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Structural and magnetic characterization of Co/Au(100) epitaxial multilayers

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Abstract :

Many studies have been devoted to the Co/Au system. Only the hcp Co(0001)//Au(111) epitaxial relationship has been investigated so far. We present a new bct Co(100)//Au(100) multilayered structure. Structural and preliminary magnetic measurements are discussed.

Introduction :

Over the last years, ferromagnetic metallic layers have been widely used as components of thin films, sandwiches and multilayer structures. Many new magnetic properties have emerged from these materials¹ : large perpendicular anisotropy, giant magnetoresistance effect, oscillation in the coupling between ferromagnetic layers through non-magnetic spacers.

In the scope of these features, Au/Co multilayers have already attracted much interest²⁻⁵. Deposition of the layers has been carried out by sputtering techniques²⁻³ as well as Molecular Beam Epitaxy⁴⁻⁵ (MBE). In these reports, the structure of the deposits is always the cfc (111) Au // hcp (0001) Co epitaxial relationship. It has been found⁶ that the easy magnetization axis is perpendicular to the surface when the Co thickness does not exceed 1.1 nm. The magnetoresistance ratio is then larger than that obtained with thicker films, when the magnetization lies in the plane. Also, a large Hall effect in zero applied magnetic field has been reported⁷⁻⁸.

The present paper reports preliminary results obtained with a new cfc (100) Au // bct (100) Co epitaxial relationship. Our purpose in investigating this system was to determine how another structure of the Co and Au layers would affect the magnetic results summarized above.

Elaboration and structural characterization :

The MBE equipment has been described elsewhere⁹. Reflection High Energy Electron Diffraction (RHEED) is performed during the growth. The source is operated at 40 kV and the angle of incidence is set around 1-2°.

The MgO wafers are obtained from Sumitomo. They are solvent-cleaned and transferred in ultra-high vacuum (UHV) where they are heated. It has been shown that the growth of Au directly on MgO(100) processes by islanding¹⁰. Therefore, we followed a technique which had

already been used with success to grow Au(100)/Ni(100)⁹ as well as Ag(100)/Fe(100)¹¹ multilayers. A thin Fe 1-2 nm-thick layer is first deposited at room temperature (RT) on the MgO surface. Because there is a $\sqrt{2}$ -ratio between the lattice parameters of the bcc iron structure ($a_{\text{Fe}} = .286 \text{ nm}$) and that of the fcc structure of MgO (NaCl structure, $a_{\text{MgO}} = .421 \text{ nm}$), an in-plane lattice agreement may be found with the following relationship :

$$\text{Fe}(100) [110] // \text{MgO}(100) [100].$$

Figure 1a shows the RHEED pattern obtained with a 2 nm-thick iron layer. The beam is along the [001] direction. The Au buffer was deposited at room temperature with a growth rate of about 0.1 nm/s. We found that higher substrate temperatures led to disordered layers showing a very diffuse RHEED pattern. When about 5 nm have been deposited at RT, the surface produces a well-defined (1X5) reconstruction¹², which becomes of higher quality with increasing thickness. The Au orientation is the same as that of MgO, their lattice parameters being close ($a_{\text{Au}} = .408 \text{ nm}$).

Fig. 1a

Fig. 1b

Figure 1b shows the (1X5) RHEED pattern of a 400 nm-thick Au buffer layer, in the [011] azimuth.

Co was deposited at RT with a growth rate of 0.04 nm/s. RHEED streaks remain very well defined during the deposition of the first Co monolayers, and oscillations can be recorded, displaying a 2D growth till about 6 layers. These oscillations are used to calibrate precisely the growth rates in terms of ML deposition. However, the diffuse background is enhanced when the Co thickness increases. After a 3 nm-thick Co layer has been grown, the RHEED pattern shows faint spots and Au deposition produces polycrystalline rings.

Two different superlattices have been grown, with Co thicknesses of respectively 2 and 4 ML. The thickness of the Au interlayers is 15 ML in both cases. 61 bi-layers have been deposited for the first specimen and 41 for the other. We will refer to them as (Au15Co2) and (Au15Co4) in the following.

Magnetic properties :

The magnetic properties of these two samples have been investigated both by SQUID magnetometry (at 280K and 5K) and ferromagnetic resonance (at room temperature).

Figure 2 shows the in-plane and out-of-plane hysteresis loops of the sample (Au15Co2) at 280K. Clearly, the magnetization is harder to saturate when the field is applied perpendicular to the film plane indicating an in-plane anisotropy. This is in contrast to the case of thin Co layers deposited on Au(111) for which a perpendicular anisotropy is observed for Co thickness up to 11Å.

