SIMULTANEOUSLY THERMIonic VACUUM ARC DISCHARGES IN OBTAINING FERROMAGNETIC THIN FILMS*

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Received June 29, 2010

Abstract. High quality granular ferromagnetic thin films on glass and silicon wafer substrates were obtained using the original thermionic vacuum arc (TVA) method developed at National Institute for Laser, Plasma and Radiation Physics. We report obtaining of 77-200 nm thick Cu/Ni/Fe structures. An experimental set-up, with three simultaneously discharges, was made to obtain the desired thin film composition and thickness.

The structural and morphological properties of the prepared nanostructured films were analyzed by AFM (Atomic Force Microscopy), XRD (X-ray Diffraction) and SEM (Scanning Electron Microscopy). Magneto-Optical Kerr Effect studies were made in order to infer the changes in polarization of an optical radiation due to the magnetic field influence. In order to obtain concluding electrical measurements results, cooper electrodes were deposited using the same method before and after the actual three material thin structures. In this way significant changes in the electrical resistance behavior were noticed and were correlated with TVA plasma parameters used for film preparation.

1. INTRODUCTION

The granular ferromagnetic thin films are composite materials that consist usually in nanometric magnetic grains (Fe, Co) uniformly distributed within a nonmagnetic matrices (cooper, silver or gold). These films are able to produce both positive and negative [1] electrical resistance variation under the influence of an external magnetic field [1-5]. The electrical conductance between magnetic granules is determined by the density of the quantum levels in the both parts of the

* Paper presented at the Annual Scientific Session of Faculty of Physics, University of Bucharest, June 8, 2010, Bucharest-Magurele, Romania.
electrical barrier. When majority spin electrons from a granule tunnel the barrier and find their corresponding levels in the other granule empty the electrical resistance is low, while in the case these levels are occupied the resistance is high [6]. Because of its magnetic properties, these structures are called soft ferromagnetic materials having important applications in the microelectronic devices like magnetic sensors, reading hard drives heads, microwave devices, magnetic shielding and transformers.

One of the most proper base materials used for this kind of sensors is the granular NiCuFe layer which combines the AMR (anisotropic magnetoresistance), low signal with high electric conductivity and low coercivity. It was used cooper to suppress the magnetoresistive signal below of the NiFe [7–8]. The optimal concentration of the thin film structures included 30%-65% Ni, 5%-25% Fe and 10%-40% Cu. For this thin films Ni atoms can make solid connection with the Cu atoms, generating possible granular alloys of Cu-Ni and Fe-Ni, respectively [9].

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For this study was used Thermionic Vacuum Arc (TVA) discharge which ignites in vacuum conditions in the vapors of the anode material, continuously generated by the electron bombardment of the anode [10–13]. The electrons, emitted from a heated tungsten cathode, are accelerated towards the anode, by a D.C. high voltage applied across the electrodes. Because the deposition of the thin films using TVA technology is obtained in high vacuum conditions and also under the energetic ions bombardment of the depositing thin films, we obtained a high quality compact nanostructures having high purity and increased adhesion.

2. EXPERIMENTAL SET-UP AND METHOD

The materials used for combined deposition were Fe and Ni as magnetic materials and Cu as the nonmagnetic matrix. The TVA deposition method is described elsewhere [14] and is based on an intense anode evaporation by electrons emitted from a heated cathode filament and accelerated towards the anode. The innovation of this work consists in the third TVA gun which made possible all three simultaneously discharges of Fe, Ni and Cu, respectively.

The experimental arrangement is shown in Fig. 1 and consists of three cathodes made from tungsten wires, each of them mounted in three different focusing Wehnelt cylinders configurations. The electrons emitted by the cathodes are accelerated toward the high voltage anodes. Each anode consists in a graphite crucible filled with a Ni, Cu and Fe chunks, respectively. The position of each heated gun can be controlled using an exterior rotator device. One geometrical parameter is the distance \( d \) between the electrodes.
The external parameters of the discharge were the current value that heats the cathodes and the respective anode voltage. The relative concentration control between all of the three components of the thin film were made by choosing the proper electric parameters for discharges and by proper positioning of the substrates in respect with the three plasma sources. Due to the different distances of the substrates to the three anodes the concentration of the magnetic metal in the Cu will increase or decrease constantly with $x$ position [14].

The studied discharges were in nickel, cooper and iron vapors. In order to achieve a correlation of the discharge parameters was used an electronic signal conditioning and data acquisition for current and anodic voltage. The software to program the devices was LabView.

