

## The Influence of Residual Gas on the Magnetic and Magnetoresistive Properties of Co/Cu Films

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**Abstract:** The article deals with the influence of residual gas on the magnetic and magnetoresistive properties of polycrystalline Co/Cu/Co films after annealing at a temperature  $T_{an}=240^{\circ}\text{C}$ . It is found that samples damaged by residual gases show low proportion antiferromagnetically ordered regions and a high remanence. Magnetoresistive effect on the samples obtained are virtually absent. Thermal annealing reveals sharp peaks at the antiferromagnetic layer thickness of copper equal  $d_{Cu}=1.2\text{nm}$  and  $2.2\text{nm}$  and  $3\text{nm}$ , which are consistent with literature data. The reasons for the increase in the magneto resistance effect are discussed. There is a correlation between the proportion of ant ferromagnetic related field and saturation of the samples.

**Key words:** Magnetron spray • Giant magnetoresistance • Antiferromagnetic coupling • Three-layer films  
• The temperature annealing

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### INTRODUCTION

Multilayer magnetic structures with alternating ultrathin magnetic and non-magnetic layers are of great interest among many researchers, due the ability to produce films with different electrical, magnetic, superconducting and mechanical properties, such as giant magneto resistance, perpendicular anisotropy, the exchange interaction, etc.

Intensive study of magnetoresistive properties of multilayer structures is connected with the discovery of giant magneto resistance in them. The size of giant magnetoresistive effect strongly depends on various parameters: deposition technique, the substrate material, the thickness of the magnetic and non-magnetic layers, the thickness of the buffer layer, etc.

It is known that the giant magneto resistance and the interlayer interaction Co/Cu multilayer sensitive to structural defects. In particular, for the successful preparation of good quality structures high-vacuum conditions are necessary. The most destructive gases affecting the ant ferromagnetic interaction and magneto resistance effect in films are  $\text{H}_2\text{O}$  and  $\text{O}_2$  [1, 2].

In the work [3], it is shown that the samples had a significant ant ferromagnetic coupling and high GMR, while, examples by damaged gas, show a decrease magneto resistance, significant permanence. The changes GMR are due to changes of bonds between ferromagnetic ones.

In the papers [4-8] giant magnetoresistance multilayer films associated with the formation of antiferromagnetically ordered regions in the ferromagnetic layers.

This paper concedes the effect of residual gases on the magneto resistive effect in deposited and annealed Co/Cu films.

**The Experimental Procedure:** Samples were prepared at vacuum apparatus QUADRA 500TM, by method of quadruple magnetron spraying under constant current in the atmosphere of working gas Ar at  $P_{Ar} = 5 \cdot 10^{-3}$  Torr in vacuum system with a basic pressure of  $\sim 8 \cdot 10^{-6}$  Torr. At this pressure,  $\sim 90\%$  of residual gas mainly  $-\text{N}_2$  and residues  $\text{H}_2\text{O}$  with  $\text{O}_2$ . [3]. The thickness of Co layers  $d_{Co}=12\text{nm}$  and the thickness of the copper layer  $d_{Cu}$  changes from 0 to 4 nm.

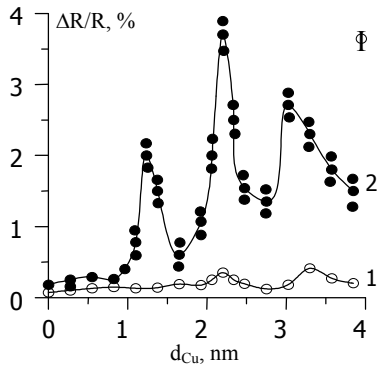


Fig. 1: Dependence of  $(\Delta R/R)_{\max}$  on the thickness of the copper layer for the deposited samples (curve 1) and after annealing at temperature  $T_{\text{an}} = 240^\circ\text{C}$  (curve 2).

The magnetoresistance was measured by a compensation method at the two-point scheme. Films were annealed in vacuum of  $1 \cdot 10^{-5}$  Torr in a magnetic field of 16kA/m applied in the plane of the film in the direction of the axis of easy magnetization. The annealing temperature varied between 20-400°C. The annealing time at each temperature was 30 minutes.

The crystal structure of the films was investigated by electron microscopy, phase composition was investigated-by electron micro-diffraction; magnetic structure was investigated-by Lorentz microscopy. Effective magnetization  $I_{\text{ef}}$  was measured by induction method at automated vibrating sample magnetometer in a field  $H=80$  kA/m and the coercive force  $H_c$  were measured by the induction and magneto-optical methods. The magnetoresistance was measured by a compensation method at the two-point scheme [9, 10].

