Growth of giant magnetoresistance multilayers: Effects of processing conditions during radio-frequency diode deposition

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The magnetotransport properties of giant magnetoresistance multilayers are significantly affected by the atomic-scale structure of the interfaces between the nonferromagnetic conducting and ferromagnetic (FM) metal layers. The interfacial roughness and the extent of intermixing at these interfaces are both known to be important. A combination of experimental and multiscale modeling studies have been used to investigate control of interface structure during multilayer growth by rf diode deposition and the consequences of such control for magnetotransport. Experiments were conducted to evaluate the dependence of the magnetotransport properties of NiFeCo/CoFe/CuAgAu multilayers upon the growth conditions (background pressure, input power), and to link the roughness of vapor-deposited copper layers to the same process parameters. These experimental studies reveal the existence of intermediate background pressure (20 mTorr) and plasma power (175 W) that resulted in the highest magnetoresistance and a strong sensitivity of copper layer surface roughness to both the power and pressure at which deposition was conducted. By using a combination of modeling technologies, the deposition process conditions have been linked to the atomic fluxes incident upon the sample surface. This was then used to determine the atomic-scale roughness of the film. Energetic metal atoms (and inert gas ions) were found to have very strong effects upon interfacial structure. The models revealed an increase in interfacial roughness when metal (or inert gas ion) translational energy was decreased by either reducing the plasma power and/or increasing the background pressure. Because high-energy metal impacts activated atomic jumping near the impact sites, high plasma power, low background pressure process conditions resulted in the smoothest interface films. However, these conditions were also conducive to more energetic Ar ion bombardment, which was shown by molecular dynamics modeling to induce mixing of the FM on the copper interface. Intermediate plasma powers/background pressures result in the most perfect interfaces and best magnetotransport. The insights gained by the modeling approach indicate a need to avoid any energetic ion bombardment during the early growth stages of each new layer. This could be accomplished by operating at low power and/or high pressure for the first few monolayers of each layer growth and may provide a growth strategy for further improvement in magnetotransport performance. © 2001 American Vacuum Society.

I. INTRODUCTION

Giant magnetoresistance (GMR) metal multilayers consisting of alternating ferromagnetic and nonferromagnetic layers can exhibit large changes in their electrical resistance when a magnetic field is applied.1–10 Though the phenomenon was only discovered about 12 years ago, it has become the basis for a growing number of important technological applications. For example, multilayers based upon NiFeCo/CoFe/CuAgAu/CoFe/NiFeCo are used for magnetic-field sensors,4,6 and related devices are being investigated for magnetic random access memories (MRAM).5 GMR-based MRAM has many potentially attractive features, including nonvolatility, low-power consumption, and high access speed. Both classes of applications have motivated interest in the design of economical vapor deposition processes that can produce thermally stable GMR devices with high GMR ratios (defined as the maximum resistance change divided by the resistance at magnetic saturation) and low magnetic saturation fields.

The best GMR multilayers appear to require the growth of metal multilayers with well-controlled interfaces.1–3,10–13 Interfacial roughness results in Néel coupling, electron scattering, and weakened antiferromagnetic coupling, which are all deleterious to the GMR properties.2,5 Interfacial (chemical) mixing results in dead (nonmagnetic) layers at the ferromagnetic (FM)–nonmagnetic metal spacer layer interfaces, reduced FM layer exchange coupling, and reduced GMR properties.2,5

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Recent atomistic modeling studies have shown that interfacial structural imperfections are effected by the depositing vapor atom’s incident energy, the deposition rate, and the characteristics of other (e.g., inert gas ion) fluxes incident upon the substrate. Since these are governed by the deposition process and process conditions, the magnetotransport properties are expected to depend upon the method and conditions used for device growth by vapor deposition.

Here, we experimentally investigate the dependence of the magnetotransport properties upon the background pressure and input plasma power during the growth of a GMR sensor multilayer system. A simplified multiscale modeling approach is used to relate these conditions to the atomic fluxes incident upon the growth surface. Atomistic modeling is then used to investigate the atomic assembly mechanisms during film growth as a function of growth conditions. The dependence of interfacial roughness and mixing upon the growth conditions is identified and compared with ex-
perimentally measured performance of the devices. The study supports the developing view that no existing vapor deposition technology is ideally suited to the growth of GMR multilayers. The modeling aided insights developed here are used to suggest alternative growth strategies for the growth of improved GMR materials.