The ignited thermionic vacuum arcs were running stable and smooth. The currents used to heat the cathode filaments were between 45–55 A for each electron gun. The discharge current was 100 mA for Ni, 40 mA for Cu and 70 mA for Fe and the applied high anode voltage was 2 kV for Ni, 1.5 kV for Cu and 1 kV for Fe. The deposition rate and film thickness were measured using a FTM7 quartz microbalance. The thickness of the obtained structures was of about 200 nm. The pressure in the deposition chamber was $1 \times 10^{-5}$ Torr.

3. RESULTS AND DISCUSSION

In order to obtain the desired ferromagnetic thin film structures using the TVA method, an optimization of the discharge parameters was made. The TVA
plasma discharge is characterized in having a high energy, equivalent with a high deposition rate. Because this work consisted in three simultaneously discharges, the deposition rate was controlled in such a way of obtaining the proper concentration of the structure. The energy supplied for the thermo emitted electrons from the heated cathode had to be diminished to values for which the deposition rate was of few angstroms/s, controlled by the variation of the discharge voltage and the heating cathode current. The optimization consisted in getting the deposition to start at the point of ignition, but no further more, obtaining in this way the total control for the deposition rate. In the figures below (Fig. 2 a, b and Fig. 3 a, b) it can be seen the optimization made for the cooper discharge. The distance between the electrodes and the heating filament current are two important parameters in the TVA discharge. As it can be seen in Fig. 2 maintaining the same inter-electrodes distances for low heating filament currents implies a higher energy amount introduced into the system. The explanation of this is that the local vapor pressure above the anode is lower for lower heating filament current so that is needed a larger energy amount to be introduced into the system. A high values of the heating filament current is equivalent with a larger number of electrons accelerated towards the anode, likewise for the same heating filament current and different distance between the electrodes (Fig. 3 a, b). Because the concentration of the obtained structure had to be a precise one, the optimization went to the point of controlling the ignition moment (Figs. 2, 3b).

![Graph](image)

Fig. 2 – $I-V$ characteristics for same inter-electrodes distance, different heating filament current, before (a) and after (b) optimization.
In this way, the coating was made mainly by thermal evaporation, the deposition rate and the film thickness being fully controlled. The ignition point for all of the material used in this work, Ni, Cu and Fe was studied. Having the control of the ignition point for each material made possible on one hand of controlling the deposition rate and on the other hand of the relative elemental concentration of the desired structure.

The coating was made on glass and silicon substrates placed at a distance of 250 mm of the anode-cathode systems. The structure is presented in Fig. 4. Ni, Cu and Fe evaporators were running all in the same time.

Fig. 3 – I-V characteristics for same filament current, different inter-electrodes distance before (a) and after (b) optimization.

Fig. 4 – Simultaneously coating structure and electrodes view.
In order to obtain significant results of the electrical measurements, two cooper electrodes were coated on the substrates before, and after the deposition. These electrodes were coated using the same thermionic arc method, but using special masks. The thickness of these two electrodes was of about 500 nm each.

The topographical AFM images and the subsequent statistical data analysis, including the calculation of the root mean square roughness ($R_q$), gave detailed information about the surface morphology. In Fig. 5 a, b it is shown the morphological AFM images of the samples $P_1$ and $P_2$, where sample $P_1$ was placed over the evaporating point of the nickel, and $P_2$ was placed over the evaporating point of the iron. The distance between these two substrates was 150 mm. As mentioned in a previous work [14] the concentration of each sample varies in respect with the distance between the anode-cathode discharges and the substrates position. As sample $P_1$ was over the Ni evaporator, it is expected a higher concentration of Ni in respect with the sample $P_2$ which has a lower concentration of Ni but a higher concentration of Fe.

As the concentration in the cooper matrix of Ni is decreasing and the concentration of Fe increasing, it can be seen from the morphological structure AFM view that the $R_q$ roughness of the sample $P_1$ has values of 20 nm decreasing to values of 10 nm for the samples $P_2$ with a lower concentration of nickel. The in-plane size of the surface roughness was found to be almost the same.
XRD analyses were performed to establish the presence of crystalline phases in the ferromagnetic structure.

In the XRD patterns of the samples with a higher concentration of Ni (P₁) to those with a higher concentration of Fe (P₂) the 2θ diffraction peaks were assigned to (111) and (200) planes of Cu, as seen in Fig. 6. The Ni and Fe lines were inserted from the database JCPDS – International Centre for Diffraction Data and allowed us to assign the presence of Ni and Fe in the Cu matrix. As shown in Fig. 6, copper diffraction lines are present for all range of the concentration obtained. The intensity of each line is higher for the samples placed in the middle of the nickel and iron anode-cathode systems, where the copper discharge took place. This intensity has lower values as the concentration of copper matrix decreases in a proportional way (Fig. 6). Because of the substrates position, a large variation of concentrations was obtained. Each concentration was studied, in the idea of getting different electromagnetic effects for different concentrations.

