

Biased Target Ion Beam Deposition of Spin-valves

H. N. G. Wadley (haydn@virginia.edu), X. W. Zhou, and J. J. Quan
Department of Materials Science and Engineering, University of Virginia, Charlottesville, VA 22903

T. Hylton, and D. Baldwin
4Wave, Inc. Sterling, VA 20166

Abstract— A further reduction of defect concentration in spin-valve multilayers is difficult in today's growth processes. Multilayers with better layer thickness uniformity, lower contamination and reduced interfacial roughness and interlayer mixing can have significantly improved properties. Atomistic simulations revealed that a modulation of the energy of depositing atoms during deposition of each material layer or the application of very low energy inert gas ion assistance could reduce both interfacial roughness and interlayer mixing. These concepts, unfortunately, cannot be implemented in the conventional physical vapor deposition (PVD) or ion beam deposition (IBD) processes currently 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 the conventional IBD, it uses low energy ion source. The high ion energy required for the sputtering is obtained by applying a negative bias voltage to the metal target. This system enables the 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 enables control of the atomic assembly at interfaces. We have used this approach to grow Ta (40 Å)/Ni₈₀Fe₂₀ (40 Å)/Co (15 Å)/Cu (t_{Cu})/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin-valves and show improved GMR ratio and coupling field over traditional IBD grown multilayers.

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1. INTRODUCTION

Giant magnetoresistive (GMR) multilayers^[1,2] consisting of two thin (20 – 100 Å) ferromagnetic (e.g., Ni, Fe, Co or their alloy) layers separated by a thin (10 – 30 Å) conductive spacer (e.g., Cu) layer are designed to allow an external magnetic field to switch the magnetic moments of the two magnetic layers between anti-parallel and parallel alignments. Due to spin-dependent electron scattering^[3], such materials exhibit a large drop in electrical resistance upon the application of an external magnetic field. In simple GMR sandwich structures, the anti-parallel moments can only be achieved by the exchange coupling between the two magnetic layers^[2,4]. However, this coupling is usually so strong that a large magnetic field is required to align the moments and obtain the desired GMR effect. In spin-valve multilayers, an anti-ferromagnetic (e.g., FeMn) layer is added to pin the magnetic moment of one of the magnetic layer^[5]. A continuous adjust of the moment alignment between the two magnetic layers can then be achieved by rotating the moment of the free magnetic layer using the external field. This eliminates the need for the exchange coupling. As a result, spin-valves can use a thicker spacer layer that induces little exchange coupling. Their GMR effect is hence much more sensitive to the external field.

Spin-valve structures have been extensively used as read head sensors^[5] in the magnetic recording industry, and 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] that promise non-volatility, radiation hardness, low power consumption, densities comparable to dynamic random access memory and access speeds comparable to static random access memory. 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. A deposition process that can improve these properties is the key for spin-valves to be further successfully used in various applications. This paper describes a new biased target ion beam deposition (BTIBD) technology developed to incorporate the insights

obtained from atomistic simulation of vapor phase deposition of GMR multilayers. This new technology has been applied to deposit spin-valves and is shown to significantly improve the GMR properties over those achieved using the conventional ion beam deposition (IBD) method.

2. ATOMISTIC SIMULATIONS: OPTIMAL DEPOSITION CONDITIONS FOR SPIN-VALVES

Fundamental studies have quickly pointed out the desired microstructures for spin-valve multilayers. Extensive experiments have shown that the properties of GMR multilayers are extremely sensitive to the thickness of the spacer layer. Interfacial roughness can sharply reduce the GMR ratio^[8]. For spin-valves, the optimal spacer layer thickness lies between 25 and 35 Å. Depending on the design, the magnetic layer thickness can be between 20 and 100 Å. Under this structural constraint, interfacial roughness on the scale from nanometers to tens of nanometers can lead to a magnetic coupling of dipolar origin called “orange peel” coupling. This coupling increases the difficulty of rotating the moment in one magnetic layer relative to that in the other. It was also established that the majority electrons in Cu traveling nearly parallel to the layers can be totally reflected at the Cu/Co interfaces in the Co/Cu/Co multilayers because the majority Fermi surface of Co is slightly smaller than the Fermi surface of Cu. This will happen on both sides of the Cu layer if the moments in the ferromagnetic layers are aligned but on only one side if they are anti-aligned. Because these electrons stay in the Cu layer where the resistivity is low, the parallel resistance is significantly decreased and the GMR is increased. This “wave guide” effect requires atomically smooth interfaces^[9].

