Journal of Magnetism and Magnetic Materials 421 (2017) 194–206

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Magnetic and magnetoresistance studies of nanometric electrodeposited Co films and Co/Cu layered structures: Influence of magnetic layer thickness

S. Zsurzsa*, L. Péter, L.F. Kiss, I. Bakonyi

Wigner Research Centre for Physics, Hungarian Academy of Sciences, Konkoly-Thege út 29-33, Budapest, H-1121 Hungary

ARTICLE INFO

Article history:
Received 5 May 2016
Received in revised form
12 July 2016
Accepted 5 August 2016
Available online 6 August 2016

Keywords:
Magnetic properties
Magnetoresistance
Electrodeposition
Nanoscale magnetic layers

ABSTRACT

The magnetic properties and the magnetoresistance behavior were investigated for electrodeposited nanoscale Co films, Co/Cu/Co sandwiches and Co/Cu multilayers with individual Co layer thicknesses ranging from 1 nm to 20 nm. The measured saturation magnetization values confirmed that the nominal and actual layer thicknesses are in fairly good agreement. All three types of layered structure exhibited anisotropic magnetoresistance for thick magnetic layers whereas the Co/Cu/Co sandwiches and Co/Cu multilayers with thinner magnetic layers exhibited giant magnetoresistance (GMR), the GMR magnitude being the largest for the thinnest Co layers. The decreasing values of the relative remanence and the coercive field when reducing the Co layer thickness down to below about 3 nm indicated the presence of superparamagnetic (SPM) regions in the magnetic layers which could be more firmly evidenced for these samples by a decomposition of the magnetoresistance vs. field curves into a ferromagnetic and an SPM contribution. For thicker magnetic layers, the dependence of the coercivity (H_c) on magnetic layer thickness (d) could be described for each of the layered structure types by the usual equation $H_c = H_{co} + a/d^n$ with an exponent around $n = 1$. The common value of n suggests a similar mechanism for the magnetization reversal by domain wall motion in all three structure types and hints also at the absence of coupling between magnetic layers in the Co/Cu/Co sandwiches and Co/Cu multilayers.

1. Introduction

The application of the giant magnetoresistance (GMR) effect [1,2] in magnetic field sensors has been greatly advanced by the introduction of the exchange-coupled GMR spin-valve concept [3]. The basic structure of a spin-valve is a sequence of a ferromagnetic (FM) layer 1, a non-magnetic (NM) spacer layer, an FM layer 2 and an *anti*ferromagnetic (AF) layer (schematically, FM1/NM/FM2/AF), each sublayer being in intimate contact with the neighboring layer (s) [4]. A typical sequence is for example Ni–Fe/Cu/Co/Ni-Mn. The two FM layers are uncoupled or only weakly coupled through the spacer layer. Therefore, whereas FM layer 2 is strongly pinned by the AF layer and keeps its magnetization orientation fixed as determined by the FM2/AF interface, the orientation of the magnetization of the magnetically soft FM1 layer (also called "free" layer) can be easily changed by a relatively small external magnetic field. In this manner, the magnetizations of the FM1 and FM2 layers can be aligned at practically any inclination angle. Specifically, a parallel alignment corresponds to a low-resistance state whereas in the antiparallel aligned state, due to the GMR effect in the FM1/NM/FM2 structure, the resistance is significantly higher. The resistance difference of the two alignment states can be used for the detection of a magnetic field (e.g., the stray field between differently oriented magnetic regions) and this is the basis for using the GMR spin-valve structure, e.g., in read-out heads of hard-disk drives [5].