Magneto Optic Kerr Effect (MOKE) were performed for three of the samples, starting with sample P₁ with a higher concentration of Ni (Fig. 7 a), P_intermediate with a medium concentration of Ni and Fe (Fig. 7 b) and P₂ sample with a higher concentration of Fe (Fig. 7 c).
Fig. 7 – MOKE measurements for different samples with different relative elemental concentrations.

Fig. 7 a, b, c shows the MOKE signal on the analyzed samples which has a descending tendency from a higher concentration of nickel to a higher concentration of iron. The saturation magnetization for sample P2 has the greatest value; sample P\textsubscript{Intermediate} has a middle value. The lowest value of the saturation magnetization was found for the P1 sample.

4 points electrical measurements were performed on the samples to avoid the possible errors that could appear at the contacts between the substrates and the thin films. Two of the contacts were used to insert a constant direct current and the other two were used for the reading of the dropping voltage. The electrical 4 points measurements scheme is shown in Fig. 8. The contacts were made using silver paste on the two cooper electrodes coated under and above the ferromagnetic structure.
Current-voltage characteristics were performed on the samples. First, the samples were measured in the absence of the magnetic field. The DC current established in the ferromagnetic thin film was increased from a lower value of 100 µA to 1 mA. It was observed an increase of the dropping voltage and the electrical resistance of the sample.

The higher values of the electrical resistances were found for the samples with a higher concentration of nickel. The variation of the electrical resistance in respect with the position of the samples and with the concentration is shown in Fig. 10.
Electromagnetic measurements were also made on the ferromagnetic samples. The induced DC current through the structure was kept to a constant value of 1 mA the only parameter which varied being the magnetic field. The dropping voltage was read for each value of the magnetic field. For the samples with a higher concentration of Ni it was obtained a negative magneto resistance (MR) effect of –3%. The MR effect was calculated using the formula:

$$\frac{\Delta R}{R} = \frac{R - R(0)}{R(0)}, \quad (1)$$

where $R(0)$ is the value of the electrical resistance when the applied magnetic field is 0 T. For the high nickel concentration samples the MR negative effect obtained was of –4.5%. (Fig. 11a). As the value of the Ni concentration is decreasing the MR effect is increasing having a value of –1.8% as shown in Fig. 11b. Due to the different concentrations of each sample a different MR effect was obtained. As it can be seen in Fig. 11c the obtained MR effect was positive, with a value of 4.8%. For the samples with a lower concentration of nickel and a higher concentration of iron, the MR effect had a maximum value of 5% (Fig.11d). The magnetic field was varied from a positive value of 0.3 T to a negative value of –0.3 T. The MR effect was obtained for both of the magnetic field values, positive and negative. The temperature at which was measured the effect was of 295 K, the room temperature.

When the electric current passes through the samples, the electronic spins of the current electrons meet in their way the electronic spins of the magnetic domains of the structure. The spins of the magnetic domains have a chaotic orientation. When a magnetic field is applied, the magnetic domain electron spins are oriented on the direction of the field. If the electronic spins of the electric current have a parallel orientation with the electronic spins of the magnetic domains, the resistance of the sample has a low value equivalent with a low electric resistance. On the other hand, if the electronic spins of the inductive current have an anti

Fig.10 – Electrical resistance behavior in respect with the x position.
parallel orientation in respect with the electronic spins of the electronic magnetic domains the electric resistance has a greater value. Depending on the densities energy levels, by applying a magnetic field, it can be obtained a positive or a negative MR effect. As it had been seen in Figs. 11 it was obtained a positive and negative effect in respect with the concentration of each thin ferromagnetic structure.

Fig. 11 – MR negative effect obtained on high Ni concentration samples (a, b).

Fig. 11 – MR positive effect obtained on low Ni concentration samples (c, d).

4. CONCLUSIONS

Thermionic Vacuum Arc (TVA) method proved its efficiency in obtaining thin films with a ferromagnetic structure. Because of the optimization made on the parameters involved on the TVA discharge, it had been achieved the control of the
deposition rate and the thickness of the prepared films, two of the most important parameters of the electromagnetic sensible structures. On the same deposition process were obtained thin films with concentration which varied in respect with the position between the evaporators and the substrates. Films having relative concentrations of Ni (starting from 13wt% to 6.5wt%) and Fe (8wt% to 15wt%) in a Cu matrix were prepared. It had been achieved different MR effects on different samples with different concentrations. For samples having Ni concentration, of 12.5wt% and low Fe concentration (8.8wt%) the MR effect had a negative value of –3.5%. By decreasing Ni concentration to values of 8.81wt% and high Fe concentration (13wt%) the MR effect had a positive value of 4.9%.

REFERENCES