Atomic mixing at the magnetic and non-magnetic layer interfaces can significantly damage the GMR properties^[10,11]. For instance, when Ni diffuses into Cu, it can lose its moment and become a center for spin-independent scattering that reduces the GMR. Fe or Co diffused into the Cu can have an even worse effect because they will maintain their moment, but may cause spin-flip scattering. Significant mixing can virtually change the spacing between the two magnetic layers due to the creation of magnetic dead-layers at the interfaces^[12]. It is hence essential to minimize both interfacial roughness and interlayer mixing in order to improve the spin-valves.

Thermodynamics analysis indicated that at least for binary systems, nanoscale multilayers with all interfaces smooth are not stable^[13]. They hence cannot be obtained under equilibrium growth conditions. This in part account for the experimental observation that high-energy deposition methods such as magnetron sputtering and ion beam deposition (IBD) produce much better GMR multilayers than the equilibrium vapor energy molecular

beam epitaxy (MBE) method^[13] even though the latter can be carried out in a higher vacuum and cleaner chamber. To compare different deposition systems, processing conditions defined in the tool control panel must be converted to more fundamental expressions such as adatom energy, adatom incident angle, deposition rate, and substrate temperature. To identify microstructures at an atomic level, high-resolution transmission electron microscope needs to be used. Because both of these are not easily achieved, the identification of optimal growth conditions for GMR multilayers is difficult to explore experimentally.

Using an embedded atom method potential database^[14] to accurately define the interatomic forces between a variety of metal elements, molecular dynamics (MD) can be used to simulate the growth of metal multilayers. Because Newton’s equation of motion is used in MD to track the position of each atom during deposition, MD can realistically revealed the formation of atomic structure as a function of vapor deposition condition. Extensive MD simulations of vapor deposition of GMR multilayers properties have been carried out, resulting in a number of interesting findings^[14,15,16,17,18,19,20].

First, MD simulations have been used to study the effects of adatom energy on the interfacial roughness and interlayer mixing^[14,15]. Fig. 1 shows an example result for the vapor deposited CoFe/Cu/CoFe/NiFe multilayer, where the left panel was obtained from simulation at an adatom energy of 3.0 eV, an adatom incidence perpendicular to the growth surface, a substrate temperature of 300 K, and a deposition rate of 1nm/ns, and right panels were obtained from three dimensional atom probing of a sputter sample. Excellent agreement can be seen between simulation and experiment, as they both indicated a more diffused CoFe-on-Cu interface than the Cu-on-CoFe interface.

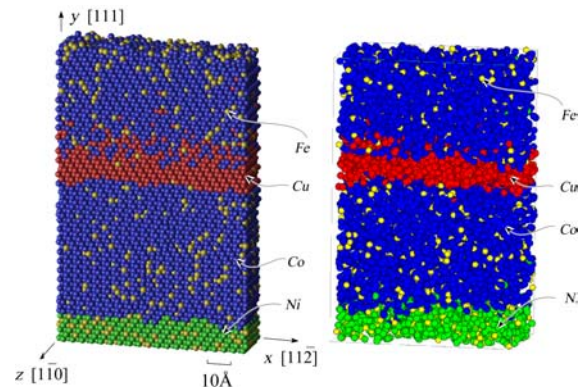


Fig. 1. Atomic Co₉₀Fe₁₀/Cu/Co₉₀Fe₁₀/Ni₈₀Fe₂₀ multilayers. The left panel is obtained from simulation at an adatom energy of 3.0 eV; the right panel is obtained from atom probe experiment of a sputtered sample^[15].

Most significantly, these studies ^[14,15] revealed that the interfacial roughness and interlayer mixing are very sensitive to the adatom energy. Increasing adatom energy enhances the reconstruction of local surface asperities during adatom impact, resulting in surface flattening. A flat surface then evolves into a flat interface after a new overlayer is deposited. 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 species. Cu atoms have a lower cohesive energy and a larger size than Co and Fe atoms, and therefore they tend to segregate on surface to reduce surface energy and surface tensile stress. Because of this, Co or Fe atoms impacting a Cu surface are more likely to exchange with the underlying Cu atoms than when Cu atoms impact a CoFe surface. As a result, the CoFe-on-Cu interface is seen to be more mixed than the Cu-on-CoFe interface in Fig. 1. The continuous exchange of the Cu atoms to the surface during deposition of the CoFe layer also results in Cu atoms to be mixed in later deposited CoFe layer.

