## ORIGINAL PAPER

# Giant magnetoresistance in electrodeposited Ni-Cu/Cu multilayers and anisotropic magnetoresistance in pulse-plated Ni<sub>x</sub>Cu<sub>1-x</sub> alloy films

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Abstract Ni-Cu/Cu multilayers and Ni<sub>x</sub>Cu<sub>1-x</sub> alloy films were electrodeposited on Au-coated glass substrates from a sulfamate-based electrolyte. The roomtemperature magnetoresistance of the multilayers was studied as a function of the ferromagnetic Ni-Cu layer thickness. Giant magnetoresistance was observed, which peaked at about 1% for a Ni-Cu layer thickness around 15 Å. Magnetoresistance measurements of Ni<sub>x</sub>Cu<sub>1-x</sub> films of different composition were obtained at both room temperature and 77 K. Anisotropic magnetoresistance of up to 1.1% and 1.7% was observed at room temperature and 77 K, respectively.

**Keywords** Alloys · Anisotropic magnetoresistance · Electrodeposition · Giant magnetoresistance · Multilayers

#### Introduction

Materials showing giant magnetoresistance (GMR) continue to attract considerable interest because of their numerous technological applications, although the discovery of GMR took place more than a decade ago. The observation of GMR in multilayers was followed by its observation in heterogeneous granular

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W. Schwarzacher H. H. Wills Physics Laboratory, University of Bristol, Bristol, BS8 1TL, UK alloy films and this has simulated considerable interest in these metastable, artificial systems [1, 2]. A variety of techniques have been used to fabricate multilayers and alloy films and the structure and therefore properties depend closely on the preparation method [3].

Electrodeposition, which is a relatively inexpensive technique, is an alternative to vacuum-based methods such as evaporation, sputtering, or molecular beam epitaxy, and may be used to fabricate multilayer and heterogeneous alloy films by appropriate control of the experimental parameters. Electrodeposited multilayers may be prepared from a single electrolyte containing ions of all the metals to be deposited [4, 5], by alternating either the current density or the substrate potential between two values at which the different metals are deposited. Heterogeneous alloy films may be prepared by co-deposition of two immiscible metals [6, 7] or by pulse electrodeposition [8].

#### Experimental

In the present work a single electrolyte was used containing Ni sulfamate (2.3 mol), Cu sulfate (0.05 mol), and boric acid (0.49 mol per litre purified water). The pH and temperature of the electrolyte were adjusted to  $2.0\pm0.1$  and  $30\pm1$  °C, respectively. Ni-Cu/Cu multilayers with 200 periods were electrodeposited on Au-coated glass substrates using a computer-controlled potentiostat. The plating potential was alternately pulsed between the value corresponding to the reduction of Cu (-0.4 V/SCE) and that at which Ni and Cu were simultaneously reduced (-1.9 V/SCE). The thickness of the individual ferromagnetic layers ( $t_{Ni-Cu}$ ) and non-magnetic layers ( $t_{Cu}$ ) was varied by monitoring the current at all stages of growth and adjusting the deposition time for each layer to give the desired total charge, taking the current efficiency into account [3].

The magnetoresistance (MR), defined as  $\Delta R/R = (R_{\rm H} - R_0)/R_0$ where  $R_0$  is the resistance without a magnetic field and  $R_{\rm H}$  that in a magnetic field *H*, was measured with a home-built probe in the current-in-plane/field-in-plane configuration at room temperature at a maximum field of 3500 Oe. Four point-probes arranged in a square made contact with the sample and the MR was measured with the current predominantly parallel ["longitudinal" MR (MR<sub>1</sub>)] and perpendicular ["transverse" MR (MR<sub>1</sub>)] to the applied field [3].