II. EXPERIMENTS

The study here focused upon the influence of input plasma power and the background gas pressure on the GMR ratio and the saturation magnetic field of rf diode deposited GMR multilayers. A schematic diagram of a rf diode sputter deposition system with a plane parallel electrode geometry is shown in Fig. 1. An inert gas (argon) plasma is initiated and maintained between the target and the substrate by the rf power source. In the rf discharge, electrons respond more rapidly to the changing electrical field and tend to flow from the plasma to adjacent conducting surfaces at a faster rate than the ions. Plasma potentials are, therefore, developed at both the target and the substrate. The positive working gas

![Graph](image1)

**Fig. 5.** Dependence of deposition rate for the four targets, (a) shows the effect of the background pressure at a fixed input power (175 W), and (b) shows the effect of the input power at a fixed background pressure.

![Graph](image2)

**Fig. 6.** Dependence of magnetotransport properties upon (a) and (b) the background pressure and (c) and (d) the input power for multilayers with a fixed CuAgAu layer thickness of 16 Å.
ions are accelerated across these sheath potentials to strike both the target and the substrate. The magnitude of the sheath potentials differs in an asymmetric discharge. The voltage drops across the sheaths are inversely related to the sheath capacitances, and thus the substrate and target areas. A bias voltage, therefore, exists, as shown in Fig. 2. As a consequence, one observes that the bombarding ion energy at the typically larger area substrate electrode is less than that at a smaller target electrode.

The Ar$^+$ ions accelerated across the target sheath bombard the target surface and result in sputtering of metal atoms from the target. These sputtered atoms are then transported to the substrate and deposited. The system geometry, the rf power applied to the plasma, and background pressure determine the energies and fluxes of the ions incident upon the target. These, together with the properties of the target, determine the energy, angular distribution, and flux of the atoms emitted from the target. Scattering of the sputtered metal atoms by working gas atoms during diffusional transport to the substrate modifies these quantities. The final energy and angular distributions of the metal atoms incident upon the substrate, therefore, depend on the target-to-substrate distance, the working gas type, rf power, and background pressure. Working gas ions are also accelerated across the substrate sheath and strike the substrate with the energies in the 40–140 eV range.$^{11,12}$ The energy and flux of both the metal and argon ions as they strike the substrate can have potentially important effects upon the resulting GMR properties.$^{1-13}$

To evaluate the significance of these effects, GMR multilayers were grown at Nonvolatile Electronics, Inc. (NVE) (Eden Prairie, MN). The architecture and composition of the antiferromagnetic coupled multilayers are shown in Fig. 3. A Randex model 2400-6J rf diode system was used to deposit the metal films. Figure 4 shows the internal layout of the rf diode sputtering deposition chamber. The diameter of the targets was 20.32 cm, and the distance between substrate and target was 3.81 cm. Four different composition targets were installed in the chamber. 4-in.-diam silicon wafers were used for the substrates. An amorphous 2000 Å Si$_3$N$_4$ film was grown on top of each wafer using a chemical-vapor-
deposition method. The Si₃N₄ film acted as an electrically insulating and diffusion inhibiting layer between the silicon wafer and the subsequently deposited metal film. The rms roughness of the Si₃N₄ film was 1.5 Å.

The deposition rate and composition for each target under the different deposition conditions had been measured previously and are shown in Fig. 5. A 40 Å Ta seed layer was deposited at 20 mTorr and 175 W on the substrate prior to the start of multilayer growth. Over most of the pressure range studied, the deposition rate decreased slightly with background pressure, Fig. 5(a). The deposition rate increased with input power. Figure 5(b) shows that the variation was almost linear. The layer thicknesses were, therefore, controlled by timing the deposition. The substrate was then moved under each target to deposit a multilayer structure. Figure 6 shows the dependence of the measured GMR ratio and saturation magnetic field (H₉₀) upon the background pressure and input power. The maximum GMR ratio was achieved at an intermediate background pressure (20 mTorr) and input power (175 W). The saturation magnetic field (H₉₀) of this condition (20 mTorr and 175 W) was also moderate and reasonably well suited for magnetic sensing applications.