The tradeoff between interfacial roughness and interlayer mixing accounts for the observation that the best GMR multilayers are all obtained at an intermediate adatom energy in rf diode sputtering ^[21], magnetron sputtering ^[12], and IBD ^[22]. Simulations further showed that this energy lies between 1-3 eV. The insights obtained in the simulations also indicated an improved approach. If a low energy is used to deposit the first a few monolayers of a new material layer, the mixing at the interface can be reduced. A high energy can then be used to deposit the remainder of the layer to flatten the surface without causing mixing. While no previous deposition systems were designed to apply intralayer energy modulation, such a scheme has been used in simulation to grow the same CoFe/Cu/CoFe/NiFe multilayer as shown in Fig. 1. An example of the deposited structure is shown in Fig. 2, where the energy scheme is indicated, and other conditions are the same as those used to obtain Fig. 1. Fig. 2 proves that the modulated energy deposition has the potential significantly further reduce both interfacial roughness and interlayer mixing.

MD simulations have also been used to study the effects of adatom incident angle on the deposition of a Ni/Cu/Ni multilayer ^[16]. Fig. 3 shows typical results of the multilayer structures obtained with and without substrate rotation at an incident angle of 1.0 eV, an oblique incident angle of 50°, a substrate temperature of 300 K, and a deposition rate of 10 nm/ns. It can be seen that deposition at an oblique adatom incident angle could cause significant surface roughness during deposition of the Ni layer (corresponding to a rough Cu –on–Ni interface) due to a shadowing effect ^[16]. Substrate rotation can reduce the effect of shadowing. As a result, the greatly reduce the surface/interface roughness created the during the oblique angle deposition condition.

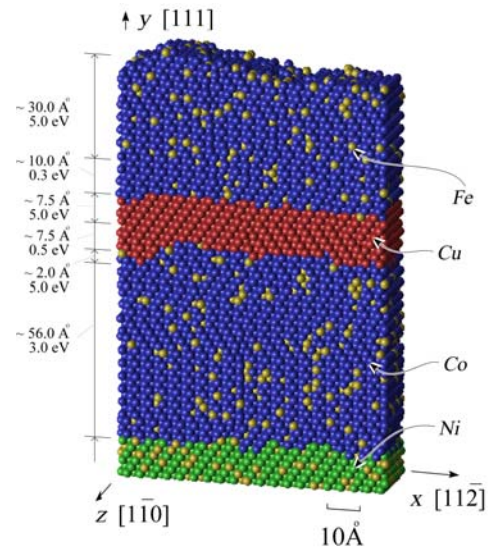


Fig. 2.
Atomic Co₉₀Fe₁₀/Cu/Co₉₀Fe₁₀/Ni₈₀Fe₂₀ multilayer obtained from simulation using an intralayer energy modulation scheme ^[15].

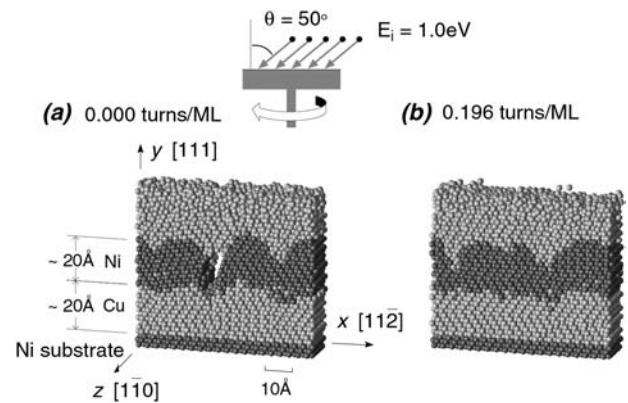


Fig. 3.
Simulated Ni/Cu/Ni multilayer deposited at 1.0 eV and an oblique incident angle of 50°. (a) No substrate rotation and (b) substrate rotation rate 0.196 turns/ML.

