Microstructure of columnar crystallites in Ni$_{80}$Fe$_{20}$/Cu magnetic multilayers

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(Received 17 September 1999; accepted for publication 14 December 1999)

We have used electron microscopy to investigate the microstructure of Ni$_{80}$Fe$_{20}$/Cu magnetic multilayers which were synthesized by dc magnetron sputtering. Columnar structure was found in the specimen with and without giant magnetoresistance (GMR). All the columnar crystallites (CCs) originate from the Fe buffer layer on silicon wafer or glass substrate and penetrate through all the multilayers up to the surface of the film. The lateral size of the CCs ranges from 10 to 30 nm. Cross-sectional high-resolution electron microscopy study shows that the CCs are single-crystal-like with fcc structure resulting from the epitaxial growth of NiFe and Cu sublayers. Electron diffraction contrast imaging and electron energy filtered elemental mapping confirmed that multilayer nature is maintained throughout the entire NiFe/Cu film. Grain boundaries between CCs can be the most likely place where NiFe or Cu bridging will occur. Columnar structure was also found in a Ta/NiFe/Cu/NiFe/FeMn/Ta spin valve film. The possible influence of the columnar crystalline structure on the GMR related problems is discussed. The microstructure results revealed in this article provide useful information for the GMR property investigation of NiFe/Cu based metallic multilayers. © 2000 American Institute of Physics. [S0021-8979(00)07406-5]

I. INTRODUCTION

Magnetic multilayers (MLs) with giant magnetoresistance (GMR) effect have been intensively investigated owing to their promising applications in miniaturized sensors and high-density digital magnetic recording devices. NiFe/Cu based MLs with high MR ratio and field sensitivity are regarded as good candidates for magnetoelectronic device applications. Upon the successful synthesis of NiFe/Cu based MLs and spin valves by ion-beam, magnetron sputtering, and electron-beam evaporation methods, extensive structure and property studies of the MLs have been performed. Microstructure of MLs plays an important role in oscillatory exchange coupling between magnetic layers and in spin-dependent electron scattering. It has been reported that the GMR effect is strongly dependent on the microstructure of the NiFe/Cu films. But for NiFe/Cu MLs, detailed microstructure characterization, especially the direct observation of cross-sectional structure at atomic level, are far more behind the physical property study. For example, it is proposed that pinholes through the Cu spacer layers in NiFe/Cu MLs may responsible for the ferromagnetic coupling of neighboring magnetic layers. But there is no microstructure investigation to evaluate the possible occurrence of such pinholes in NiFe/Cu MLs.

In present work we have used high-resolution transmission electron microscopy (HRTEM) to study the microstructure of NiFe/Cu MLs with different substrate and/or varied spacer thickness. Columnar crystallites (CCs) were found to be a prominent structure characteristic in these films. The CCs start from the Fe buffer and penetrate all the sublayers upward to the surface of the film. Similar CCs were also found in NiFe/Cu based spin valves. If some of the CCs are single crystal phase of NiFe, they would provide local ferromagnetic bridges in the multilayer film and result in a possible decrease of GMR. Our experiment results show that the multilayer structure remained within all the CCs. Grain boundaries between CCs can be the most likely place where NiFe or Cu bridging will occur. With the understanding of the microstructure of the CCs, we discussed the possible influence of the columnar crystalline structure on the GMR related problems.

II. EXPERIMENT

Four types of NiFe/Cu MLs of N$_1$, N$_2$, N$_3$, and N$_4$ were prepared by dc magnetron sputtering. The nominal structures of these multilayer films are Si(001)/SiO$_2$/Fe(10 nm)/[Ni$_{80}$Fe$_{20}$(15 Å)/Cu(20 Å)]$_{20}$; glass/Fe(10 nm)/Ni$_{80}$Fe$_{20}$(15 Å)/Cu(20 Å)]$_{20}$; and glass/Fe(10 nm)/[Ni$_{80}$Fe$_{20}$(13 Å)/Cu(10 Å)]$_{25}$, which are denoted as N$_1$, N$_2$, and N$_3$, respectively. Film N$_4$ has the same substrate as N$_1$ but with slightly thinner Ni$_{80}$Fe$_{20}$ (13 Å) and thicker Cu (23 Å) sublayers. The GMR measurements of the films with similar structure can be found in another article. Cross-section specimens for TEM observation were prepared by using a standard method. High-resolution electron microscopy was carried out in a JEM-2010 TEM with point resolution of 1.9 Å, and the energy filtered parallel electron energy loss spectroscopy (PEELS) imaging of the MLs was

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The CCs ranges from 10 to 30 nm. When the Cu spacer is and thus form vertical column boundaries. The lateral size of the growth on the buffer layer up to the surface of the film almost unchanged lateral width from the very beginning of the structure. The CCs extend throughout the entire film with the prominent columnar crystallite feature of these MLs is the structure inside some columnar crystallites could be identified, as indicated by arrows. The diffraction contrast is complex in the images. performed in a CM200-FEG TEM equipped with GATAN imaging filter (GIF) with spatial resolution of 1 nm.

