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LETTER TO THE EDITOR

Microscopic evidence for pseudo-epitaxy in FCC (100) Co/Cu multilayers

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Abstract. We have grown FCC (100) Co/Cu multilayers on (100) silicon substrates, using a diode-RF sputtering technique. The in-plane texture of the films was studied *in situ* using reflection high-energy electron diffraction (RHEED). The microscopic layout of the (100) cobalt planes was investigated by means of nuclear magnetic resonance (NMR). Plots of the original NMR spectra against the direction of the applied static field are presented as microscopic evidence of a pseudo-epitaxial stack. The RHEED patterns obtained along the [011] and [001] azimuth of the copper planes support this interpretation.

In a previous paper [1], we reported on magnetic properties of FCC (100) Co/Cu multilayers deposited on silicon (100). Cubic in-plane anisotropy was observed and attributed to a strong in-plane texture of the cobalt planes. The epitaxial growth of (100) Cu on (100) Si has been shown possible using electron-beam evaporation [2], diode-RF [3] or magnetron [4] sputtering, assuming a 45° rotation of the (100) copper planes. In this case, the lattice mismatch between Cu and Si is reduced to 5.9%. On the other hand, due to small lattice mismatch (2%), epitaxy between cobalt and copper is expected. Accordingly, we found from hysteresis loops measured by SQUID magnetometry, that [001] and [011] Co axes were respectively the hard and easy directions for the magnetization. In this letter, we give direct experimental evidence for single-crystallinity in our Co/Cu multilayers. NMR and RHEED measurements are used to correlate microscopic and macroscopic aspects of this structural state.

The samples were prepared at LEP using a diode-RF sputtering system. The sample analysed in detail in this study has a (20.9 Å Cu/12.6 Å Co) × 30 structure and was deposited on a 300 Å Cu buffer layer epitaxially grown on a (100) silicon substrate [3]. This buffer layer, deposited in special conditions is used to initiate the epitaxy of the Co/Cu multilayer. The period thickness was determined using grazing x-ray reflection at the copper K-α line with an accuracy better than 1%. However, the respective thicknesses of cobalt and copper, deduced from the relative deposition times are given to within 1 Å. Detailed x-ray studies have shown a FCC (100) growth with crystallite size along the growth direction as large as four periods [3].

The RHEED measurements were performed under ultrahigh vacuum just after the deposition of the multilayers. Figure 1 presents the RHEED patterns obtained along two azimuths at 45°. Sharp streaks are observed and give evidence for an epitaxial growth of the multilayer. The distance between the streaks is inversely proportional to the distance between the atomic rows parallel to the electron beam. Then, among the possible azimuths of the (100) cobalt planes, we can attribute the patterns (*a*) to [011] and (*b*) to [001]. According to the cubic symmetry of the (100) cobalt planes, we observe similar RHEED patterns by rotating the sample around its surface normal. The sharpness of the streaks is characteristic of a high degree of texture and of a smooth surface. Comparison with the RHEED patterns obtained just after the deposition of the Cu buffer layer [3], shows clearly that the structural quality of the surface is improved during the growth of the multilayer.

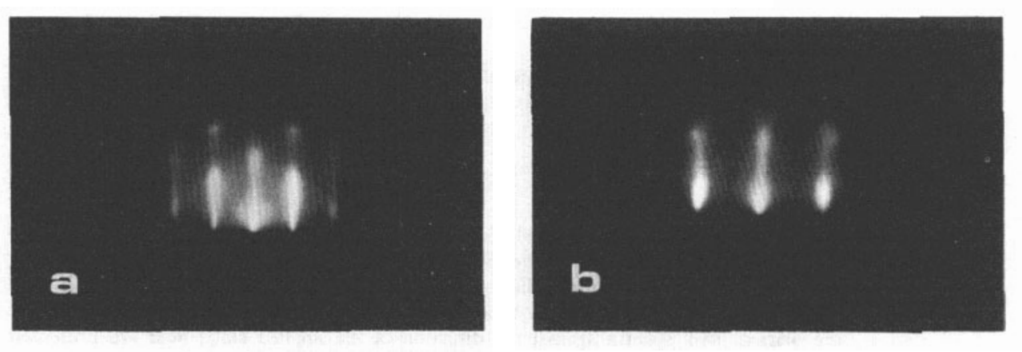

Figure 1. RHEED patterns of the (100) Co/Cu multilayers: (*a*) along the [011] azimuth and (*b*) along the [001] azimuth of the (100) cobalt planes. The sharp streaks give evidence for a smooth and epitaxial growth.

The NMR measurements were carried out at 2 K using a variable frequency spin-echo apparatus. We cut two pieces 9 mm × 20 mm along two direction at 45° corresponding to the easy and hard axis of the cobalt planes. They were placed in a Dewar tail around which was fitted an exciting coil, so that the RF magnetic field was parallel to the film and lengthwise to the sample. A DC magnetic field was applied in the plane of the film and perpendicular to the RF field.