MD approaches have been set up to study the effects of low ^[17] and high ^[19] energy inert gas impacts on the GMR multilayer structures. Fig. 4 indicates that during low energy Xe irradiation on a rough Ni-on-Cu surface, the roughness can be reduced by increasing Xe energy from zero to 15 eV. However, the impact induced mixing starts to become significant at a Xe energy above 6 eV. The tradeoff between roughness and mixing appears to indicate an optimal Xe 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 that are similar to GMR multilayers ^[23]. It is consistent with the experimental finding that ion assisting energies above 20 eV reduce GMR properties ^[24].

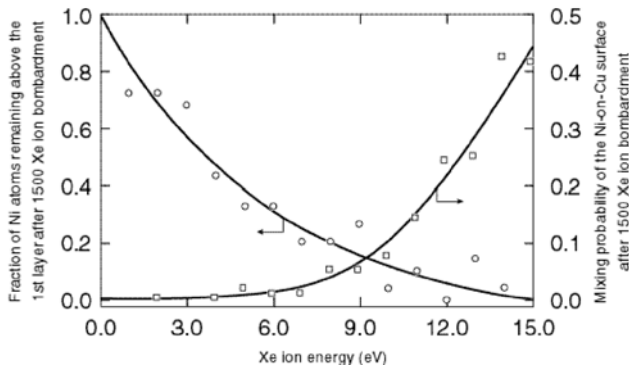


Fig. 4.

A characterization measure of surface roughness and exchange mixing on a model Ni-on-Cu surface as a function of Xe ion impact energy.

During magnetron (or rf diode) and IBD processes, a fraction of the high-energy inert gas ions used for the sputtering at the target can be reflected as neutrals. These neutrals can have energies in excess of 50 eV and can reach the growth surface [20]. These reflected neutrals were found to be a major cause for significant interlayer mixing in the sputtered GMR multilayers [19]. MD simulations of sputtering indicated two major methods to reduce this type of mixing [20]. First, the use of heavier inert gas ions such as Xe^+ rather than Ar^+ ions for the sputtering can greatly reduce both energy and flux of reflected neutrals. Second, the use of a sputtering inert gas flux that impact the target at a normal incident angle also reduces both energy and flux of reflected neutrals. Due to the triangular geometry of ion beam gun, target and substrate, the normal ion incident angle at the target cannot be achieved in conventional IBD systems, As a result, the use of Xe instead of Ar as the sputtering gas has been found to be essential to produce good GMR multilayers [25] during traditional IBD deposition.

3. CONVENTIONAL ION BEAM DEPOSITION

The above discussions indicated that a controllable adatom energy in a range between 1 and 5 eV is the key to grow good GMR multilayers. Up to date, magnetron (or rf diode) sputtering and IBD are the two deposition methods most successfully applied to deposit GMR multilayers [13]. However, none of them is ideally suited for optimal processing of atomically engineered interfaces required by devices. For instance, magnetron and rf diode systems operate at high pressures and the adatom flux is substantially scattered by the low energy background gas during transport from target to substrate. Adatoms hence have relatively low energies. These adatoms are well suited for deposition at interfaces for avoiding interlayer mixing, but are poorly suited for deposition of other parts, as they cannot effectively flatten the surface. The conventional IBD, on the hand, can only be operated using relatively high sputtering ion energies with no

substantial scattering from the background gas. As a result, the adatoms in IBD can have significantly higher energies. While they are better suited to the need for flattening the surface, they are poorly suited to the need for reducing the mixing at the interfaces. In order to explore ways to improve IBD by incorporating the knowledge obtained from atomistic simulations, we analyze the IBD system using a schematic illustration shown in Fig. 5.

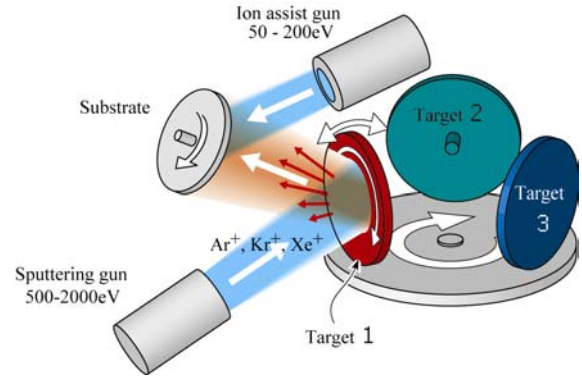


Fig. 5.

Conventional ion beam deposition approach.

