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Drastic change of giant magnetoresistance of Co/Cu multilayer by decreasing residual impurities in sputtering atmosphere

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In order to clarify the influence of residual impurities in the sputtering atmosphere on the microstructure and the giant magnetoresistance (GMR), Co/Cu multilayers were fabricated by changing the chamber pressures, P_b, just before introducing processing gas. P_b was controlled by changing the pumping time after venting the chamber with air. A drastic change of magneto resistance (MR) ratio from 48% to 14% was observed, when P_b was changed slightly from 7×10^{-8} to 3×10^{-8} Torr. In that P_b region, the root mean square (rms) roughness increased discontinuously from 3.7 to 4.6 Å as P_b was lowered. The abrupt drop of the MR ratio was accounted for by the decrease of the antiferromagnetic coupling accompanied by the increase of interfacial roughness. © *1999 American Institute of Physics.* [S0021-8979(99)73608-X]

I. INTRODUCTION

The giant magnetoresistance (GMR) effect in multilayers has been activity investigated by many researchers. However, multilayers with high magnetoresistance (MR) ratio are not consistently produced even at present because the microstructure of the multilayer is not carefully controlled. It is widely known that the purity of the sputtering atmosphere strongly affects the magnetic properties accompanied by significant change of microstructure in thin film media.[1] For multilayers, we have reported the significant change of MR ratio with increasing the impurities in the sputtering atmosphere by introducing air after pumping the chamber down to the base pressure.[2] Taking into account this fact, the differences in the purity of the respective sputtering atmospheres could be one of the causes of poor reproducibility of GMR in multilayers, because the purity of the sputtering atmosphere is easily changed by the base pressure of the chamber. The base pressure of the sputtering chamber usually used is in 10^{-7} Torr order, and the residual impurities, mainly H_2O, are not excluded enough. When the base pressure becomes lower than 10^{-9} Torr, the content of residual H_2O decreases and H_2 becomes a major element. In the present study, in order to clarify the influence of residual impurities on GMR, Co/Cu multilayers were fabricated by changing the base pressure of the sputtering chamber, and the correlation between their MR ratio and microstructure was investigated, especially focusing on the interfacial roughness.

II. EXPERIMENTAL PROCEDURE

Multilayers, of the form substrate/$(\mathrm{Co}\,d_{\mathrm{Co}}/\mathrm{Cu}\,d_{\mathrm{Cu}})_{30}$/Cu 20 Å were deposited on Si (100) wafers with a thermally oxidized layer at room temperature using a dc magnetron sputtering machine which enables to pump down to 2×10^{-11} Torr. The Co thickness, d_{Co} was 10 and 20 Å. The Cu layer thickness, d_{Cu} was varied within the range from 8 to 11 Å, where the so-called ''first peak'' of the GMR oscillation is expected. The thickness of each layer was controlled by changing the deposition time. The amount of residual impurities in the sputtering atmosphere was controlled by changing the chamber pressure, P_b, just before introducing sputtering gas. P_b was varied by changing the pumping time after venting the chamber with air. For the process gas, ultraclean Ar gas (UC-Ar), whose moisture level is less than 1 ppb,[1] was used. The flow rate of the UC-Ar gas during sputtering was 54 sccm to make the pressure ~0.6 mTorr. The microstructure of the multilayers was analyzed by x-ray diffraction (XRD) with a Cu $K\alpha$ radiation source and by transmission electron microscopy (TEM). $M-H$ loops were measured by a VSM at room temperature. The magnetoresistance was measured by a dc four-point probe method in a magnetic field up to 13 kOe at room temperature. The MR ratio was defined as $\Delta\rho/\rho_{13\,\mathrm{kOe}}\equiv(\rho_0-\rho_{13\,\mathrm{kOe}})/\rho_{13\,\mathrm{kOe}}$, where ρ_0 is the maximum resistivity at around zero field and $\rho_{13\,\mathrm{kOe}}$ is the resistivity under the applied field of 13 kOe, respectively. The saturation field, H_s was defined as the magnetic field where the resistivity is 1% larger than $\rho_{13\,\mathrm{kOe}}$ in the magnetoresistance curves which were corrected for the contribution of the forced effect in the high magnetic field. An antiferromagnetic (AF) coupling energy, J between adjacent Co layers in the multilayer was calculated as $M_sH_sd_{\mathrm{Co}}/4$, where M_s is the saturation magnetization.[3]

