-dependent scattering

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of conduction electrons at interface [14]. The discovery of GMR was a great breakthrough in the condensed matter physics, quickly gained widespread applications in the field of information storage, and won the 2007 Nobel Prize. Next, Xiao et al. also found a large negative magnetoresistance in Co-Cu granular films, which indicated that the GMR effect was not restricted to multilayer-structured films [15]. The large GMR effect in granular films was originated from spin-dependent scattering at interface [16,17]. Compared with multilayer systems, the granular films also received much attention because of the simple preparation method and isotropy in nature, which was very important for practical application. In the next few years, much work has focused on the GMR effect in granular films, such as Co-Cu [18], Co-Ag [19], Fe-Cu and Fe-Ag [20], all of which were no solubility in the solid state. Meanwhile, people also expected to observe the large GMR effect in Fe-Cr granular films because of its stronger spin-orbit coupling. However, there were only few of work about the GMR in Fe-Cr granular films because it was difficult to obtain the Fe-Cr granular films by the conventional preparation method due to the considerable solid solubility limit between Fe and Cr at room temperature [21]. Fujimori et al. prepared the Fe-Cr heterogeneous alloy by rf sputtering method and observed a maximum magnetoresistance value of 37.3% at 4.2 K [22,23]. Meanwhile, Brüx et al. also observed a 26% GMR value in Fe-Cr quenched-state bulk alloys at 5 K [24]. The large magnetoresistance observed in Fe-Cr heterogeneous alloy confirmed that the Fe-Cr granular films may also possess large magnetoresistance value. Hence, it is necessary to develop a new method to prepare the Fe-Cr granular films and explore their electrical transport properties.

On the other hand, the AHE was another spin-dependent transport property in magnetic materials. In the recent years, more and more attention have been devoted to the AHE in the magnetic materials because of the widely application prospect in magnetic field sensor and magnetic random access memory [3]. In 1995, Pakhomov et al. found the saturated Hall coefficient in co-sputtered Ni-SiO_2 granular films was almost four orders of magnitude greater than that of pure nickel and named as giant Hall effect (GHE) [10]. The large Hall coefficient and small temperature coefficient of resistance made the granular films to be a good candidate for high sensitivity Hall sensors. More important, a lot of research results indicated that the scaling relation in granular films deviated from the classical scaling law, but the physical mechanism was still in dispute [25]. Therefore, the AHE in the inhomogeneous Fe-Cr granular films was also an interesting subject.

In this work, the $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films with different Cr atomic fraction (x) were fabricated by using a plasma-gas-condensation cluster deposition (PGCCD) system. The uniform Fe nanoclusters obtained by plasma-gas-condensation technology and coated with a Cr layer by conventional magnetron sputtering *in situ* in a vacuum, were soft landed onto the substrate and assembled into Fe-Cr granular films with Fe cluster distributed in the nonmagnetic Cr matrix. The influence of Cr atomic fraction (x) on the temperature-dependent resistivity, MR and AHE in Fe-Cr nanocluster-assembled granular films were investigated systemically. The physical mechanism of AHE and the scaling relation in inhomogeneous granular films was discussed. The reason for small MR observed in Fe-Cr nanocluster assembled granular films was also explored.

2. Experiment

2.1. Preparation of the samples

The $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films with different Cr atomic fraction (x) ranging from 0 to 100 were prepared by using a PGCCD system. The equipment schematic is shown in Fig. 1. This equipment has three functional chambers: sputtering chamber, filtering region and deposition chamber. The three chambers are connected by gas dynamic nozzles (5 mm in diameter), and each chamber has independent pumping system to maintain the pressure difference. Two pieces of Fe targets (purity 99.99%) with a diameter of 76.2 mm and thickness of 2 mm were fixed face to face with a distance of about 100 mm in sputtering chamber. A Cr target (purity 99.99%) with a diameter of 76.2 mm and thickness of 4 mm was installed on the conventional magnetron sputtering target located at the top of the deposition chamber. After the background pressure was evacuated below 6×10^{-4} Pa, the Ar gas (purity 99.999%) with a flow rate of 400 sccm adjusted by a fine mass flow controller, was continuously injected into the sputtering chamber to prepare the Fe nanoclusters. Meanwhile, 20 sccm of Ar gas was also injected into the deposition chamber to sputter the Cr films. During the sputtering process, the work pressure was maintained at about 90 Pa for sputtering chamber and 0.4 Pa for deposition chamber. A direct current (DC) power supply with an output power of 400 W was applied to generate the Fe vapor from the facing targets. After colliding with the inert Ar atoms, the Fe vapor was cooled down, nucleated and formed clusters. The detailed preparation process could be seen elsewhere [26]. Then, the insert gas accompanied with the as-formed Fe clusters were extracted from the sputtering chamber through the nozzle by the pressure difference of the adjacent chambers. When the Fe clusters flow through the glow discharge area of Cr target located on the top of the deposition chamber (area in dotted box), the Fe clusters was coated by a layer of Cr *in situ* in a vacuum. The Cr atomic fraction (x) ($0 \le x \le 100$ *at.%*) can be adjusted by controlling the power of the Cr target without changing the preparation parameters of the Fe clusters. Finally, the Fe-Cr clusters accompanied by a few of Cr atoms were soft landed onto the substrate and random self-assembled into $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films. The deposition rate of Fe and Fe-Cr nanoclusters ware measured by a quartz oscillator type thickness monitor installed on the top of deposition chamber. The thicknesses of $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films for Cr power of 0 W, 12 W, 30 W, 60 W, and 110 Ware 448 nm, 557 nm, 680 nm, 898 nm, 1067 nm respectively. It should be noted that all the $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films contain the same amount of Fe.

2.2. Structural characterization

The microstructures of $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films were characterized by X-ray diffractometer (XRD, Bruker D8-Advance) with Cu Kα radiation. The transmission electron microscope (TEM, JEM-2100&F30) and selected area electron diffraction (SAED) were used to determine the morphology and the crystalline phases of films. The morphology and the compositions of the films were analyzed by a scanning electron microscope (SEM, SU-70). The samples patterned into Hall bar with five contacts by stainless steel cover template, was used to measure the longitudinal resistivity (ρ_{xx}) and Hall resistivity (ρ_{xy}) simultaneously (see inset of Fig. 1). The Quantum Design physical property measurement system (PPMS-9) operated in the magnetic field range -50 k Oe < H < 50 k Oe was used to test the magnetic and electrical properties with temperatures ranging from 5 to 300 K. The thickness of $Fe_{100\text{-}x}Cr_x$ nanocluster-assembled granular films was determined by a surface profiler (Alpha-Step D-100) and confirmed by the SEM cross-section images.

3. Results and discussion

Fig. 2(a) shows low-magnitude TEM image of the pure Fe clusters deposited onto the Cu grid with effective thickness of