In the conventional IBD system shown in Fig. 5, 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 can sputter metal atoms off from the target surface. These metal atoms are then transported to the substrate (as the depositing flux). Growth occurs when these atoms are condensed on the substrate surface. Multiple targets can be installed in the rotational 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, substrate is allowed to rotate. Normally, the energy of 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, but often within a limited range.

There are a number of drawbacks with the design shown in Fig. 5. First, the lowest energy normally achievable in commercially available grid ion beam gun is about 50 eV. This is significantly higher than the ideal energy desired for ion-assisted growth of GMR multilayers. Second, the primary sputtering ions must impact the target at an oblique incident angle that may cause high energy and flux of reflected neutrals. Third, while IBD systems are designed to contain all sputtering ions on the target, a fraction of the ions may miss the target in practice. These high-energy ions may sputter off 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 targets. Because the majority of the ions are focused only near the center of the target, this results in highly concentrated target wear at its center and poor utilization of the target material. The concentrated sputtering at the target center also significantly reduces the deposition thickness uniformity that is essential to the GMR multilayers. Fourth, it is difficult to apply lower adatom energy desired for the interface deposition as described above. This is because at low beam voltages, it is more difficult to focus the sputtering ion beam, resulting in larger overspill contamination. In addition, 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 to affect the properties. This has been verified by the experimental observation that the GMR properties always increase as the deposition rate is increased during IBD deposition [25]. Processing at low beam voltages also results in low deposition rates and low wafer output. Fifth, in a conventional IBD process, a instantaneous change of deposition energy required by the intralayer energy modulation scheme cannot be achieved because a change of the sputtering ion beam gun energy will necessarily be accompanied by the adjustment of the corresponding change of ion beam plume.

Saito et al. [22] have measured the GMR ratio of the $[\text{Ni}_{80}\text{Fe}_{20}(20\text{Å})/\text{Cu}(50\text{Å})/\text{Co}(10\text{Å})/\text{Cu}(50\text{Å})]_{10}$ multilayers Deposited using IBD at various Ar^+ acceleration voltages. To further examine IBD systems, their results are reproduced in Fig. 6. Fig. 6 indicates that the best GMR ratio is obtained at an ion acceleration around 600 – 800 eV, in agreement with other report [26]. The adatom energy corresponding to this ion acceleration voltage is much higher that 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 to attributed to the overspill contamination and the residual gas contamination during the reduced deposition rates.

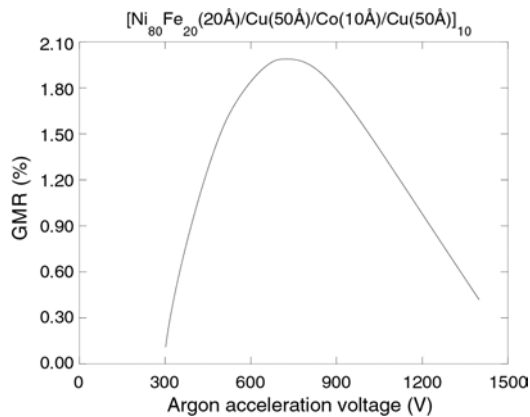


Fig. 6.
GMR ratio as a function of Ar acceleration voltage.

4. BIASED TARGET ION BEAM DEPOSITION

Based upon the insights discussed above, a new biased target ion beam deposition (BTIBD) technology [27] has been developed to overcome the problems of the conventional IBD method. The first such a BTIBD system has been successfully assembled at 4Wave, Inc., and is now being operated at University of Virginia.

The concept of the BTIBD system is schematically shown in Fig. 7. Six different targets can be installed in the target assembly so that multilayers of up to six different layers 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 is allowed to rotate along its axis. A magnetic field along the growth surface can be applied to deposit layers with desired magnetic orientation.

