Biased Target Ion Beam Deposition of GMR Multilayers

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Abstract—Multilayers like those used in giant magnetoresistive spin-valves can be improved if better layer thickness uniformity, lower contamination levels, reduced interfacial roughness and less interlayer mixing can be achieved. Atomic simulation studies have revealed that optimization of the energy of the depositing atoms and the application of very low energy inert gas ion assistance reduce both interfacial roughness and interlayer mixing. Modulation of the energy of these fluxes as each layer growth progresses has been predicted to give even better quality interfaces. Unfortunately, these concepts cannot be implemented in conventional physical vapor deposition (PVD) or ion beam deposition (IBD) processes used to deposit these materials. A new biased target ion beam deposition (BTIBD) system that enables these conditions to be achieved has recently been developed. Unlike conventional IBD, it uses a low energy ion source. The higher ion energy required for the sputtering is obtained by applying a negative bias voltage to the metal targets. This system enables low energy ion assistance at the growth surface. By modulating the bias voltage during each layer growth, it is also possible to change the average energy of the depositing atoms and therefore enable control of the atomic assembly at interfaces. We have used BTIBD to grow model Ta (40 Å)/Ni$_{80}$Fe$_{20}$ (40 Å)/Co (15 Å)/Cu (t$_{Cu}$/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin-valves that show improved GMR ratios and coupling fields over traditional IBD grown multilayers.

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1. INTRODUCTION

Giant magnetoresistive (GMR) multilayers $^{[1,2]}$ consisting of two 20 – 100 Å thick ferromagnetic (e.g., Ni, Fe, Co or their alloy) layers separated by a thin 10 – 30 Å thick conductive spacer (e.g., Cu) layer are designed to allow an external magnetic field to switch the magnetic moments of the two magnetic layers from an anti-parallel and to a parallel alignment. Due to spin-dependent electron scattering $^{[3]}$, these materials exhibit a significant (~5-10%) drop in their electrical resistance if a sufficiently large external magnetic field is applied to achieve full domain reversal. In simple GMR sandwich structures, the anti-parallel magnetic moments result from exchange coupling between two closely separated magnetic layers $^{[2,4]}$. However, this coupling is strong and a large applied magnetic field is required to align the moments and obtain the desired GMR effect. In a spin-valve device, an additional anti-ferromagnetic (e.g., FeMn) layer is deposited to pin the magnetic moment of one of the magnetic layers $^{[5]}$. A continuous adjustment of the moment alignment between the two magnetic layers can then be achieved by rotating the moment of the free magnetic layer using an external field. This eliminates the need for exchange coupling. As a result, spin-valves can be made with a thicker spacer layer that induces little exchange coupling, and as a result, the GMR effect can be achieved at a much lower external magnetic field.

Spin-valve structures have been extensively used as read head sensors by the magnetic recording industry $^{[5]}$. They have significantly boosted hard drive disk storage capacity over that projected for competing read head technologies $^{[6]}$. Pseudo-spin-valves are also being explored for a new type of magnetic random access memories (MRAM) $^{[7]}$. For these applications, the materials must have a high GMR ratio (defined as the maximum resistance change divided by the resistance at magnetic saturation), a low saturation magnetic field, a high sensitivity (relatively large change in resistance for small change in applied field), a near zero coercivity, a weak temperature dependence and a high thermal stability. These properties can be improved when the multilayers have sharp flat interfaces $^{[8,9,10,11]}$. Nanoscale multilayers with all interfaces smooth are not stable and cannot be obtained under equilibrium growth conditions $^{[12]}$. This in part accounts for the experimental
observation that low temperature deposition methods such as magnetron sputtering and ion beam deposition (IBD) produce better GMR multilayers than the molecular beam epitaxy (MBE) method [9] even though the latter can be carried out in a higher vacuum (cleaner) chamber. These deposition processes also have other important differences. Perhaps the most significant are the average energy, the average incident direction of adatoms when they strike the growth surface, and the presence of energetic assisting ions. MD simulations indicated that increasing adatom energy promotes flat interfaces due to an impact induced surface flattening effect [13,14]. However, high-energy adatoms are more likely to exchange with underlying atoms upon impact, resulting in mixing when they are deposited on a surface of different composition [13,4]. The tradeoff between interfacial roughness and interlayer mixing accounts for the observation that the best GMR multilayers are obtained at an intermediate adatom energy in rf diode [15] and magnetron sputtering [16]. This is consistent with MD predictions that the best adatom energy lies between 1-4 eV [13,14]. The insights obtained in the simulations also indicate an improved growth approach. If a low adatom energy is used to deposit the first a few monolayers of a new material layer, mixing at the interface can be reduced. A high energy can then be used to deposit the remainder of the layer with a flattened surface without causing mixing.