III. RESULTS AND DISCUSSION

Figure 1 shows a series of cross-sectional TEM images of Ni₈₀Fe₂₀/Cu multilayers of N₁ (a), N₂ (b), and N₃ (c). Note the pronounced columnar structure. Multilayered structure inside some columnar crystallites could be identified, as indicated by arrows. The diffraction contrast is complex in the images.

Figure 2 is a HRTEM image of a CC in Fig. 1(b). Inside the CC are many twins with the twinning (111) plan tilted about 26° from the horizontal direction. This kind of twin structure is the most popular structural defect found in CCs. The two sets of equally spaced lattice planes with an angular separation of 70.5° indicate a typical two-dimensional image of a fcc structure along the [110] axis. The multilayer structure of NiFe and Cu could hardly be identified from chemical contrast in Fig. 3 due to their close atomic numbers and therefore small difference in electron scattering ability, especially under HRTEM imaging conditions when the specimen is very thin. It has been reported that the “silver bridge” could be directly identified from HRTEM images of NiFe/Ag MLs because the lattice mismatch between NiFe and Ag is about 13% and large density of dislocations could be found at their interface. In contrast, careful examination of the digitized HRTEM images of CCs shown in Fig. 3 did not show any change in lattice spacing along the growth direction of the film, which reveals a twofold possibility. One is that the CC is possibly a single crystal of NiFe or Cu, which may result in the formation of ferromagnetic “bridges” or nonferromagnetic pinholes, respectively. Another possibility is alternate and coherent growth of NiFe and Cu layers in the CCs because NiFe and Cu are all of fcc structure with small lattice mismatch (~2%), therefore they could grow coherently over each other when each sublayer is only 1–2 nm in thickness. So, a more direct experiment measurement is needed to investigate the chemical information of the CCs.

Elemental mapping by energy-filtered parallel electron energy loss spectroscopy (PEELS) in field-emission-gun transmission electron microscope can provide chemical information at spatial resolution of 1 nm, which can be used as a promising method to solve the composition-related prob-
lems in magnetic MLs as well as in spin valves. Figure 4 is a PEELS spectrum in which electron energy loss peaks corresponding to inner-shell ionization of Fe–L$_{2,3}$, Ni–L$_{2,3}$, and Cu–L$_{2,3}$ are shown respectively. Figures 5(b)–5(d) is a series of elemental maps of Fe, Ni, and Cu by using the three-window method on above PEELS spectrum from Ni$_{80}$Fe$_{20}$/Cu MLs. Compared with the electron-diffraction contrast image shown in Fig. 5(a), where the multilayer structure within CCs could hardly be observed due to the complex diffraction contrast, elemental mapping of Ni, Fe, and Cu could clearly show the multilayer structure of CCs. This shows one advantage of elemental mapping in chemical analysis on MLs over HRTEM. It should also be noted that the Fe concentration is only 20% in each of the NiFe layers, therefore the signal intensity is not very high in Fe maps of NiFe layers. Nevertheless, the Fe buffer shows a strong intensity in Fig. 5(b). Owing to the atomic ratio of Ni to Fe of four in the NiFe layer, the layered structure of the film appears more pronounced in Fig. 5(c) of Ni maps. The Cu layer could also be clearly seen in Fig. 5(d). Because the best spatial resolution of our PEELS system is 1 nm, we could not get further detailed local chemical information at the NiFe/Cu interfaces or at CCs grain boundaries for such ultrathin metallic MLs.

Interface roughness is a key parameter related to GMR, and the roughness can be described in terms of chemical roughness or geometrical roughness. Chemically, NiFe and Cu are likely to intermix at their interface. Annealing will increase the thickness of the intermixing layer and hence decrease the GMR, because the intermixed NiFe/Cu interface is a nonferromagnetic region with high resistivity and electron scattering in this region is spin independent. The thickness of the intermixed NiFe/Cu layer is measured to be around 0.2 nm in the as-deposited spin-valve sample using dc magnetron sputtering. Such a thin layer could not be resolved by elemental mapping with spatial resolution of 1 nm. And it is also impossible to identify such a layer from HRTEM images as shown in Figs. 3 and 6. It is thought that such a thin intermixing layer will not damage the coherent-growth characteristic of the CCs.