#### **Results and discussion**

In order to investigate how the GMR depends on the individual layer thicknesses, several multilayer series were prepared. The dependence of MR<sub>1</sub> and MR<sub>t</sub> on  $t_{Ni-Cu}$  is shown in Figs. 1 and 2, respectively, for three independent multilayer series of 200 repeats with  $t_{Cu} = 10$ , 15, and 20 Å. Figure 1 shows that some of the films exhibit GMR  $(MR_1 < 0)$ , depending on the thickness of the individual layers. Figure 2 shows that  $MR_t < 0$  for all films, as expected. A maximum GMR of about  $1\% (0.95 \pm 0.1\%)$ was achieved for a multilayer consisting of 200 bilayers of Ni-Cu(13 Å)/Cu(15 Å). This result is comparable with the GMR of  $\sim 2\%$  at much larger magnetic field (20.6 kOe) observed for a multilayer consisting of 1700 bilayers of Ni-Cu(20 Å)/Cu(20 Å) grown galvanostatically on Ti from a citrate-based electrolyte [9]. However, it is significantly less than the  $\sim 6\%$  reported for sputtered Ni/Cu multilayers by Kubota et al. [10]. This fact suggests either that the electrodeposited multilayers have structural defects not present in the sputtered multilayers, which suppress the GMR, or that the incorporation of Cu in the ferromagnetic Ni-Cu layer has an adverse effect. Given that in the present work a significant GMR is recorded even for  $t_{Cu}$  as small as 10 Å, which would not be the case if the layer structure had too many defects, the second may be the more likely explanation.

If the quantity of metal deposited in each cycle of a multilayer is reduced, at some point the layers will become discontinuous, and if it is reduced still further, the deposit will approach a homogenous Ni<sub>x</sub>Cu<sub>1-x</sub> alloy [8]. In the second part of the present work we have studied the magnetotransport properties of Ni<sub>x</sub>Cu<sub>1-x</sub> alloys prepared by the alternate deposition of submonolayer quantities of Cu and Ni-Cu for the composition range x = 0.32-0.81 at 300 K and 77 K.



**Fig. 1** Dependence of the maximum percentage room temperature "longitudinal" MR (MR<sub>1</sub>) on the ferromagnetic Ni-Cu layer thickness ( $t_{\text{Ni-Cu}}$ ) for three independent Ni-Cu/Cu multilayer series, each with a constant  $t_{\text{Cu}}$ . A negative sign for MR<sub>1</sub> indicates a GMR effect



Fig. 2 Dependence of the maximum percentage room temperature "transverse" MR (MR<sub>t</sub>) on the ferromagnetic Ni-rich layer thickness ( $t_{Ni-Cu}$ ) for the three independent Ni-Cu/Cu multilayer series of Fig. 1

Figure 3 shows the variation of the anisotropic magnetoresistance (AMR) [defined as AMR = $(MR_t-MR_1)/2$ ] as a function of the Cu content for a series of pulse-plated  $Ni_xCu_{1-x}$  alloy films prepared as described [8]. AMR of up to 1.1% and 1.7% was observed at room temperature and 77 K, respectively. Figure 3 shows that the AMR decreases with increasing Cu concentration at both temperatures and for a given concentration of Cu it increases with decreasing temperature. Note, however, that we are still measuring significant AMR for values of x much smaller than those at which we would expect a homogeneous Ni-Cu alloy to be non-magnetic ( $x \approx 0.7$  at 300 K,  $x \approx 0.5$  at 77 K). This suggests that the alloys are in fact heterogeneous, with Ni-rich regions responsible for the AMR.



Fig. 3 Anisotropic magnetoresistance as a function of Cu content (at%) for pulse-plated Ni<sub>x</sub>Cu<sub>1-x</sub> alloy films at 300 K and 77 K

This finding agrees with earlier work on the magnetic properties of such alloys [11].

# Conclusions

In summary, we have shown that essentially the same method, alternate deposition of Cu and Ni-Cu layers, can be used to produce either multilayers showing the GMR characteristic of such structures, or  $Ni_xCu_{1-x}$  alloys showing AMR. The persistence of AMR at small *x* suggests that the alloys are heterogeneous.

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