III. MULTISCALE MODELING

A multiscale modeling approach, Fig. 7, has been used to investigate the origin of the relationships between the experimental observations and growth conditions. The details of the approach are described in Refs. 11, 12, and 13. A CFD analysis of the reactor yielded the velocity and pressure distribution of the argon working gas flow in the sputtering chamber and, in particular, between the target and the substrate. A one-dimensional plasma model was used to deduce the bias voltage, the argon ion energy, and flux incident upon both the target and the substrate. A three-dimensional (3D) MD analysis of sputtering was used to quantify the yield, the energy, and the angular distributions of the copper atoms sputtered from the target by argon ion impact. A DSMC atom transport model was then used to track the propagation of metal atoms from the target to the substrate. This enabled calculation of their incident energy and angle as a function of working gas pressure, spacing, temperature, and the plasma power. The outputs of these transport calculations were finally used as inputs in a two-dimensional (2D) KMC model recently developed to simulate the atomic assembly and thus structure of a film as a function of the process conditions.

IV. SIMULATION RESULTS

Figure 8 shows a comparison between the measured and plasma model simulated bias voltage (i.e., the magnitude of the difference between the two sheath voltages), as a function of the background pressure and input power during deposition of copper film. Good agreement between the simulations and the experimental measurements performed with the NVE rf diode sputtering chamber was obtained.
Fig. 10. Model predicted average energy of copper atoms and argon ions incident upon the substrate. (a) shows the effect of background pressure at a fixed input power (175 W), and (b) shows the effect of input power at a fixed background pressure of 20 mTorr.

Figures 9(a) and 9(b) show the pressure dependence of the Ar\(^+\) ion current densities (or ion flux) and the ion energy at both electrodes computed using the plasma model. The energy and ion flux at the target are seen to be about a factor of 10 higher than those at the substrate. Even so, significant ion fluxes with energies in the 50–100 eV range impact the substrate under the conditions used to grow the GMR films. The energy of the ions at both electrodes decreased sharply with pressure. However, the Ar\(^+\) ions fluxes at both electrodes are seen to rise slightly as the pressure increased from 10 to 50 mTorr. The reason for the decrease in ion energy with pressure resulted from more collisions between ions and working gas across the target sheath. The collision frequency also increased with the pressure, and therefore, the probability of argon gas ionization ratio also increased. Figures 9(c) and 9(d) show that increasing the input power also significantly increases the Ar\(^+\) ion current density (flux) and energy. The energy of the Ar\(^+\) ion was governed by the voltage at the sheath, which is proportional to the input power. The results also show that the energy of the Ar\(^+\) ions at the substrate was much smaller than the Ar\(^+\) ion energy at the target. The ion flux increased 2.5 times as the power was increased from 50 to 350 W. Typically, the ion flux impacting the substrate was about a tenth of that impacting the target.

A DSMC transport model was used to track the paths of representative sputtered atoms of specified energy and emission direction as they propagated through the background gas. This enabled the likelihood that an atom reached the substrate to be determined together with atoms incident energy and the incident angle. More than 10,000 atoms were analyzed to obtain statistical representations of these data. The sputtered atom average energy was then deduced at a fixed distance (3.81 cm) from the target for different chamber pressures and input powers. The energy of sputtered copper atoms after propagating from the target to substrate are shown in Fig. 10, together with that for the Ar\(^+\) ion. The gas pressure controlled the frequency of argon atom collisions with sputtered atoms as they traveled from the target to the substrate. The number of collisions was proportional to the pressure, since these collisions transfer energy from hot copper atoms to the thermalized argon gas. This leads to a reduction in both copper atom kinetic energy and deposition efficiency as the gas pressure increases, Fig. 10(a). It is also noted that the Ar ion flux incident upon the substrate also has an incident energy that drops sharply with background pressure. The power dependence of the copper atom and Ar\(^+\) ion energies are shown in Fig. 10(b). The copper atoms’ energy rose weakly with power because of the rise of the emitted atom energy with power was overcome by energy transfer to the working gas during transport to the substrate.

The metal flux, its energy, and incident angle distributions were used as inputs in a 2D KMC model to predict surface morphology of copper films, see Figs. 11 and 12.\textsuperscript{22–25} The KMC model implements hot atom effects (reflection, resputtering, bias, and athermal diffusion) deduced from MD calculations and is described in Ref. 22–25. The KMC analysis reveals that the roughness of copper films increased as the plasma power was decreased and/or the background pressure was increased and was corroborated by atomic-force microscopy images of copper films. To compare trends, we normalized the experimental and simulated data by dividing by the first data point of each data set. Figure 13 shows that the normalized roughness deduced from experiments and by simulation had a similar dependence upon the power and pressure used during deposition.

