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Giant Magnetoresistance and Coercivity of electrodeposited multilayered FeCoNi/Cu and CrFeCoNi/Cu

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ABSTRACT

The effect of Cr addition on electrodeposited multilayered nanowires CrFeCoNi/Cu was investigated from a magnetic property perspective: current perpendicular to the plane-Giant Magnetoresistance (CPP-GMR) and Coercivity (BH loops). The magnetic behavior of multilayered nanowires of CrFeNiCo/Cu was also affected by the alloy deposition potential, alloy pulsing time (layer thickness) and number of bilayers. Furthermore, the addition of Cr influenced both the nanowires GMR and Coercivity. Cr addition to the ferromagnetic FeCoNi layer induced a reduction in the room temperature GMR from 10.64% to 5.62%; however, the magnetic saturation field decreased from 0.45 to 0.27 T. The increase in the number of bilayers, from 1000 to 2500, resulted in a higher GMR value, 14.56% with 0.35 T magnetic saturation field. Addition of Cr to the ferromagnetic layer decreased the coercivity from 0.015 to 0.0054 T. Low saturation field CPP-GMR nanowires showing low coercivity at room temperature opens a new door for magnetic sensing devices. To the best of our knowledge, this is the first study on electrodeposited CrFeCoNi/Cu multilayered nanowires.

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1. Introduction

The discovery of Giant Magnetoresistance (GMR) has brought a revolutionary change in the field of magnetic storage and sensing devices [1,2]. GMR based devices are able to detect the presence of weak magnetic fields, which makes them “sensitive sensors” materials, opening a new research direction in the field of spintronics [1,2]. Fert et al. [3] were the first to observe (CIP-GMR), current in plane of multilayers, in an alternating magnetic and non-magnetic Fe/Cr thin films obtained by vapor technique (Molecular Beam Epitaxy). Thin films only allow CIP-GMR measurements, where the GMR phenomenon, is limited by the mean free path (λ) of the electrons [4] and the multilayers' angstrom size thickness. However, multilayered nanowires inherent geometry allows for the CPP-GMR (current perpendicular to the plane) measurement, which depends on the electrons' spin diffusion length [4], which allows the fabrication of thicker layers and shows higher signal to noise ratio. Valet and Fert theoretical model [4] anticipated that the current perpendicular to the plane (CPP-GMR) would lead to larger changes in resistance with magnetic fields. The GMR effect is based on the scattering of electrons at multilayers' interfaces [2,5]. In the presence of a magnetic field, the majority charge carriers easily pass through the magnetic multilayers when the lines of magnetization are parallel to the majority carriers, therefore contributing to a low resistivity. In the absence of a magnetic field, the ferromagnetic layers anti-ferromagnetically couple, and both majority and minority carriers are scattered strongly in the alternating ferromagnetic layers, thus leading to a higher resistivity [2]. High-aspect-ratio structures such as multilayered nanowires are ideal candidates for CPP-GMR measurements. Multilayered nanostructures having a sandwich-like arrangement (alternating the ferromagnetic layer separated by a nonmagnetic spacer) favors the CPP geometry, showing higher values than the thin film CIP-GMR [6]. The enhanced value of CPP-GMR motivated the interest towards the GMR magnetic sensors, which have various applications, such as contactless sensors without mechanical gear [7], detection of magnetic beads with functionalized biomarkers by integrating into microfluidic devices [8], position detection of magnetized patterned objects with application in robotic field, magnetometers, compasses [9,10], etc. Furthermore, due to their inherent nanometric size, nanowire based GMR devices have the advantage of nanoscale magnetic sensing applications, specifically in the detection of high density stored magnetic bits for computer hard drives [10,11]. Spallas et al. [12] fabricated, tested and

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compared GMR devices and reported that the CPP response (39%) was twice the CIP response. Furthermore, CIP-GMR films for read heads are reaching their performance limits, with GMR ratios of 10%–20%. A CIP-GMR film with 20% MR can be applied to a 100–200 Gbpsi recording density; however, CPP-GMR films show promise for the next generation of read heads with recording densities well over 500 Gbpsi [13].

Thin films represent an alternative geometry, which includes convoluted vapor techniques such as chemical vapor deposition, molecular beam epitaxy, magnetron sputtering and the melt spinning methods [1,2,6]. Compared to other mentioned techniques, electrodeposition is the method of choice for fabricating high-aspect-ratio structures [6,14]. The use of electrodeposition technique in developing multilayered nanowire for CPP-GMR structures has shown not only the ease and convenience in fabricating nanowires but also the cost effectiveness [6,14]. Although, electrodeposited GMR values are lower compared to vapor techniques [15], electrodeposition offers the advantage of a single electrolyte, controlled layer composition, room temperature fabrication, the ability to deposit metal in deep nanometric grooves and achieve thousands of layers for high-aspect-ratio nanostructures with high signal to noise ratio [6,14,15].