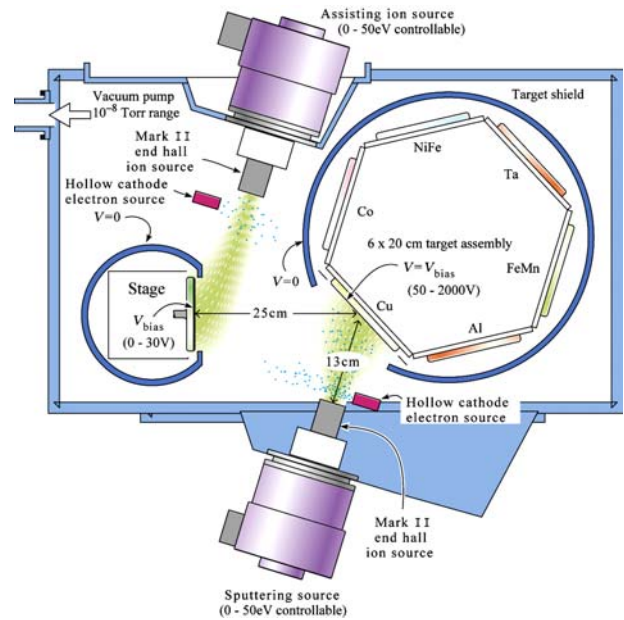


Fig. 7.
Biased target ion beam deposition approach.

The key to this BTIBD system is that instead of using the conventional grid ion beam gun, it uses novel low energy ion sources that combine an end-Hall ion source [28] and a hollow cathode electron source [29]. 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). In Fig. 7, one such a ion source is constructed to face the substrate stage and is used to create low energy assisting ions (0-50 eV) ideal for GMR multilayer deposition. Another similar ion source is used to provide a low energy (0-50 eV), and high density of 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. Depending on the bias voltage, the impact energy of the ions on the target can be easily controlled between 50 and 2000 eV that is suitable for the sputtering. Since the other hardware (such as target shield and chamber wall etc.) is grounded, most ions will strike the negatively biased target. Even some ions miss the target, they will have only the un-accelerated low energy that is not sufficient to cause the overspill contamination. This not only eliminates the need for an effort to capture all of the ions on the target, but also allows the use of an ion beam that is much broader than the target. As a result, the entire target can be uniformly illuminated, and both the target material utilization efficiency and deposition thickness uniformity are significantly improved. Because overspill contamination is reduced even when the ion beam is not focused, and a much higher ion density can compensate 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 increased the deposition rate desired for IBD deposition of GMR multilayers.

A plasma sheath develops at the surface of the negatively biased target. Because the sheath is very small (~2mm) 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 eV) can be used while maintaining reasonable deposition rates. This facilitates a smooth control of target bias voltage and a 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), allowing some control of the adatom scattering from the background gas. For these reasons, BTIBD is ideal for processes that seek to control adatom energies over a wide range.

5. GROWTH OF SPIN-VALVES

Preliminary runs have been carried out to evaluate the novel biased target ion beam deposition technology. For all the tests, the base pressure was about 4×10^{-8} Torr, the working pressure was about 4.5×10^{-4} Torr, the depositing flux is about 55° off from the normal of the growth surface, and the bias voltage was 300 V. The adatom incident angle of 55° is far from ideal, and the bias voltage was thought to be low from conventional IBD viewpoint (e.g., see Fig. 6). However, both conditions are illustrative of the capabilities of the BTIBD method.

Pure Ta, Ni₈₀Fe₂₀, Co, Cu, and FeMn films were deposited to a thickness between 1000 and 2000 Å to

calibrate the deposition rates, resistivity and thickness uniformity. We found that Cu has a deposition rate around 0.55 Å/sec. This is almost one order of magnitude increase over IBD operated at the same ion energy (300 eV). The Cu resistivity is about $2.6 \mu\Omega \cdot \text{cm}$ at 1010 Å thickness. The relatively low resistivity is an indication of no or extremely small overspill contamination. Of all the films deposited, we obtained deposition thickness variation of less than 3% over an entire 150 mm wafer surface (around 0.5 % in most cases). This high deposition thickness uniformity was achieved without using a deposition flux shaper. Previously, the 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 as compared to IBD.

Deposition of example Ta (40 Å)/Ni₈₀Fe₂₀ (40 Å)/Co (15 Å)/Cu (t_{Cu})/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin-valves has been carried out. The GMR curve measured for a Ar sputtered spin-valve with a Cu layer thickness of 33 Å is shown in Fig. 8. A relatively high GMR ratio of 6.972% was obtained. It has been well known that Xe produced much better GMR properties than Ar as the sputtering gas in IBD^[26,30]. Nonetheless, the GMR value obtained in Fig. 8 is close to that obtained by IBD using Xe gas^[26]. It is also significantly improved over a GMR value of 2.5% obtained for a similar spin-valve, Ta (25 Å)/Ni₈₀Fe₂₀ (60 Å)/Co (15 Å)/Cu (33 Å)/Co (15 Å)/NiFe (30 Å)/FeMn (80 Å)/Ta (50 Å), deposited by IBD using Ar gas at the same sputtering energy of 300 eV^[30].