These MD studies also show that when the adatom incidence is far from the growth surface normal (oblique deposition), significant surface/interface roughness develops due to shadowing [17]. Substrate rotation can minimize this shadowing, and as a result, greatly reduce the surface/interface roughness created during oblique angle deposition. Rotation also reduces thickness variations (especially if flux shapers are used).

MD simulations [18] further indicated that during low energy Xe$^+$ ion assistance, a rough surface can be smoothed if the ion energy is between 5 and 15 eV. However, impact induced mixing starts to become significant at an ion energy above 6 eV. The tradeoff between roughness and mixing appears to indicate an optimal Xe ion energy around 9 eV. This is in excellent agreement with a recent report that inert gas ion assistance with a low energy of 9 eV produced the best Cr/Sc X-ray mirror multilayers [19]. It is also consistent with experimental findings that ion assisting energies above 20 eV reduce GMR properties [20].

During sputtering processes, a fraction of the high-energy inert gas ions striking the target are reflected as neutrals. These neutrals can have energies in excess of 50 eV and are often able to reach the growth surface [21] where they cause significant interlayer mixing in multilayers [22]. MD simulations of sputtering indicated two major methods to reduce this type of surface damage [21]. First, the use of heavier inert gas ions such as Xe$^+$ rather than Ar$^+$ can greatly reduce both the energy and flux of reflected neutrals. For instance, during 500 eV normal incidence ion sputtering of a (111) Ni surface, the reflection probability and average energy of reflected neutrals are ~0.5 and ~58.87 eV respectively with Ar$^+$ ions. They drop to ~0.15 and ~6.57 eV respectively with Xe$^+$ ions. Second, the use of a sputtering inert gas flux that impacts the target at a normal incident angle also reduces both the energy and flux of reflected neutrals. During 600 eV Ar$^+$ sputtering of a (111) Ni surface, the reflection probability and average energy of reflected neutrals are ~0.95 and ~200 eV at a ion incident angle of 60°, and they drop to ~0.5 and ~20 eV respectively at a normal ion incident angle.

In a conventional IBD system, Fig. 1, a primary grid ion beam gun is used to generate high-energy inert gas ions (Ar$^+$, Kr$^+$ and Xe$^+$ etc.) that are directed at a depositing metal target. These ions sputter metal atoms from the target surface. These metal atoms are then transported to the substrate (as the depositing flux). Growth occurs when these atoms condense on the substrate surface. Multiple targets can be installed in the target assembly and targets can be switched during deposition of multilayers. A second grid ion beam gun can be used to create low-energy inert gas ions that are directed at the substrate to assist the growth. During deposition, the substrate is allowed to rotate. Normally, the energy of the sputtering ions, the energy of assisting ions, the incident angle of sputtering ions at the target, and the incident angle of assisting ions at the substrate can all be controlled. However, the geometry of the ion beam, target and substrate means that a normal ion incident angle at the target cannot be achieved. To minimize the effect of the resulting neutrals, Xe is used instead of Ar as the sputtering gas. This has been found to be essential to produce high quality GMR multilayers using IBD deposition [23].

![Fig. 1. Conventional ion beam deposition.](image-url)
Traditional deposition processes (e.g., molecular beam epitaxy, magnetron sputtering, and ion beam deposition) are not designed to accommodate the optimized GMR growth conditions identified above \[8\]. This paper describes a new biased target ion beam deposition (BTIBD) technology that enables some of these conditions to be achieved. This new growth technology has been applied to deposit a model spin-valve system and is shown in preliminary operations to significantly improve the GMR properties over those achieved using a conventional ion beam deposition (IBD) method.