The GMR effect originally discovered in FM/NM multilayer structures [1,2] is the highest when the adjacent layer magnetizations are antiparallel aligned [6–8]. In physically deposited multilayer structures, this can be achieved by choosing spacer layer thicknesses ensuring an AF coupling between adjacent layers which occurs at the so-called AF maxima [9–11]. Specifically, for Co/Cu multilayers, at the first AF maximum (at about 1 nm spacer thickness), the AF coupling is very high and strong magnetic fields (as high as 5–10 kOe) [9,11] can only reverse the magnetizations to achieve the parallel alignment (low-resistance state). At the second AF maximum for the same multilayers (typically at 2 nm spacer layer thickness), the GMR is reduced by a factor of two, but since the saturation field is reduced by a factor of 10, there is a

* Corresponding author (Ph.D. student at Eötvös University, Budapest).
E-mail address: zsurzsa.sandor@wigner.mta.hu (S. Zsurzsa).

http://dx.doi.org/10.1016/j.jmmm.2016.08.019
0304-8853/

significant gain in field sensitivity [9,11]. However, the saturation field is still typically 0.5 kOe here whereas even lower saturation fields are required for sensor applications [5].

A possible solution to comply with this requirement was the elaboration of the spin-valve structure [3] which has, indeed, found successfully application in sensors. Another concept was also proposed to reach high GMR at sufficiently low magnetic fields by the so-called pseudo spin-valve [12,13]. Such a structure can be formed by the repetition of a [FM_s/NM/FM_h/NM] quadrilayer [12] to build up a multilayer or can simply consist of a FM_s/NM/FM_h trilayer sandwich [13]. In both pseudo spin-valve versions, the coercivity of the FM_s layer (soft layer) is smaller than the coercivity of the FM_h layer (hard layer) whereas the NM layer thickness is chosen to exhibit a coupling between the FM1 and FM2 layers as small as possible. After saturating both FM layers in one direction and changing the magnetic field direction to the opposite, first the FM_s layer magnetization will reverse whereas the FM_h layer magnetization remains in the original orientation until its coercive field is reached. Thus, in the magnetic field range between the FM_s and FM_h coercivities, the magnetizations of the two kinds of magnetic layers are antiparallel aligned and a significant GMR effect can occur when passing a current through this pseudo spin-valve structure. The different coercivities can be achieved either by appropriately choosing the individual magnetic layer thicknesses or their composition (e.g., Co and Ni–Fe).

In Section 6.3 of Ref. [4], a summary was given on the attempts to produce a spin-valve sandwich structures by electrodeposition [14–19]. According to the basic idea of Attenborough et al. [14–16], in the core of the sandwich, an artificial antiferromagnet (AAF) was designed by preparing a Co(2.7 nm)/Cu(3.2 nm)/Co(2.7 nm)/Cu(3.2)/Co(2.7 nm) layered structure with thin Co layers and thin Cu layers, the latter intended to ensure a strong AF coupling between the Co layers. On both sides of this core structure, a thick Co layer (10 nm) was grown which was separated by a thick Cu layer (4.7 nm) from the core, the latter employed with the purpose of magnetically decoupling the outer thick Co layers from the core structure. The whole structure indeed exhibited a pseudo spin-valve behavior in that a clear plateau could be observed in the *MR (H)* curve (with a maximum GMR of about 5%) and clear steps in the *M(H)* curve [14–16]. However, a critical evaluation [4] of these results led to the conclusion that actually an AAF structure was not formed in the core. This conclusion is mainly supported by the summary of experimental results on electrodeposited Co/Cu multilayers [20,21] which reveals that the GMR magnitude does not exhibit an oscillatory behavior, but rather a monotonous increase with increasing Cu spacer thickness. Therefore, the thin spacer layer thickness (2.3 nm) used by Attenborough et al. [14–16] is not expected to mediate an AF coupling. Shima et al. [17] prepared the same layered structure with almost equivalent layer thicknesses and on an identical substrate. Although distinct MR switching curves were observed for both positive and negative magnetic fields, the maximum magnetoresistance was only slightly above 1%. Therefore, Shima et al. [17] concluded that their observed magnetoresistance may have stemmed from domain wall magnetoresistance. Pasa and coworkers [18,19] have also attempted the preparation of similar electrodeposited sandwich structures as pseudo spin-valves, but their results neither showed convincingly a plateau behavior of the *MR*(*H*) curves.