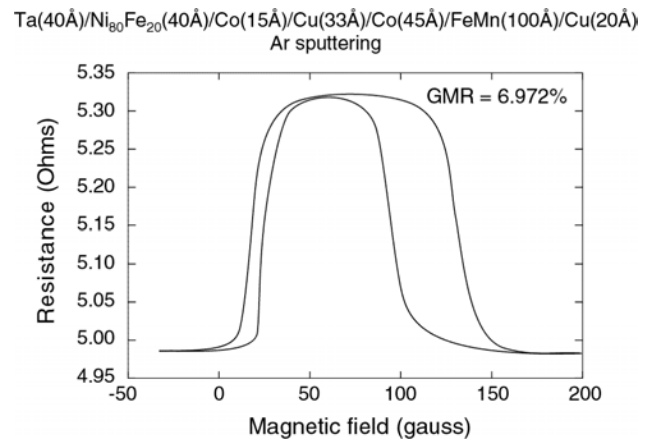


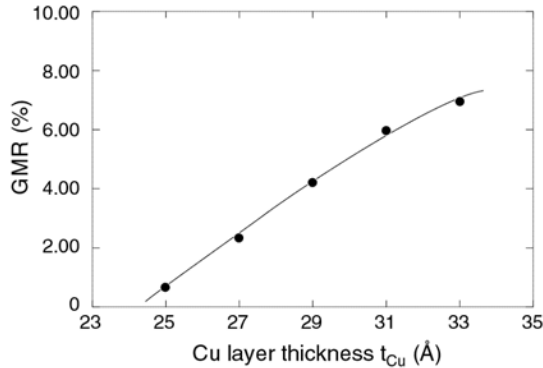
Fig. 8.
Resistance vs. field for an Ar sputtered spin-valve.

The effects of spacer layer thickness were also explored. The GMR ratio measured as a function of the spacer layer thickness is shown in Fig. 9(a) for Ar sputtered spin-valves and Fig. 9(b) for Xe sputtered spin-valves. Fig. 9(a) indicates that 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. Fig. 9(b) shows that the Xe sputtered spin-valves reached a GMR ratio above 8 % at a Cu

spacer thickness of about 27 Å. While the GMR ratio may be further increased by fine-tuning the spacer layer thickness, the GMR ratio of 8.0 % is at least 5% increase in the GMR ratio 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-valve deposited using IBD.

Ta(40Å)/Ni₈₀Fe₂₀(40Å)/Co(15Å)/Cu(*t*_{Cu})/Co(45Å)/FeMn(100Å)/Cu(20Å)

(a) Ar sputtering



(b) Xe sputtering

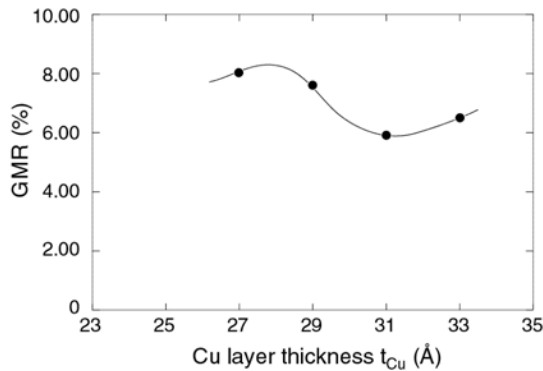


Fig. 9.

GMR ratio as a function of Cu layer thickness of a spin-valve. (a) Ar sputtering and (b) Xe sputtering.

We point out that this is our first set of runs and we have not yet started to optimize the deposition conditions.

6. CONCLUSIONS

Atomistic simulations indicated that the conventional IBD does not provide ideal conditions for GMR spin-valve deposition. The 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. Based on these analyses, a novel biased target ion beam deposition system has been constructed to overcome all these problems. Preliminary runs using BTIBD easily produced better GMR properties

over those obtained from IBD. BTIBD appears to have the potential to be significantly optimized.

7. ACKNOWLEDGEMENTS

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