2. CONVENTIONAL ION BEAM DEPOSITION

The above discussions indicate that a controllable adatom energy in the range between 1 and 5 eV is important for the growth of good GMR multilayers. To date, magnetron (and rf diode) sputtering and IBD are the methods most successfully applied to deposit GMR multilayers \[12\]. However, none of them is ideally suited for optimal processing of atomically engineered interfaces required by devices. For instance, rf sputtering systems usually operate at high pressures (\(10^{-1} – 10^{-3}\) Torr), and the adatom flux is substantially scattered (thermalized) by the low energy background gas during transport from target to substrate \[24\]. Adatoms hence have relatively low energies. These adatoms are well suited for deposition at interfaces for avoiding interlayer mixing, but are poorly suited for effectively flattening the surface. Conventional IBD, on the hand, can only be operated using relatively high sputtering ion energies and relatively low pressure with no substantial scattering from the background gas. As a result, the adatoms in IBD can have significantly higher energies. For instance, MD simulations of the sputtering of (110) Ni surface indicated that the average energy of the sputtered Ni atoms is around 15 and 12 eV respectively during 600 eV Xe\(^+\) and Ar\(^+\) impacts \[21\]. While these atoms are better suited for flattening the surface, they are poorly suited for avoiding mixing at interfaces.

There are a number of other drawbacks with the IBD design shown in Fig. 1. First, the lowest inert gas ion energy normally achievable in commercially available grid ion beam guns is about 50 eV. This ion assistance energy is significantly higher than the ideal energy desired for ion-assisted growth of GMR multilayers (although atomistic simulations have shown that by inclining the gun so that the ions make oblique impact with the substrate and by using lighter atomic weight ions such as neon, the optimum ion energy can be increased \[23\]). Second, the primary sputtering ions impact the target at an oblique incident angle, which results in many high energy reflected neutrals unless Xe is used. Third, while IBD systems are designed to contain the sputtering ions on the target, in practice, a fraction of the ions always miss the target. These high-energy ions then sputter undesired materials from some of the vacuum system hardware resulting in “overspill” contamination of the growth film. In order to reduce beam overspill, high performance IBD systems generally employ large expensive targets. The majority of the ions are focused to the center of the target resulting in highly concentrated target wear and poor utilization of the target material. The concentrated sputtering at the target center also significantly reduces the deposition thickness uniformity that is essential for device performance. Fourth, IBD is constrained to high sputtering beam voltages (>500 V). This is because a low voltage beam is difficult to focus which is essential for reducing overspill contamination. Deposition rates in IBD systems (typically 0.1 to 2 Å/sec.) are lower compared to other sputtering deposition techniques. It is then critical to increase the deposition rate so that the contamination from residual gases in the vacuum chamber does not exceed the threshold value that affects properties. Experimental observations indicated that the GMR properties always increase as the deposition rate is increased during IBD deposition \[23\]. Processing at low beam voltages is thus prohibited because it results in low deposition rates and low wafer output. Under high beam voltage conditions, the lower adatom energy desired for the interface deposition as described above cannot be achieved. For instance, the average energy of sputtered atoms is only reduced from 22 to 15 eV when the beam voltage is reduced from 1000 V to 600 V \[21\]. If scattering with the background gas is considered, this translates into an adatom energy range of 9-13 eV \[21\]. Fifth, in a conventional IBD process, a dynamic change of deposition energy required for the intralayer energy modulation cannot be achieved because a change of the sputtering ion beam gun energy will necessarily be accompanied by a change in the ion beam plume characteristics.