III. RESULTS AND DISCUSSION

A. GMR and magnetic properties

Figure 1 shows the MR ratio and $\rho_{13\,\mathrm{kOe}}$ of multilayers as a function of P_b. Here d_{Cu} was optimized 9–10 Å to make the MR ratio maximum. In the multilayers with $d_{\mathrm{Co}}=10$ Å, the MR ratio, which was about 39% around $P_b=3\times10^{-7}$ Torr, gradually increased with lowering P_b and reached about 48% at $P_b=7\times10^{-8}$ Torr. However, when P_b was lower than 7×10^{-8} Torr, the MR ratio drastically dropped and became about 14% at $P_b=3\times10^{-8}$ Torr. Further lowering P_b, the MR ratio decreased, and vanished at $P_b=7\times10^{-11}$ Torr. $\rho_{13\,\mathrm{kOe}}$ was constant at about 23 $\mu\Omega$ cm over the whole range of P_b in this study. These results indi-

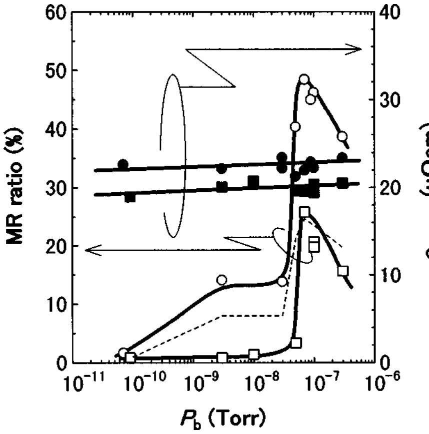

FIG. 1. Changes of MR ratio and $\rho_{13\,\mathrm{kOe}}$ as a function of the chamber pressure, P_b, for the multilayers with $d_{\mathrm{Co}}=10$ Å (circles) and $d_{\mathrm{Co}}=20$ Å (squares). Also shown as a dashed line is the calculated MR ratio of the multilayers with $d_{\mathrm{Co}}=20$ Å by using the results of $d_{\mathrm{Co}}=10$ Å.

cate the change of MR ratio was dominated by the magnetoresistance, $\Delta\rho$. In the multilayers with $d_{\mathrm{Co}}=20$ Å, we can see the similar dependence of MR ratio and $\rho_{13\,\mathrm{kOe}}$ against P_b, although with lower absolute values. The differences of MR ratio between the multilayers with $d_{\mathrm{Co}}=10$ Å and $d_{\mathrm{Co}}=20$ Å are explained by a shunting effect of current in the Co layers as follows. Using both $\rho_{13\,\mathrm{kOe}}$ values of the multilayers with $d_{\mathrm{Co}}=10$ Å and $d_{\mathrm{Co}}=20$ Å, we can estimate the resistivity of the Co layer, ρ_{Co} at 16.7 $\mu\Omega$ cm. From the experimental MR ratio of the multilayer with $d_{\mathrm{Co}}=10$ Å and the ρ_{Co}, we can calculate the MR ratio of the multilayer with $d_{\mathrm{Co}}=20$ Å (dashed line in Fig. 1). The agreement of the experimental value and the calculated one means that the MR ratio is diluted by shunting current with increasing d_{Co} in the case of the multilayers with $d_{\mathrm{Co}}=20$ Å.

Figure 2 shows the volume fraction of AF coupled regions of the Co layers at zero field, $1-M_r/M_s$, as a function of P_b for the multilayers with $d_{\mathrm{Co}}=10$ Å shown in Fig. 1. Remanent magnetization ratio, M_r/M_s was determined from an $M-H$ loop along the axis of easy magnetization in the multilayers. J of multilayers with $d_{\mathrm{Co}}=10$ Å was also shown. Here, d_{Cu} was optimized 8–9 Å to make H_s maximum. $1-M_r/M_s$, which was 0.45 around $P_b=3\times10^{-7}$ Torr, gradually increased with lowering P_b and reached a peak value of 0.80 at $P_b=7\times10^{-8}$ Torr, while, $1-M_r/M_s$ decreased steeply with the further lowering P_b reaching a value of 0.24 at $P_b=3\times10^{-8}$ Torr. This change of $1-M_r/M_s$ corresponds well to the change of the MR ratio shown in Fig. 1. In other words, the change in the MR ratio was caused by the change in the amount of antiparallel alignment of magnetization of the Co layers at zero field. J, which was 0.17 erg/cm^2 at $P_b=3\times10^{-7}$ Torr, gradually increased with lowering P_b and took a value of 0.20 erg/cm^2 at $P_b=7\times10^{-8}$ Torr. Further lowering P_b, J decreased similarly to $1-M_r/M_s$ reaching roughly half its maximum value, 0.13 erg/cm^2 at $P_b=3\times10^{-8}$ Torr. Therefore, the rapid drop of MR ratio between the multilayers fabricated under $P_b=7\times10^{-8}$ Torr and $P_b=3\times10^{-8}$ Torr was caused by diminishing the amount of antiparallel alignment of the magnetization of the Co layers, associated with the decrease of AF coupling energy, J. Such a change of J of the multilayers fabricated under various P_b is anticipated to be caused by microstructural change.

