

Enhancement of spin-dependent scattering and improvement of microstructure in spin valves by delayed deposition

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Si/Ta/Co/Cu/CoFeMn (or NiO) spin valves were prepared by a delayed sputtering procedure, in which depositions of Ta and followed layers were intervened by 1 h. The spin valves are found to have better coherent growth with stronger (111) preferred orientations in Co and Cu layers, compared with the traditional continuous method. At the same time, the giant magnetoresistance ratio and spin-dependent scattering are enhanced by a factor of 20%–30%. These results indicate that the coherent growth and the (111) preferred orientation can enhance the spin-dependent scattering of spin valves. © 2005 American Institute of Physics. [DOI: 10.1063/1.1949729]

Since the discovery of giant magnetoresistance (GMR) in magnetic multilayers and spin valves, extensive experimental and theoretical investigations have been carried out.^{1–7} For specific magnetic multilayers, the strength and the sign of interlayer coupling between neighboring ferromagnet (FM) layers strongly depend on the microstructure. Antiparallel alignment cannot be acquired at low magnetic fields and the angle between neighboring FM magnetizations varies with microstructure.^{4,6–8} Thus, the variation of the GMR ratio with microstructure might originate from the angle instead from spin-dependent scattering. In order to clarify the effect of microstructure on the spin-dependent scattering, spin valves must be used.

Many attempts have been made to enhance the GMR ratio of spin valves.^{9,10} In order to have a high GMR ratio and high exchange field, a two-step sputtering procedure was employed.¹¹ In this letter, a delayed deposition method was adopted to fabricate spin valves, in which the buffer layer was first deposited and the followed layers were prepared after an interval of 1 h. For comparison, spin valves were prepared by traditional continuous procedure without any delay.

Spin valves of Si (001)/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/NiO (25 nm) and glass/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/FeMn (9 nm) were prepared by a magnetron sputtering system with two different methods. As the traditional continuous method, fabrication of constituent layers was carried out continuously. As the delayed deposition method, fabrication procedure was intervened by 1 h between the buffer Ta layer and the followed layers. The base pressure was 2×10^{-5} Pa and the argon

pressure 0.3 Pa during deposition. A magnetic field of about 133 Oe was exerted to induce a uniaxial anisotropy. The metallic and NiO layers were sputtered by dc and rf sputtering, respectively. The deposition rates of metallic and NiO layers were 0.1–0.2 and 0.04 nm/s, respectively. X-ray diffraction (XRD) and high-resolution transmission electron microscopy (HRTEM) were carried out. The morphologies of Co and Cu surfaces at Co/Cu and Co/Cu interfaces were characterized using atomic force microscopy (AFM). The GMR ratio was measured using standard dc four-point probe method with the current and magnetic field perpendicular to each other and both parallel to the film plane.

Figure 1 shows the room-temperature resistance vs the external magnetic field for the NiO-pinning [Fig. 1(a)] and FeMn-pinning [Fig. 1(b)] spin valves prepared by the two methods. The GMR ratios in the NiO-pinning spin valves are 6% and 7.6% by the continuous method and the delayed deposition one, respectively. The GMR ratios in the FeMn-pinning spin valves are 4% and 5% for the two fabrication methods, respectively. Apparently, the GMR ratio can be enhanced by a factor of 20% to 30% using the delayed deposition method. In experiments, the GMR enhancement was also found for other FeMn-pinning and NiO-pinning spin valves with various Cu spacer layer thickness deposited on glass or Si. The GMR ratio and its enhancement for the NiO-pinning spin valve are larger than those of the FeMn-pinning one.

From the results in Fig. 1, one can find that there is a flat platform for high resistance as a signature of antiparallel alignment. Since the magnetizations of the FM layers are switched from the parallel to the antiparallel alignment with external magnetic field, the GMR enhancement is not caused by a change in the angle between the FM magnetizations. More remarkably, the resistances at saturation fields are equal to each other for the two methods. For example, for the

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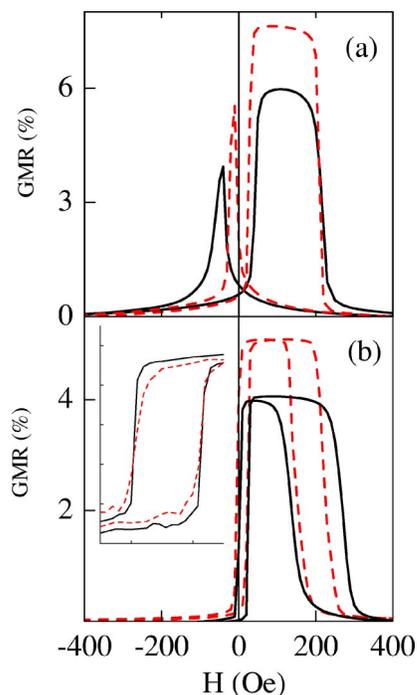


FIG. 1. (Color online) R vs H curves at room temperature for (a) Si/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/NiO (25 nm) and (b) glass/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/FeMn (9 nm)/Ta (5 nm). The inset shows the minor loop of the free Co layers in the FeMn-based spin valves. The solid and dashed lines refer to the results for the continuous and the delayed sputtering methods, respectively.