**The Main Part:** Films Co / Cu / Co deposited on a glass substrate at a temperature  $T=20^\circ\text{C}$  were investigated. Fig. 1 shows the dependence  $(\Delta R/R)_{\max}$  on the layer thickness after the deposition of Cu films (curve 1) and after annealing at temperature  $T_{\text{an}}=240^\circ\text{C}$  (curve 2). It is seen that after deposition ant ferromagnetic maximums are completely absent in the second and third films but the first ant ferromagnetic maximum is not found.

Fig. 2 shows loops of magnetic (a) and magneto resistive (b) hysteresis of films damaged by the residual gases with a thickness of the nonmagnetic layer  $d_{\text{Cu}}=2.2\text{nm}$ . It can be seen that the residual magnetization is high; the squareness of the hysteresis loop is in this case equals  $I_r/I_s=0.99$ . The share of ant ferromagnetic ordered fields AF in these films is equal to 1%. Magneto resistive effect, which is proportional to the fraction of ant ferromagnetic ordered fields, is 0.25 % (Fig. 2b).

After annealing the films at temperature  $T_{\text{an}}=240^\circ\text{C}$  on the curve  $(\Delta R/R)_{\max}=f(d_{\text{Cu}})$  (Fig. 1 curve 2) three ant ferromagnetic maximums are clearly distinguished. There may be several reasons for this behavior  $(\Delta R/R)_{\max}$ : increase of the angle of dispersion of the magnetization vector, due to an increase in grain size, separation of Cu and Co and the formation of a sharper interphase boundary, the removal of internal tension caused by the diffusion of impurity atoms (atoms of the residual gas) to sinks (intergrain boundaries, the surface of the film).

Giant magnetoresistance  $(\Delta R/R)$  occurs when there are fields with ant parallel orientation of magnetization vector in the adjacent ferromagnetic layers. The value of the GMR effect depends on the share of areas related in ant ferromagnetic way and the strength of the ant ferromagnetic bond [11]. To determine the proportion of ant ferromagnetically ordered fields of Co layers and indirect exchange bond loops of the magnetic and the magneto resistive hysteresis have been constructed.

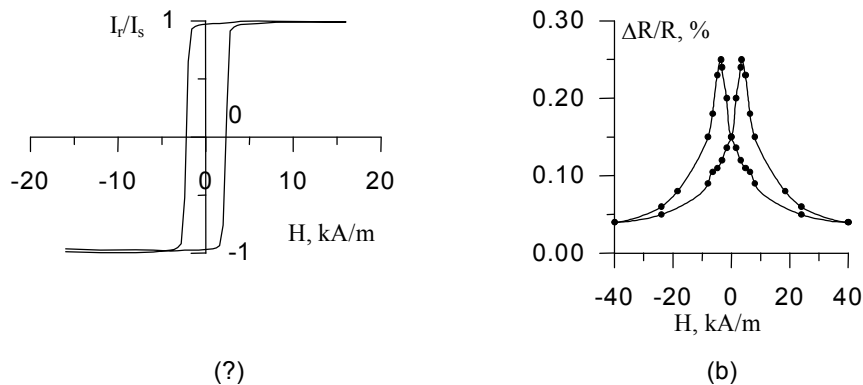


Fig. 2: Magnetic (a) and magneto resistive (b) of the hysteresis loop of the films, deposited on glasses and damaged by the residual gases at a thickness of the nonmagnetic layer  $d_{\text{Cu}}=2.2\text{nm}$ .