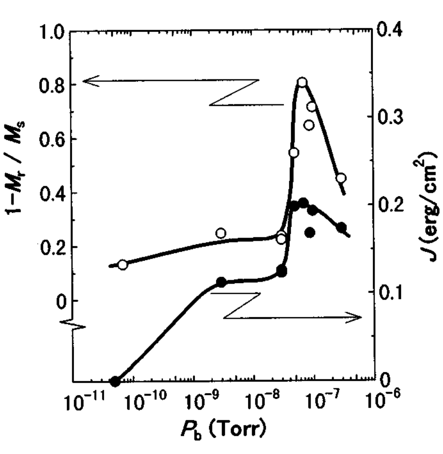

FIG. 2. Changes of $1-M_r/M_s$ and the AF coupling energy, J, as a function of the chamber pressure, P_b, for the multilayers with $d_{\mathrm{Co}}=10$ Å.

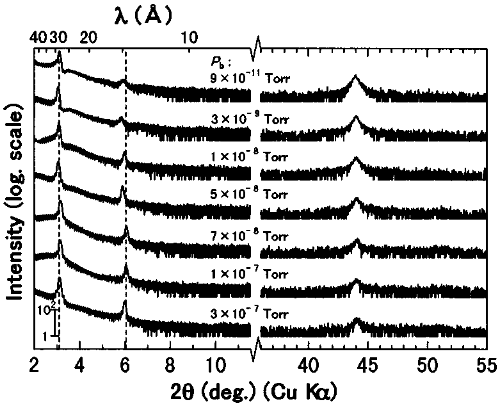

FIG. 3. Changes of XRD profiles of the multilayers with $d_{\mathrm{Co}}=20$ Å fabricated under the various chamber pressures, P_b.

B. Microstructure

Figure 3 shows the changes in XRD profile of multilayers with $d_{\mathrm{Co}}=20$ Å and $d_{\mathrm{Cu}}=9.5$ Å fabricated under various P_b. In the region of $2\theta=36°$–$55°$, interference peaks diffracted from Cu (111) and Co (111) planes are observed around $2\theta=44°$. The intensity of the interference peak became larger on lowering P_b, indicating the enlargement of grain size and the decrease of defects in the multilayers. The decrease of grain boundaries and defects means the decrease of scattering center and results in the enhancement of the AF coupling energy, J, because the coupling between the adjacent magnetic layers arises through conduction electrons. It is speculated that the enlargement of grain size and the decrease of defects induced the increase of J in the range of $P_b=3\times10^{-7}$–7×10^{-8} Torr as shown in Fig. 2.

On the other hand, in low angle region, the first and the second order diffraction peaks originated from the artificial period are observed around $2\theta=3°$ and $6°$, respectively. The

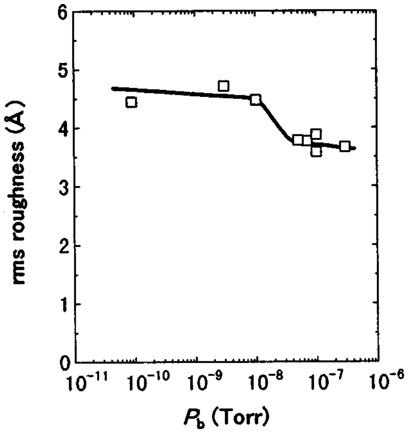

FIG. 4. Changes of the rms roughness as a function of the chamber pressure, P_b, for the multilayers with $d_{Co}=20$ Å.

observed positions of these peaks corresponded to the calculated ones from the reflectivity of the multilayer (dashed lines). Both the diffraction peaks and the finite-size peaks, which appears as a high frequency oscillation in the low angle region, diminished with lowering P_b, indicating the interfacial flatness deteriorates with lowering P_b.