Since after the breakthrough of GMR phenomena [1–3], several research groups have investigated various elemental systems in electrodeposited multilayered nanowire array [16–18,21–26]. One of the pioneer research group, Piraux et al. [16] studied the electrodeposition of multilayered nanowires of Co/Cu system using commercially available polycarbonate membranes, and observed a 15% GMR at room temperature. Co/Cu system was also studied by Liu et al. [17] and Blondel et al. [18] who reported 11% and 14% room temperature GMR, respectively. The addition and combinations of various elements in the ferromagnetic layer was shown to have the potential for higher CPP-GMR [6]. Specifically, when Ni was added to the Co rich layer not only that the corrosion resistance improved [19], but also it reduced coercivity, an essential factor for sensitive sensor behavior. Magnetic materials having Ni in its alloy show high permeability, large saturation and remnant magnetization, and lower coercivity, which vary with grain size and magnetic domain orientation [20]. Schwarzacher et al. [21] and Blondel et al. [22] examined the addition of Ni into Co layer in electrodeposited CoNiCu/Cu multilayered nanowires and observed high GMR at room temperature, 22% and 20%, respectively. With the addition of Fe to the CoNi alloy, a more complex system of FeCoNi showed an anomalous co-deposition behavior [23]. The quaternary multilayered thin films CoNiFeCu/Cu were electrodeposited by Huang et al. [24] who reported a CIP-GMR of 6% at room temperature but at high saturation fields. High GMR values were reported at an extremely high magnetic saturation field, which makes the GMR material impractical for industrial applications. [25,26]. Piraux et al. [26] observed a decrease in magnetic saturation fields for the trilayered nanowires obtained in aluminum oxide templates. At low temperature (4.2 K) and high magnetic fields (9 T), 80% CPP-GMR was obtained for the typical multilayered NiFe/Cu system, while a 19% CIP-GMR, at low saturation fields, was observed for the trilayered nanowires [26]. In our study, the highest applied magnetic field was 1 T, even though all our samples saturated at an even lower magnetic field of less than 0.5 T and at room temperature. Osaka [27] observed high saturation magnetic flux density and low coercivity in electrodeposited FeCoNi thin films and reviewed the relation between composition and the magnetic properties of CoNiFe ternary alloys. Davis et al. [28] were the first to study CoNiFeCu/Cu in the form of nanowires and measured 20% CPP-GMR at low saturation fields (0.2 T) and room temperature.

Thin films containing Cr were fabricated by Dolati et al. [29] who studied the FeCrNiMo alloy thin films and observed that a higher Cr content lead to fine-grains, smooth and compact alloy deposition. Xin-Kuai et al. [30] investigated the electrodeposition of Cr from a trivalent bath and also observed smooth, fine (less than 100 nm) grains, which result in discrete layers and lower coercivity. Another practical consideration of these GMR magnetic sensing materials is the percentage change in resistivity with magnetic field (the slope of GMR versus H), which is referred as sensitivity. Daniels and Clemens [31] studied the effect of Cr in Fe/Cr multilayers GMR obtained by a sputtering technique, and observed that the magnetic saturation field decreased with Cr doping to the Fe layer and the sensitivity increased in these films. They reported that an increase in preferential spin-independent scattering at Cr sites was due to the introduction of more scattering centers in the Fe layer. Segregation of Cr to grain boundaries resulted in an increase of spin-dependent scattering. Another effect of Cr alloying in the magnetic layer is that the required magnetic saturation field decreases, since the antiferromagnetic coupling strength weakens and it is consistent with the reduction in the magnetic moment. Their study concluded that the magnetic field required to saturate the FeCr/Cr multilayers decreases with the inclusion of Cr due to the decrease in the strength of the antiferromagnetic coupling. By alloy Fe with 20% Cr, the slope of the GMR versus H curve increased, which translated to lower coercivity and higher sensitivity. As the average grain size in nano-crystalline materials can approach critical magnetic length scales such as domain wall thickness or the ferromagnetic exchange length, the magnetic behavior changes. When the grain size is reduced to the extent that the domain wall thickness is comparable to the grain size, the coercivity is found to dramatically decrease. Softening should occur as soon as the structural correlation length or grain size becomes smaller than the ferromagnetic exchange length, which is in the order of the domain wall width [32,33].

To the best of our knowledge, CrFeCoNiCu alloy has not been yet electrodeposited and this is the first study of alternating layers of CrFeCoNi and Cu in confined deep recesses such as nanowires. Furthermore, this study also presents the CrFeCoNi/Cu magnetic behavior of these multilayered nanowires. Due to the shape anisotropy, in the case of parallel nanowire array, the magnetic dipolar interactions drastically modifies their magnetic behavior, resulting in a decrease of coercive field [34], which is a desirable feature for magnetic sensors. The goal of our study is to electrodeposit nanowires with higher CPP-GMR and low coercivity. The less coercive force is an essential factor for sensitive magnetic sensing devices. The present study on multilayered nanowire of CrFeCoNi/Cu focuses on investigation of the Cr addition effect on the magnetic layer and the resulting magnetic properties. Magnetic CrFeCoNi alloy layer deposition potential, pulsing time and bilayers number were investigated from a GMR and Coercivity prospective.

2. Experimental

The electrodeposition technique was used to fabricate multilayered FeCoNi/Cu and CrFeCoNi/Cu nanowires. Commercially available nanoporous templates of anodized aluminum oxide (Whatman 20 nm pore, 60 μm thick) were used as templates for fabricating nanowires. The surface of the membrane was sputtered with gold for 10 min with a current of 20 mA depositing a (10 nm) thin layer of gold for electrical contact acted as the working electrode. The gold coated membranes were fixed in an in-house fabricated Polyetheretherketone (PEEK) stationary holder exposing an area of 2.25 cm^2 of the membrane. A platinum mesh (99.99% pure) was used as the anode (counter electrode) and a saturated calomel electrode was used as the reference