Geometrical roughness could be viewed on a larger scale. From Figs. 1, 2, and 5, it is clear that NiFe and Cu
layers run across all the CCs, and the layers are flat in some CCs but curved or tilted in some others. The NiFe and Cu sublayers are generally flat at the early stage of growth and may gradually become wavy during the period when the film grows to be thick, as can be seen in Fig. 2. So surface roughness does not necessarily mean that the under-surface interfaces are rough. From the profile of the sublayers shown in Fig. 1, the growth surface of the film on silicon oxide is more flat than that grown on glass. The waviness of the sublayers is a general feature in many ML systems, which may lead to magnetostatic coupling ("orange-peel" coupling) between the ferromagnetic (FM) layers across the spacer layer. At column boundaries, the sublayers are more likely distorted, which can be seen from the grain boundary regions labeled by short arrows in Fig. 2.

The continuity of the layer structure of the NiFe/Cu MLs is of special interest because discontinuous MLs will greatly change the GMR properties. The break of NiFe and/or Cu layers is most likely to happen at the CC boundaries. Figure 6 is a HRTEM image showing a defective CC grain boundary region with lattice distortion, defects, and strains. Region a shows the gradual lattice transition from one CC to another by a slight tilting of the lattice plane and lattice mismatch at the boundary. Region b indicates that a twin in one CC can extend into another. The grain boundary at the left-hand side of the CC shown in Fig. 6 is a more disordered structure. It is believed that Cu diffuses along grain boundaries in Ni$_{80}$Fe$_{20}$/Cu MLs by annealing and then isolating NiFe layers. The break of the NiFe layers is in favor of increasing the GMR. On the contrary, NiFe pinholes will lead to FM coupling and result in annihilation of GMR. To our knowledge, there is no direct observation of such "copper bridge" or NiFe pinholes in the literature. Though we could not find any direct evidence of Cu-bridge or NiFe pinholes from the above HRTEM images of the as deposited NiFe/Cu MLs, CC boundary can be the most likely place where bridging will occur.

Columnar morphology can also be found in many other ML systems. For example, in Co/Au multilayers, the CCs are mosaics composed by many small domains with very small orientational difference. While in NiFe/Ag multilayers the CC is semicoherent and contains many dislocations due to large lattice mismatch between magnetic and nonmagnetic layers, in the present case, the columnar crystallite is single crystalline with coherent growth of NiFe and Cu layers. Modak et al. found that the MR increases with increasing grain size in Co/Cu MLs. Therefore, enhancement of MR in Co/Cu MLs was attributed to an increase of the electron mean free path. This may also hold for the present case in NiFe/Cu magnetic MLs. Larger CC grain size will decrease the density of CC boundaries in the film and thus could reduce the chance for pinholes to occur.

A spin valve (SV) is a more promising structure that could be developed for real application as GMR read heads. CC could also be found in a NiFe/Cu based spin valve of Ta/NiFe/Cu/NiFe/FeMn/Ta, as shown in Fig. 7. The CCs show coherent growth of fcc structure, in which (111) lattice planes are generally parallel to the film plane, but different CCs will be slightly rotated with respect to the growth direction and then show different lattice images. Electron energy loss spectroscopy was also used to investigate the multilayer characteristics of the SV. Figure 8 shows a bright field image of a cross-section view of a SV and corresponding elemental mapping of Ni, Fe, and Mn, which could be used to identify.

FIG. 6. HRTEM image of a columnar crystallite and its column boundaries in Ni$_{80}$Fe$_{20}$/Cu multilayers shown in Fig. 1(a).

FIG. 7. Cross-sectional HRTEM image of a columnar crystallite in a Ta/NiFe/Cu/NiFe/FeMn/Ta spin valve film.
IV. CONCLUSION

In conclusion, there is prominent columnar structure in Ni$_{80}$Fe$_{20}$/Cu magnetic multilayers and Ta/NiFe/Cu/NiFe/FeMn/Ta spin valves prepared by magnetron sputtering. The multilayered structure within the columnar crystallites was confirmed by elemental mapping, though it cannot be distinguished by HRTEM investigation. Both NiFe and Cu sublayers grow alternately in a columnar crystallite with a coherent fcc lattice. (111)-twin structure was found to be a dominant defect in the columnar crystallites in the MLs. Orientational difference between columnar crystallites results in disordered column boundaries which are roughly parallel to the growth direction of the film. The CC boundary can be the most likely place where Cu bridge or NiFe pinholes will occur. Impact of the columnar crystallites on GMR properties was discussed.

ACKNOWLEDGMENTS

This work was supported by Chinese Academic of Sciences (Grant No. KJ951-A1-401).