NiO-pinning spin valves, the sheet resistance changes from 21.36 to 20.16 Ω for the continuous method, and from 21.78 to 20.27 Ω for the delayed deposition method when the external magnetic field is swept. The resistances at the parallel alignment are close to each other for the two methods and the change of the resistance for the delayed deposition method is larger than that of the continuous method. Therefore, the enhancement of the GMR ratio stems from a change of spin-dependent scattering.

In order to understand the mechanism of the GMR enhancement, it is necessary to get information about the difference of the microstructure between the two fabrication methods. As one of the major factors, the interfacial roughness at Co/Cu and Cu/Co in the spin valves has great influence on the GMR ratio.^{5,12} For Si/Ta (7 nm)/Co (3 nm) and Si/Ta (7 nm)/Co (3 nm)/Cu (3 nm), prepared by the delayed deposition and the continuous methods, the roughnesses at Co and Cu surfaces were detected, respectively. AFM measurements show that for these samples the rms of roughnesses of the Cu surface are 0.145 and 0.157 nm for the delayed deposition and the continuous methods, respectively. The rms of surface roughnesses of the bottom Co surface with the delayed deposition and the continuous methods are 0.166 and 0.110 nm, respectively. Apparently, the interfacial roughness at Co and Cu surfaces for the delayed deposition method is close to those of the continuous method. This coincides with the phenomenon that the resistance at saturation fields is equal to each other for the two fabrication methods. Although the interfacial roughness may increase or suppress the GMR effect,¹² the interfacial roughness cannot be used to explain the GMR enhancement in the present work.

Figure 2 shows the XRD spectra of NiO-pinning and FeMn-pinning spin valves. For the NiO-pinning spin valve, a

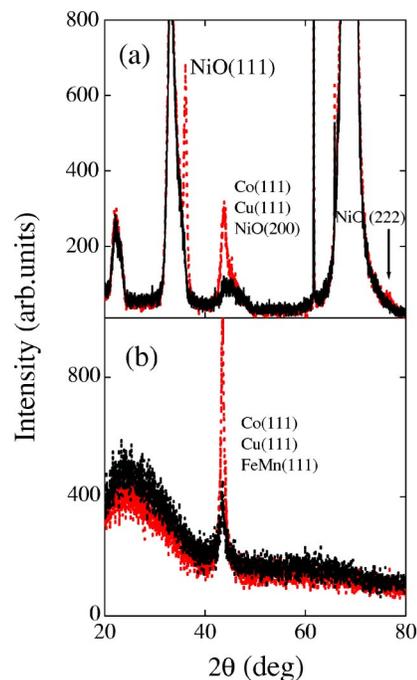


FIG. 2. (Color online) X-ray reflection spectra at high angles for (a) Si/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/NiO (25 nm) and (b) glass/Ta (7 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/FeMn (9 nm)/Ta (5 nm). The solid and dashed lines refer to the results for the continuous and the delayed sputtering methods, respectively.

strong NiO(111) peak is observed for the delayed deposition methods while no corresponding peak is found for the continuous method. For FeMn-pinning and NiO-pinning spin valves, a diffraction peak exists near $2\theta=44^\circ$. This can be attributed to Co(111), Cu(111), NiO(200), and FeMn(111). Although it is difficult to distinguish them, the Co and Cu layers are at least expected to have a stronger (111) preferred orientation for the delayed deposition method.

In order to get a more intuitive view of the microstructures of the Co and Cu layers, as an example, HRTEM imaging was carried out for Si/Ta (8 nm)/Co (3 nm)/Cu (2.5 nm)/Co (3 nm)/NiO (25 nm). Figure 3 shows the cross-section micrographs. For the delayed deposition method, the Co and Cu layers match laterally very well and the coherent growth between them occurs, which is extended into the NiO layer. Apparently, the interface between the Co and Cu layers is perfect geometrically. Moreover, for the delayed deposition method, the lateral size is as large as several tens of nanometers. This benefits from the fact that they are of fcc structure and the mismatch between the lattice constants of the Co and Cu layers is very small. One can also find that the (111) lattice planes in the Co and Cu layers are parallel to the film plane. The single-crystallite-like structure and coherent growth of fcc structure with (111) lattice plane parallel to the film plane have been observed in FeNi/Cu multilayers with Fe buffer layer and other multilayers consisting of Cu and Co.^{3,13} Figure 3(b) shows the results for the continuous method. In most regions, the (111) atomic planes in Co and Cu layers are not parallel to the film plane (marked by arrows), and some disordered regions can also be observed in Co and Cu layers. These results are in good agreement with the XRD curves.