We observed in our sputtered Co/Cu multilayered film a Co satellite line, around 197 MHz, due to Co sites which have one Cu nearest neighbour (figure 2). This is similar to the spectra observed in a dilute Co–Cu alloy [5]. For 12 Co atoms surrounding 1 Cu atom the cubic symmetry is broken and each Co–Cu direction constitutes a local symmetry axis. We call such a Co crystal site S_1. The hyperfine field H_{n1} of S_1 is just characterized by the values when the magnetic moment is along and perpendicular to the local axis. For a slightly anisotropic field, H_{n1} can be written as

$$H_{n1} = H_{iso} + H_{an}(3\cos^2 q - 1)/2$$

where H_{iso} and H_{an} are the isotropic and anisotropic parts of the hyperfine field respectively, and q is the angle between the magnetization M_s and the local symmetry axis. For ^{59}Co nuclei the connection between the resonance frequency F and the magnetic field H is given by the ratio $F/H = 1.0054$ MHz kOe^{-1}.

It is now straightforward to determine the number of magnetically inequivalent S_1 sites as a function of M_s orientation. Thus for M_s parallel to the [001] crystallographic direction there are two inequivalent sites in the proportion 1:2. For a [011] orientation there are three inequivalent sites in the proportion 1:1:4.

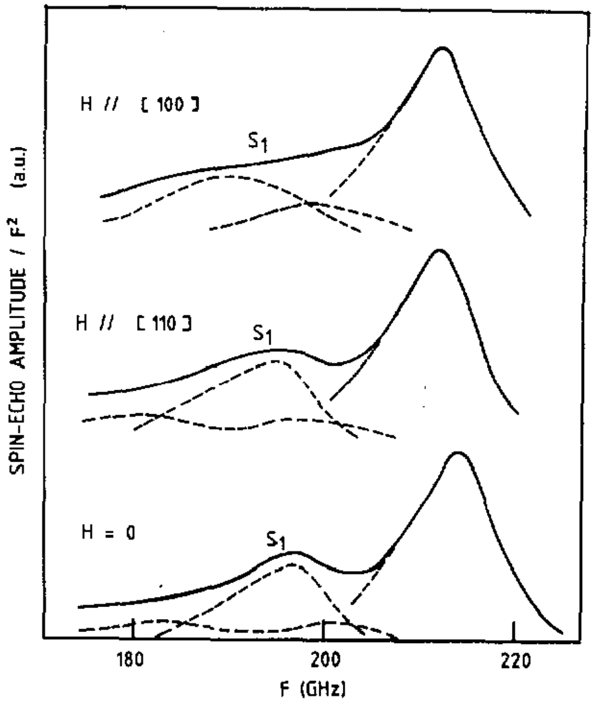

Figure 2. Co spin-echo spectra at 2 K in Co (100)/Cu multilayered films. A 2 kOe DC field H is applied parallel either to the easy [011] axis or to the hard [001] axis in the film plane. Different magnetically inequivalent sites of Co atoms having one Cu nearest neighbour are represented by the broken curves.

The Co spin-echo spectra were measured, at different orientations, on the same sample with a 2 kOe applied field, which is enough to saturate the magnetization in any direction in the film plane [1]. When the DC field H is applied along the easy axis, the whole spectrum shifts to lower frequencies at a rate close to -1 MHz kOe^{-1}. This shift without deformation indicates that the distribution of magnetically inequivalent sites S_1 remains the same as in zero external field. When H is parallel to the [001] hard axis only a -2 MHz shift of the main line is observed. The main line arises from Co sites without Cu nearest neighbours so that the hyperfine field is expected to be isotropic. As a consequence the width of the main line remains practically unchanged. In order to explain the orientation-dependence of the satellite lines we were led to choose $H_{iso} = -194$ kOe and $H_{an} = +12$ kOe within an uncertainty of 2 kOe. The resulting satellite lines are represented by the broken curves on figure 2. For H parallel to the [011] axis each individual line is simply shifted by -2 MHz. The residual intensity below 180 MHz arises from Co sites having more than one Cu nearest neighbour [5]. For H parallel to the [001] axis the amplitude of the satellite lines is slightly reduced and broadened. This line broadening reflects a dispersion in q angle which is ascribed to a slight deviation of M_s from the [001] direction. Indeed, even at 99.4% of the saturation, the angle between H and M_s can be as high as 6°.

The dipole field at S_1 is calculated to be -2 kOe along the axis. The lowering of the symmetry is expected to modify the 3d shell of an S_1 atom, giving rise to an anisotropic hyperfine field through the orbital and spin-dipolar terms. For HCP Co the observed H_{an} value of $+5.3$ kOe originates from the orbital and spin-dipolar contributions evaluated to be +3.6 and +1.3 kOe respectively [6], with negligible lattice dipole field. The present H_{an} value suggests that the distortion of the 3d shell may be important.

In conclusion we have shown that the sputtered Co/Cu multilayered films deposited on a (100) Si substrate possess (100) texture with well-defined axis in the film plane. This was checked using NMR experiments to investigate local symmetry involving Co sites having one Cu nearest neighbour resulting from about 5% Cu diffusion in Co layers. The breaking of cubic symmetry for these sites leads to a H_{an} value far above that of HCP cobalt.

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