Saito et al. \[26\] have measured the GMR ratio of IBD deposited \([\text{Ni}_{80}\text{Fe}_{20}(20Å)/\text{Cu}(50Å)/\text{Co}(10Å)/\text{Cu}(50Å)]\) multilayers as a function of Ar\(^+\) ions impacts \[21\]. Their best GMR ratio was obtained at an intermediate ion acceleration voltage of 600 – 800 V, Fig. 2. Their best GMR ratio was obtained at an intermediate ion acceleration voltage of 600 – 800 V, in agreement with other reports \[27\]. The occurrence of a GMR peak is consistent with a need to tradeoff interfacial roughness for some interlayer mixing. However, the adatom energy corresponding to 600 – 800 eV sputtering gas ions is around 9 eV \[21\]. This is much higher than that normally used in magnetron and rf diode sputtering depositions. The inability for IBD to produce better GMR properties at lower acceleration voltages must therefore be attributed to the overspill contamination and the residual gas contamination during the reduced deposition rates.

3. BIASED TARGET ION BEAM DEPOSITION

Based upon the insights discussed above, a biased target ion beam deposition (BTIBD) technology \[28\] has been developed to overcome some of the problems of the conventional IBD method.
The concept of the BTIBD approach is schematically shown in Fig. 3. Six different targets can be installed in a target assembly so that multilayers of up to six different materials can be deposited. The wafer stage is at the left side of the chamber facing the sputtering target. The wafer plane can be tilted so that the adatom incident angle can be adjusted from 0° (normal incidence) to 90°. During deposition, the wafer can be rotated about its axis. A magnetic field (not shown) can be applied parallel to the growth surface to deposit layers with desired magnetic orientations.

The key to this BTIBD system is that instead of using conventional grid ion beam guns, it uses low energy ion sources that combine end-Hall ion [29] and hollow cathode electron sources [30]. The hollow cathode electron source is used to eject electrons into the end-Hall ion source. The latter can then maintain a stable plasma. A special feature of this kind of ion source is that it can reliably produce a very high density of inert gas ions with a very low energy (from several eV up). In Fig. 3, one such a ion source is constructed to face the substrate stage and is used to create low energy assisting ions (5-50 eV) ideal for GMR multilayer deposition. Another similar ion source is used to provide a high density of low energy (5-50 eV) ions in front of the sputtering target. By applying a negative bias to the sputtering target, the inert gas ions can strike the target at a near-normal incidence. This reduces both the energy and flux of reflected neutrals. Depending on the bias voltage, the impact energy of the ions on the target can be easily controlled between 50 and 2000 eV. Since the other hardware (such as target shield and chamber wall etc.) is grounded, most ions will strike the negatively biased target. Even though some ions miss the target, they will have only the un-accelerated low energy that is below the sputtering threshold. As a result, they do not cause overspill contamination. This not only eliminates the need to capture all of the ions on the target, but it also allows the use of an ion beam that is much broader than the target. Consequently, the entire target can be more uniformly illuminated, and both the target material utilization efficiency and deposition thickness uniformity are significantly improved. Since overspill contamination is reduced even when the ion beam is not focused, and a much higher ion density can compensate for the loss of deposition rate due to low sputtering yield, BTIBD allows the use of lower sputtering energies (i.e., low bias voltages). It also has the potential to increase the deposition rate if the increased ion flux can over compensate for the reduced sputtering yield per ion.

A plasma sheath develops at the surface of the negatively biased target. Because the sheath is thin (~2 mm) compared to the spacing between the ion source and target, the target bias has no substantial effect on the ion trajectories from source to target. Hence, for constant source operation, the illumination profile and the ion current reaching the target are nearly independent of the target voltage. The target shield to target spacing is closer than a shield distance at the edges of the target to prevent penetration of the plasma. A large range of target voltages (~100 to >1000 V) can be used while maintaining reasonable deposition rates. This facilitates smooth control of target bias voltage and provides a means for the eventual realization of intralayer energy modulation during deposition. In addition, the ion sources are capable of operating over a broad range of processing pressures (10⁻⁴ to 5×10⁻³ Torr). For the geometry shown in Fig. 3, this means that the sputtered atoms are on average scattered by the background gas about once or twice at a 10⁻⁴ Torr pressure and about 50-100 times at a 5 ×10⁻³ Torr pressure. One or two collisions with the background gas is expected to reduce the energy to about 5 - 20 eV
range. However, collisions with the background gas 50-100 times should fully thermalize the adatoms so that their energies are reduced to ~0.1 eV. For this reason, BTIBD provides a means of control adatom energies over a wide range (from sub eV to several tens eV) [21].