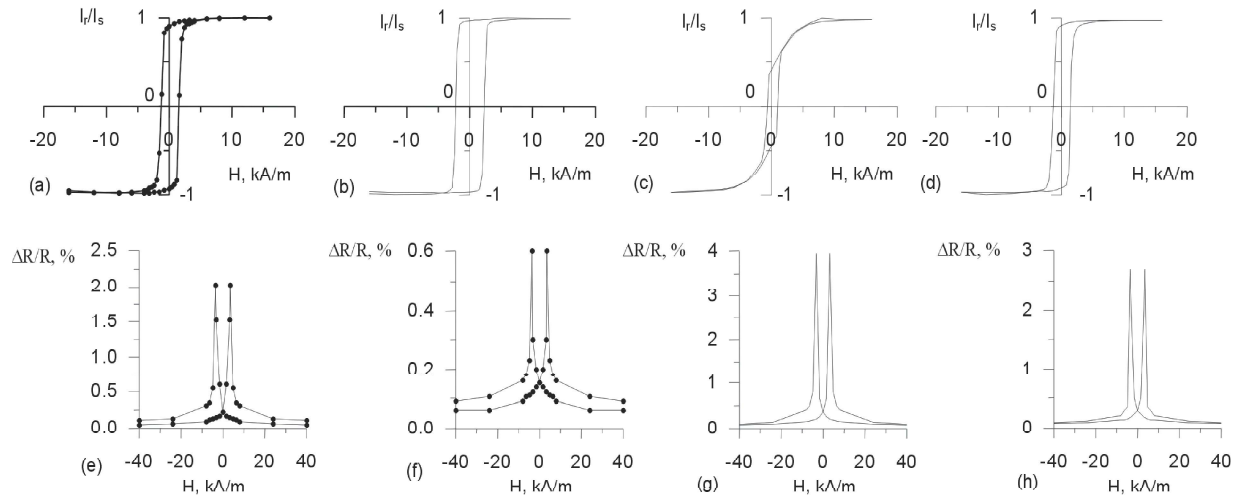


Fig. 3: Magnetic (a, b, c, d) and magnetoresistive (e, f, g, h) hysteresis loops of the films Co/Cu/Co, deposited on a cover glass and annealed at  $T_{an}=240^{\circ}\text{C}$ ,  $d_{Co}=12\text{nm}$ : a, e- $d_{Cu}=1.3\text{nm}$ , b, f- $d_{Cu}=1.6\text{nm}$ , c, g- $d_{Cu}=2.2\text{nm}$ , d, h- $d_{Cu}=3.1\text{nm}$ .

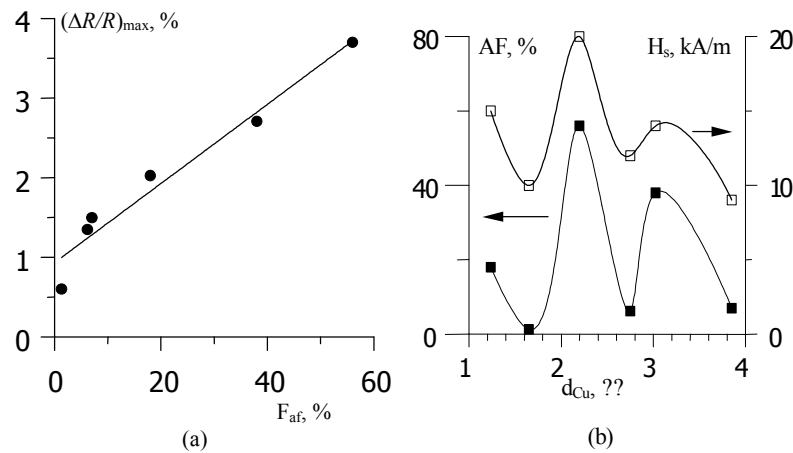


Fig. 4: The dependence of the magnetoresistance  $(\Delta R/R)_{max}$  on the share of ant ferromagnetically ordered fields of the ferromagnetic layers  $F_{af}$  of films Co/Cu/Co annealed at  $T_{an}=240^{\circ}\text{C}$  (a), the dependence of the share of ant ferromagnetically ordered fields (curve |) and saturation field (curve ?) for samples annealed at a temperature  $T_{an}=240^{\circ}\text{C}$ , the thickness of the layer of copper  $d_{Cu}$  (b).

Figure 3 shows a magnetic loop (a, b, c, d) and MR (e, f, g, h) Hysteresis loops of Co/Cu/Co, deposited on glasses coverslips and annealed at  $T_{an}=240^{\circ}\text{C}$  with thickness of copper layer  $d_{Cu}=1.2\text{nm}$  (a, e) (the first antiferromagnetic maximum),  $d_{Cu}=1.6\text{nm}$  (b, f) (antiferromagnetic minimum),  $d_{Cu}=2.2\text{nm}$  (c, g) (the second ant ferromagnetic maximum);  $d_{Cu}=3.1\text{nm}$  (g, h) (the third ant ferromagnetic maximum).  $d_{Co}=12\text{nm}$ .

The type of magnetic hysteresis loops allows to speak about the presence of both ferromagnetic and ant ferromagnetic related fields in the bulk of the film and their volume ratio varies with the thickness of the copper layer and change of the morphology of interfaces.