As noted above, the basic problem is that since there is no evidence for an AF coupling in electrodeposited Co/Cu multilayers, but rather for the absence of such a coupling [20,21], an AAF structure cannot be prepared by this technique. Therefore, for achieving a pseudo spin-valve behavior in electrodeposited layered structures, the realization of uncoupled magnetic layers with different coercivities should be pursued instead.

From an analysis of the evolution of both the GMR magnitude and the coercivity with spacer layer thickness [21], it could be concluded that in electrodeposited Co/Cu multilayers, fully uncoupled magnetic layers can be achieved above a certain Cu layer thickness only since for small spacer thicknesses an FM coupling cannot be excluded. Having uncoupled magnetic layers in a layered magnetic nanostructure, the coercivity of the magnetic layers can be controlled by their thickness since the most typical behavior in magnetic thin films is a monotonous decrease of the coercivity with increasing thickness [22,23].

Therefore, it was the purpose of the present work to study the variation of the coercivity of Co layers with thickness. For this purpose, we have prepared various electrodeposited nanometric layered structures from Co thin films via Co/Cu/Co sandwiches to Co/Cu multilayers. For the latter two structures, the Cu spacer layer thickness was chosen 5 nm which was expected to be sufficient to ensure a decoupling of the adjacent magnetic layers [21]. For comparing the behavior of the three kinds of magnetic nanostructure, magnetic and magnetoresistance measurements were carried out at room temperature for Co layer thicknesses ranging from 1 nm to 20 nm.

2. Experimental

The nanometric electrodeposited Co thin films, Co/Cu/Co sandwiches as well as the Co/Cu multilayers were electrodeposited from a aqueous electrolyte containing 0.74 mol/l $CoSO_4$, 0.010 mol/l $CuSO_4$, 0.3 mol/l Na_2SO_4, 0.25 mol/l H_3BO_3, and 0.15 mol/l H_3NO_3S. The bath composition was very similar to the one used in our previous work on studying the initial growth stages of electrodeposited Co/Cu multilayers [24].

All the samples were deposited on a [100]-oriented, 0.26 mm thick Si wafer covered with a 5 nm Cr and a 20 nm Cu layer by evaporation. The purpose of the chromium layer was to ensure adhesion and the Cu layer was used to provide an appropriate electrical conductivity for the cathode surface. Electrodeposition was carried out in a tubular cell of 8 mm × 20 mm cross section at room temperature with an upward facing substrate placed at the bottom of the cell [4,25]. This arrangement ensures a lateral homogeneity of the deposits and helps to avoid edge effects.

Based on our experience in studying the initial growth stages of electrodeposited Co/Cu multilayers [24], the electrodeposition process was always started with the deposition of a 2.5 nm thick Cu layer on the Si/Cr/Cu substrate. One aim of depositing such an initial Cu layer is to get rid of, at least partially, the influence of the native oxide of the evaporated Cu layer before the deposition of the first Co layer. Since the deposition of the magnetic layered structures of interest was always started with a Co layer, the observed detrimental influence of the native oxide layer on the Co nucleation [24] could be significantly reduced in this manner. The other beneficial effect is the reduction of the Cu content in the first Co layer due to a depletion of the electrolyte at the cathode surface before the Co deposition, hence reducing any possible difference between the first and upcoming Co layers. After completing the deposition of the magnetic layered structure in the form a single Co thin film or a Co/Cu/Co sandwich, a protective Cu layer of 5 nm thickness was immediately electrodeposited on top of it from the same bath. The same 5 nm thick Cu layer was used as spacer between the magnetic layers in both the Co/Cu/Co sandwiches and the Co/Cu multilayers (for the latter, the last 5 nm thick Cu layer served simultaneously as a protective surface layer). The 5 nm thickness of the Cu spacer layer was chosen on the basis of our previous work [21] according to which at this spacer thickness we can already expect a more or less perfect decoupling of the adjacent magnetic layers. In the nanometric structures, the magnetic layer thickness d_{Co} was varied from 1 nm to 20 nm. For the Co/Cu