In general, the crystalline structure depends strongly on the initial growth mode.¹³ The better crystalline structure for

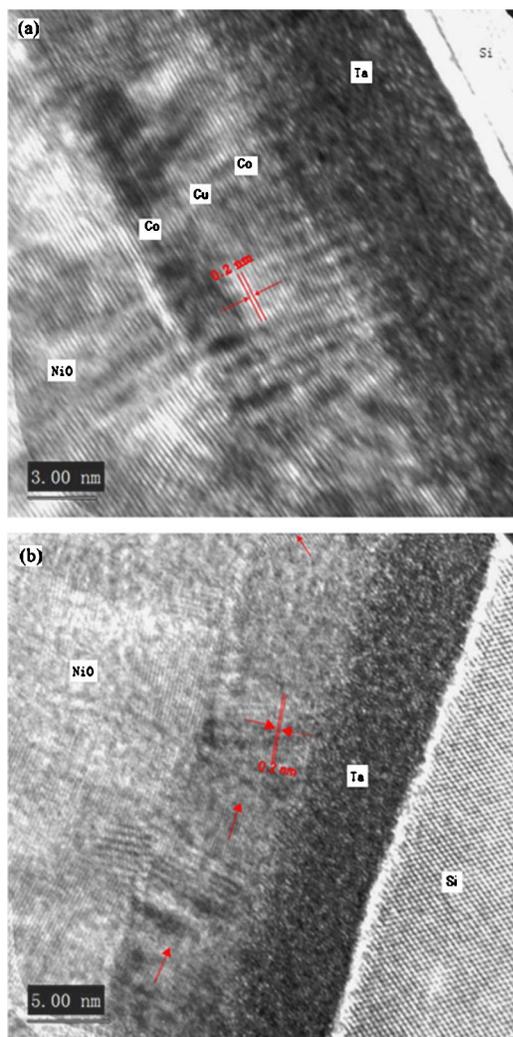


FIG. 3. (Color online) TEM images for the spin valves Si/Ta (8 nm)/Co (3 nm)/Cu (3 nm)/Co (3 nm)/NiO (25 nm) by (a) the continuous and (b) the delayed deposition methods.

the delayed deposition method can be attributed to three possible reasons.^{14,15} First, the Ta surface becomes smoother due to the motion of the Ta atoms during the interval. At ambient temperature, however, the motion is weak. Secondly, during the interval, there might be residual gases of O₂, H₂O, H₂, and so on. Molecules of these gases are adsorbed on the Ta surface and serve as a surfactant to induce the layer-by-layer growth of the Co layer. Thirdly, the nucleation density of Co atoms was enhanced significantly, which also favors the two-dimensional growth. Most likely, the third reason plays a determinant role in the better growth of Co layers for the delayed deposition method.

The enhancement of the GMR ratio and the spin-dependent scattering can be explained as a result of the coherent growth of Co and Cu layers, for to three possible reasons. First, for the delayed deposition method, the Co and Cu layers are grown in coherence, and at the same time the lateral size of the column is larger than that for the continuous method. Disordered regions are observed for the continuous method. Therefore, the mean free path is enhanced for the delayed deposition method, in comparison to the continuous method.¹³ Secondly, the chemical interfacial roughness and interfacial diffusion at Co/Cu are expected to be differ-

ent for the two fabrication methods, which might also result in an enhancement of the spin-dependent scattering in the delayed deposition. Thirdly, for the delayed deposition method, the (111) atomic planes of Co and Cu layers are mainly parallel to the film plane, having a stronger (111) preferred orientation.¹⁶ In contrast, for the continuous method, in most regions (111) atomic planes are not parallel to the film plane. This might also change the GMR ratio. In other words, it is the coherent growth that leads to the GMR enhancement for the delayed deposition method. As discussed earlier, the spin-dependent scattering is enhanced by the delayed deposition in FeMn-pinning and NiO-pinning spin valves. Because of the effect of the specular reflection, the enhancement of the spin-dependent scattering in the NiO-pinning spin valve is larger than that of the FeMn-pinning one. Therefore, the GMR enhancement in the NiO-pinning spin valve is larger than that of the FeMn-pinning one, as shown in Fig. 1.

In summary, as the continuous method, constituent layers were fabricated continuously. As the delayed deposition method, fabrication procedure was delayed by 1 h between the Ta buffer layer and followed ones. For the second method, the FeMn-pinning and NiO-pinning spin valves show the coherent growth of the Co and Cu layers with a stronger (111) preferred orientation. At the same time, the GMR ratio is enhanced by a factor of 20%–30%. It can be concluded that the spin-dependent scattering is enhanced by the coherent growth and (111) preferred orientations.

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