According to the magnetic hysteresis loop share of ant ferromagnetically related fields was estimated Co:  $F_{af} = \frac{I_{af}}{I_{af} + I_f} = 1 - \frac{I_r}{I_s}$  [4, 5], where  $I_r$  and  $I_s$  are the residual magnetization and saturation magnetization, respectively. Fig. 4a shows the dependence of  $(\Delta R/R)_{max}=f(F_{af})$  films of Co/Cu/Co annealed at  $T_{an}=240^{\circ}\text{C}$ . The observed linear dependence  $(\Delta R/R)_{max}=f(F_{af})$  in a wide range of thickness of the copper layer ( $d_{Cu}=1.3-3\text{nm}$ ) for the films deposited on glass substrates, allows to make a conclusion about an unambiguous connection of magneto resistance and share of ant ferromagnetically ordered fields in adjacent ferromagnetic layers.

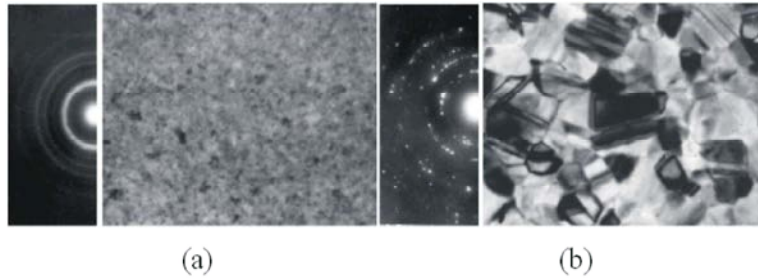


Fig. 5: Images of structures and patterns micro diffraction of films Co/Cu/Co before annealing (a) and after annealing (b).

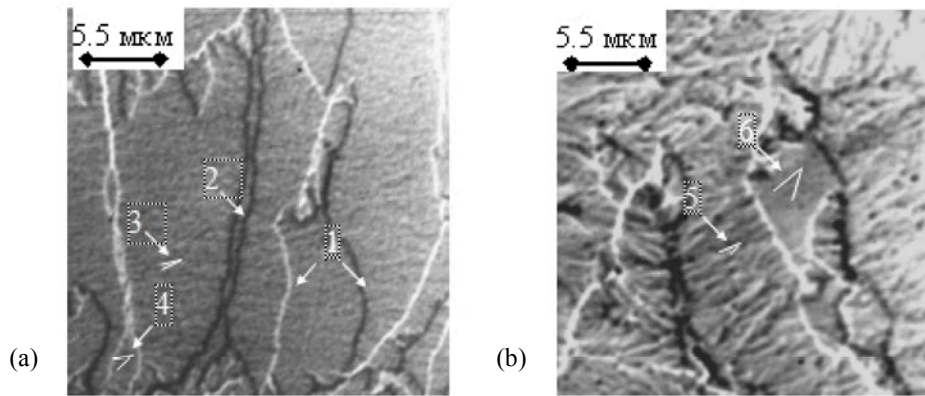


Fig. 6: Electron microscopic image of the blast furnace structure of the films Co/2.2nm Cu/Co: a-after spraying, b-annealing  $T_{an}=240^{\circ}\text{C}$ .

The saturation field  $H_s$  was determined due to magnetoresistive hysteresis loops, which is proportional to the indirect exchange bond between the ferromagnetic layers [12]. Since the " tails " of the curve of the magnetoresistive hysteresis extend to the large fields, the field  $H_s$  was determined by the intersection with the horizontal axis of the tangent drawn through the point on the curve  $\Delta R/R=f(H)$ , located at half maximum of the peak  $(\Delta R/R)_{max}$ . Fig. 4b shows the dependence of saturation field  $H_s$  and the proportions of ant ferromagnetically ordered fields ( $F_{af}$ ) of films Co/Cu/Co, deposited on a glass substrate, on the thickness of the copper layer  $d_{Cu}$ . It can be seen that the value  $H_s$  reaches the maximum meaning for the films Co/Cu/Co with a thickness of the intermediate layer  $d_{Cu} = 2.2$  and 3.1nm according to the second and third ant ferromagnetic maximums. Thus, there is a correlation of curves  $\Delta R/R=f(d_{Cu})$  (Fig. 1),  $H_s=f(d_{Cu})$  (Fig. 4b curve ?) and  $F_{af}=f(d_{Cu})$  (Fig. 4b curve |), which indicates that the strength of ant ferromagnetic bond determines the size of the giant magnetoresistance effect.