According to Ueda *et al.*,[4] the root mean square (rms) roughness of the multilayer was calculated from these low angle XRD profiles. Figure 4 shows the rms roughness as a function of P_b. The rms roughness, which was about 3.7 Å in the multilayers fabricated under $P_b=3\times10^{-7}$–5×10^{-8} Torr, became about 4.6 Å in the multilayers fabricated under $P_b=1\times10^{-8}$–9×10^{-11} Torr. The important point to note is the correspondence of the discontinuous change of the rms roughness and the drastic change of MR ratio around $P_b\sim5\times10^{-8}$ Torr.

It is widely known that the interfacial roughness of multilayer induces a ferromagnetic (*F*) coupling between the magnetic layers. According to the Néel's model, the so-called ''orange peel effect,'' the *F*-coupling energy, J_f increases like a quadratic function with heightening amplitude, *h* of the sinusoidal waving interface when the wavelength, *L*, and the intermediate layer thickness, *d*, are constant.[5] In order to estimate J_f, induced by the interfacial roughness of the multilayers with lowering P_b, TEM cross section images of the multilayers with $d_{Co}=10$ Å and $d_{Cu}=9.5$ Å fabricated under $P_b=1\times10^{-7}$ and 7×10^{-11} Torr are shown in Fig. 5. In the multilayer with $P_b=1\times10^{-7}$ Torr, a continuous layered structure was observed. In contrast, in the multilayer with $P_b=7\times10^{-11}$ Torr, it was found that the interfaces are relatively rough, because of large undulations observed around columnar boundaries. The estimated values of *L* and *h* by taking an average from several undulations in the TEM images, were $L=120$ Å and $2h=7$ Å for $P_b=1\times10^{-7}$ Torr, $L=180$ Å and $2h=20$ Å for $P_b=7\times10^{-11}$ Torr, respectively. Using the intermediate layer thickness, 9 Å, J_f of the multilayers with $P_b=1\times10^{-7}$ and 7×10^{-11} Torr are calculated as 7×10^{-3} and 5×10^{-2} erg/cm^2, respectively. The difference of these J_f values is the same in order with the change of *J* against P_b as shown in Fig. 2. Namely, we can say that the amount of antiparallel alignment of the magnetization in the adjacent Co layers at zero field is decreased by the orange peel effect, and results in a fatal decrease of GMR.

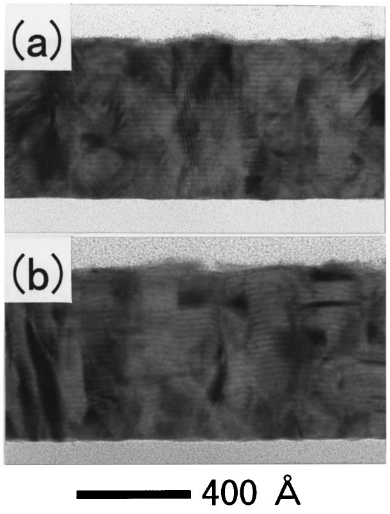

FIG. 5. TEM cross section images of the multilayers with $d_{Co}=10$ Å fabricated under the chamber pressures of (a) $P_b=1\times10^{-7}$ Torr and (b) $P_b=7\times10^{-11}$ Torr, respectively.

IV. SUMMARY

Co/Cu multilayers were fabricated by changing the chamber pressure, P_b. P_b was the pressure just before introducing sputtering Ar gas and varied by changing the pumping time after venting the chamber with air. The change of MR ratio against P_b was investigated in connection with the microstructure of multilayer. As results, we found that; (1) the MR ratio took a peak of 48% at $P_b=7\times10^{-8}$ Torr, and steeply dropped to 14% within the range of $P_b=7\times10^{-8}$–3×10^{-8} Torr; (2) in that the P_b region, rms roughness discontinuously increased from 3.7 to 4.6 Å as P_b was lowered; (3) the abrupt drop of the MR ratio was accounted for by the decrease of *J* accompanied by the increase of interfacial roughness.

We conclude that residual impurities have a strong effect on the microstructure to change drastically the magnetic property of multilayers, and that the control of impurities in the sputtering atmosphere is the important factor stabilizing the GMR.

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