Fig. 2

The in-plane hysteresis loops of the sample with thin Co layers at room temperature and 5K are plotted in figure 3. At 280K the remanent magnetization and coercive field are very weak ($M_R \cong 0.2 M_{sat}$; $H_c < 10$ Oe).

At 5K, the saturation magnetization is almost the same than at room temperature but the coercive field is larger ($H_c \cong 60$ Oe). These results suggest that these very thin Co layers have a superparamagnetic behaviour. Fitting the magnetization curve at room temperature leads to an average magnetic moment of the superparamagnetic clusters of $2100 \mu_B$, meaning a size of approximately 1200 atoms. As the temperature is decreased, the magnetic moment of these clusters freezes leading to a higher remanent magnetization and coercivity.

Fig. 3

Figure 4 shows a comparison at room temperature of the in-plane hysteresis loops of the two samples (Co 2ML versus Co 4ML). The sample with larger Co thickness exhibits a lower saturation field and a larger coercivity. However its remanent magnetization is still low. These results can be interpreted in terms of the superposition of two contributions to the magnetization: a surface contribution which is still superparamagnetic as in the sample with thin Co layers plus a volume contribution (from the inner Co layers) in which the magnetization would have a more established ferromagnetic behaviour. This point of view seems to be supported by the following FMR results.

Fig. 4

The FMR measurements have been carried out at room temperature between 0 and 18kOe using the X band (9.4 GHz). The spectra have been recorded for different orientations

of the applied field with respect to the plane of the layers (ϕ_H angle between the applied field and the film plane). These spectra represent the derivative with respect to the applied field of the absorbed microwave power.

Fig. 5

Fig. 6

Figures 5 and 6 represent typical FMR spectra obtained on the multilayers with respectively 2 and 4 Co monolayers. In the range 0-18 KOe, the spectra on the first sample exhibit a single line at very low field (a few hundred Oe). Its position depends very weakly on the field orientation with respect to the film plane (see figure 7). For the second sample two lines are observed. The first one exhibits the same behaviour as the low-field line of the first sample. These low-field lines may be caused by a non-resonant absorption of the microwave power by the unsaturated magnetization of the samples.

The second line occurs at larger fields. Its dependence on the field orientation (ϕ_H) is usual for thin ferromagnetic films with in-plane anisotropy (see fig.8). The presence of this second line for 4 monolayers of Co supports the idea that a ferromagnetic core forms in the volume of the Co layers giving rise to the coercivity observed on the hysteresis loop and to this usual ferromagnetic FMR line. We have fitted this angular dependence with a classical model¹³ which takes into account the Zeeman energy, the shape anisotropy and a magnetocrystalline anisotropy $E_a = K_1 \sin^2 \alpha + K_2 \sin^4 \alpha$ (α = angle between the magnetization and normal to the film plane). The fit leads to the following set of parameters : $4\pi M_s H_a = 6035$ Oe and $H_{a2} = 81$ Oe ($H_a = H_{a1} + H_{a2} = 2K_1/M_s + 4K_2/M_s$). Assuming for the magnetization of Co the bulk value = 1440 emu/cm³ we deduce : $K_1 = 8.6 \cdot 10^6$ erg/cm³ and $K_2 = 2.9 \cdot 10^4$ erg/cm³. This positive value of K_1 indicates a perpendicular magnetocrystalline anisotropy approximately four times smaller than the magnetostatic shape anisotropy. K_1 is two times larger and K_2 fifty times smaller than in bulk Co ($K_1 = 4.5 \cdot 10^6$ erg/cm³ along the c-axis, $K_2 = 1.5 \cdot 10^6$ erg/cm³). These differences must be ascribed to the difference in the crystallographic structures between these samples (bcc Co) and bulk Co (hcp Co) or to interfacial roughness¹⁴.

Further experiments are in progress to clarify these magnetic properties in particular by looking at samples with various Au thicknesses. In particular the existence of an antiferromagnetic coupling between Co layers through the Au spacer may contribute to the low remanent magnetization seen on fig.4.

Fig. 7 : Angular variation of the resonant field which characterizes the low field lines in the two multilayers.

Fig.8 :Angular variation of the resonant field which characterizes the high field line in the Au15Co4 multilayer. The full line is a fit according to the model of ref.14.

In conclusion, single crystal Co(100)//Au(100) multilayers have been grown on MgO(100) substrates. Unlike the Co(1000)//Au(111) structure, no perpendicular anisotropy is observed. The 2ML Co multilayer seems to exhibit a superparamagnetic behavior and the 4ML Co multilayer shows two magnetic contributions: superparamagnetic and ferromagnetic.

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