Electron-microscopic images of the films Co/Cu/Co showed that all films are grained polycrystalline with the grain size  $L=5-6\text{nm}$  (Fig. 5a). In such films the dispersion of the magnetization vector is so small that on the images

of the blast furnace structure by the Lorentz microscopy method magnetization ripple is absent.

Researches of blast furnace structure showed that in the films Co/Cu/Co with a thickness of copper layer ( $d_{Cu}=2.2\text{nm}$ ) after deposition double of Neel organized (Fig. 6a). In the deposited film those areas prevail in which the Neel boundaries are placed one above the other (this corresponds to the case when the adjacent layers Co are related ferromagnetically) ( Fig. 6a, arrow 1 ) and there are very few areas where the Neel boundaries diverge (Fig. 6a, arrow 2). Between blast furnace walls of the same contrast there are areas in which in adjacent layers Co ant ferromagnetic bond is organized.

Dispersion angle of the magnetization vector in areas related ferromagnetically was  $11^{\circ}$  (Fig. 6a, arrow 3) and in areas related ant ferromagnetically was  $34^{\circ}$  (Fig. 6a, arrow 4). After annealing at  $T_{an}=240^{\circ}\text{C}$  the size of grain increased to  $R=11-15\text{nm}$  (Fig.5b). The contrast of the magnetization vector of the ripples increased (Fig. 6b). It can be seen that dispersion angle of the magnetization vector increased. Dispersion angle of the magnetization vector in the areas where the layers Co are related ferromagnetically was  $17^{\circ}$  and in areas where there was ant ferromagnetic bond between layers Co was  $45^{\circ}$  (Fig. 6b, arrows 5 and 6).

It should be also be noted that the dispersion angle of the magnetization vector in the fields related ant ferromagnetically is  $\varphi=0,45$  rad, what is significantly greater than dispersion of the magnetization vector in the fields related ferromagnetically-where  $\varphi=0,09 \div 0,17$  rad. The conduction electrons are scattered by magnetic defects (such as magnetization ripple). In fields related ant ferromagnetically (large dispersion of magnetization vector) conduction electrons must dissipate more intense than in fields related ferromagnetically. Therefore, increasing the ant ferromagnetically related fields in adjacent layers of cobalt leads to an increase in the magneto resistance effect.

Obviously, in the deposited films in Co/Cu/Co, studied in the present work, the concentration of point defects (vacancies, the residual gas atoms, etc.) exceeds thermodynamically equilibrium one. In this case, there is a thermodynamic driving force that tends to reduce the concentration of defects to the equilibrium value. Excess defects during annealing are removed from the grain by diffusion them to the drains (grain boundaries and the surface of the film) and the recombination of vacancies and interstitial atoms. At the same time the voltage of the film partially is relieved. It can be assumed that in this case the intermediate layer Cu becomes more perfect; as a result fields in the adjacent layers Co related ant ferromagnetically appeared in the three-layer film.

On the other hand, annealing Co/Cu/Co films leads to delimitation of Co and Cu atoms [13], thereby forming a sharp interfacial boundary and the alignment of the intermediate layer Cu on thickness, which promotes the formation of the ant ferromagnetic bonds in the adjacent layers and Co accompanied by an increase of magnetoresistive effect as  $\frac{\Delta R}{R} \approx H_s = \frac{4J_{ef}}{\mu_0 I_s d_{Co}}$  [14], where

the  $H_s$ -is the saturation field,  $J_{ef}$ -is the effective energy of ant ferromagnetic bond. The share of ant ferromagnetically related fields can be determined by the magnetic hysteresis loops, as well as by electron microscopic images of the blast furnace structure.

### CONCLUSION

The study of the behavior of the magnetic parameters and the value of magneto resistance of three-layer films of Co/Cu/Co at thermal magnetic treatment has been carried out. It is shown that the change in magnetic parameters and the magneto resistance is due to the change of crystal structure. Films with a thickness of copper layer corresponding to the second ant ferromagnetic maximum are of particular interest.

At the thermal magnetic treatment not only the magnitude of the field  $H_s$  and  $\Delta R/R$  changes, but also the appearance of the magneto resistive hysteresis loops changes.

Correlation of dependencies of the magneto resistance effect and the saturation field on the annealing temperature gives us the right to say that the saturation field is proportional to the power of the interlayer ant ferromagnetic bond, determines the magnitude of the giant magneto resistance effect.

The oscillation period  $H_s$  and  $F_{af}$  coincides with the period of oscillation  $\Delta R/R$  at the change of thickness  $d_{Cu}$ .

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