4. GROWTH OF SPIN-VALVES

Preliminary runs have been carried out to evaluate the biased target ion beam deposition technology. For all the tests, the base pressure was about $4 \times 10^{-8}$ Torr, the working pressure was about $4.5 \times 10^{-4}$ Torr, the depositing flux was about $55^\circ$ off from the normal to the growth surface, the bias voltage was 300 V, and the substrate was rotated during deposition. The adatom incident angle of $55^\circ$ is far from ideal, and the bias voltage was thought to be low from a conventional IBD viewpoint (e.g., see Fig. 2). However, both conditions are illustrative of the capabilities of the BTIBD method.

Pure Ta, Ni$_{80}$Fe$_{20}$, Co, Cu, and FeMn films were deposited to a thickness of between 1000 and 2000 Å to calibrate the deposition rates, deduce the resistivity and estimate the thickness uniformity. We found that Cu could be deposited at a rate of around 0.55 Å/sec. This is almost an order of magnitude increase over that of IBD operated at the same ion energy (300 eV). The Cu resistivity was about 2.6 $\mu$Ω·cm at a 1010 Å thickness. This low resistivity is an indication of minimal contamination. For all the films deposited, we obtained a deposition thickness variation of less than 3% over a 150 mm wafer surface (around 0.5% in most cases). This high deposition thickness uniformity was achieved without using a flux shaper. Previously, comparable uniformity had not been achieved by IBD even on a 125 mm wafer. We attribute this difference in uniformity to the superior illumination provided by BTIBD.

Deposition of model Ta (40 Å)/Ni$_{80}$Fe$_{20}$ (40 Å)/Co (15 Å)/Cu (t$_{Cu}$)/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin-valves has been carried out. The magnetoresistance curve measured for an Ar$^+$ sputtered spin-valve with a Cu layer thickness of 33 Å is shown in Fig. 4. A relatively high GMR ratio of 6.972% was obtained. It is well known that in IBD, a Xe sputtering gas produces much better GMR properties than Ar$^+$ ions. While the GMR ratio may be further increased by fine-tuning the spacer layer thickness, a GMR ratio of 8.0% is at least a 5% increase over previous IBD films of the same structure. Of all the samples deposited using Xe gas, we found an average coupling field of 10 Oe. This was also improved from 15 Oe obtained from similar spin-valves deposited using IBD.

The effects of spacer layer thickness upon Ar$^+$ sputtered spin-valve were also explored. The GMR ratio was measured as a function of the spacer layer thickness and the results are shown in Fig. 5. Fig. 5 indicates that the GMR ratio of the Ar sputtered spin-valves increases with the spacer layer thickness over the entire range explored, and has not yet reached the maximum value. Many mechanisms can contribute to the spacer layer thickness dependence of the GMR ratio. For instance, the GMR ratio exhibits periodic oscillation with spacer layer thickness due to the exchange coupling between the two magnetic layers [2,4]. This effect is relatively insignificant in the spin-valve studied here where the spacer layer thickness is much thicker than the 2nd peak value. An additional interesting finding is that while we also deposited many spin-valves with spacer layer thickness less than 25 Å, no GMR effect was discovered in these films. Since the films are deposited at an oblique angle of $55^\circ$, they can have rough surface/interfaces [17]. We suspect that pinholes formed when the spacer layer is thin.
5. CONCLUSIONS

Atomistic simulations have indicated that conventional IBD does not provide ideal conditions for GMR spin-valve deposition. Significant problems include overspill contamination, poor deposition uniformity, inefficient target material utilization, high energy and flux of reflected neutrals, low deposition rate, lack of energy control at the low end, and inability to apply low energy ion assisted deposition. A novel biased target ion beam deposition system has been constructed to address all of these problems. Preliminary runs using BTIBD matched or exceeded the GMR properties of a model spin-valve device obtained with IBD.

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