The modeling of copper deposition indicates that under the conditions that led to a maximum GMR ratio (20 mTorr/175 W), the copper atoms were effectively thermalized. Thermally activated atom hopping was the principle mechanism of film growth when the power was low and/or the pressure was high. The initial rise in the GMR ratio with power is in part a result of the surface smoothing associated with more energetic metal atom impacts. The drop in GMR as the pressure is increased beyond 20 mTorr also is linked to a reduction in surface mobility as metal atom energy decreases. However, the occurrence of a peak followed by a reduction in the GMR ratio by further increasing the plasma.
power cannot be explained by a metal impact energy mechanism. The most likely cause for the reduction in the GMR ratio appears to be related to changes in the Ar$^+$ ion flux and impact energy at the substrate.

A 3D MD model was used to explore the effect of Ar$^+$ ion impacts with a model interface as a function of the ion energy (and, therefore, process conditions). Details of the method can be found in Refs. 18–21. The model geometry is shown in part (a) of Fig. 14. The light and dark balls in the Fig. 14 represent copper and cobalt atoms, respectively. The substrate is copper, which contains 120 (224) planes in the y direction, 12 (111) planes in the z direction, and 52 (220)
planes in the $x$ direction. An array of cobalt pyramids was constructed on the copper surface to simulate cobalt island nucleation. To minimize the effects of the small crystal size, periodic boundary conditions were used. A fixed boundary condition was applied to the lowest monolayer of atoms to avoid translation. The temperature of the three layers above this fixed layer was kept at a fixed substrate temperature of 300 K using a thermostat algorithm. 150 Ar$^+$ ions then bombarded the surface at a normal incident angle. Figure 14 shows that this resulted in significant smoothing and intermixing of the copper and cobalt by an exchange mechanism analogous to that described in Ref. 20. The modeled depen-
The dependence of intermixing upon the input power and the background pressure is shown in Fig. 15. The results indicate that significant intermixing (alloying) of cobalt atoms in the copper occurs, and that it increased with increasing ion impact energy, which in turn depended upon the process power/pressure. As a consequence, the intermixing fraction decreased with increasing background pressure and/or decreasing input power. Intermixing results in a dead magnetic layer at the nonmagnetic and ferromagnetic interface, which can contribute to a reduction in GMR properties.

By combining the simulations of roughening and intermixing, Fig. 16, it can be seen that the existence of a GMR ratio maximum (at 175 W/20 mTorr) results from the competing effects of increased adatom surface mobility induced flattening and ion impact induced intermixing as either the power increase or pressure decrease. The latter dominates at high power/low pressure and results in a loss in GMR ratio. Likewise, the former is promoted by high power and low pressure. The highest GMR ratio was obtained under conditions when some energetic adatom (and ion) smoothing occurred with minimal intermixing.

Simulations of metal deposition (with no Ar\(^+\) ion bombardment) have suggested processes that reduce both the interfacial roughness and the chemical intermixing, which has led to a proposed energy modulated deposition technology.\(^{18,19,20}\) During an energy modulated method, low incident metal atom energies are used to deposit the first few monolayers of each new metal layer and higher energies are used for the reminder of the layer. This strategy was found to result in an optimal combination of interfacial roughness and interfacial mixing. In rf diode sputtering, a modulated Ar\(^+\) ion energy deposition could be implemented by using low power and/or high pressure (low Ar\(^+\) ion energy) growth for several angstroms of each metal layer (with reduced intermixing) and using high power and/or lower pressure (high Ar\(^+\) ion energy) for growth of the remaining layer (with smooth interface). Such a modulated plasma power and/or pressure strategy may significantly improve the performance of giant magnetoresistive multilayers grown by rf diode sputter deposition.

**V. SUMMARY**

Multilayers grown by rf diode deposition are found to exhibit a maximum GMR ratio when grown at intermediate pressure and plasma power (175 W/20 mTorr). This is thought to be linked to the interface structure of the CoFe on CuAgAu interface. A combination of surface characterization and modeling methods has been used to analyze the rf
diode deposition of metal multilayers. As the pressure is decreased or the power increases from the optimum (20 mTorr/175 W), measurements of the copper layer deposition and calculations indicate a rise in the copper atom’s impact energy, which induces significant copper surface flattening. This is also accompanied by a sharp rise in the impact energy of Ar⁺ ions, which also contribute to smoothing. MD calculations show that their impact also causes alloying of the cobalt on the copper interface (by an impact exchange process). This mixing becomes more severe as the impact energy increases. An optimal combination of smoothing and mixing is achieved at intermediate background pressure and plasma power. Since mixing is more difficult after several monolayers of CoFe have been deposited, the use of low power to deposit the 50% of the CoFe on Cu layer followed by higher power for its completion may result in improved GMR devices.

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