Low energy ion beam assisted deposition of a spin valve

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The spin dependent electron transport in giant magnetoresistive (GMR) multilayers is significantly affected by the atomic scale structure of their interfaces. Devices with atomically flat and chemically sharp interfaces are preferred for magnetic sensor and memory applications. Recent atomic simulations of the atom-by-atom assembly of these devices indicate that near optimal interfacial structures can be created using low energy, ion assisted vapor deposition techniques with ion energies in the 5–10 eV range. A recently developed biased target ion beam deposition system has been used to experimentally test this hypothesis. Prototypical Ta/NiFe/Co/Cu/Co/FeMn/Cu spin valve structures were first grown using (simultaneous) argon ion assistance during deposition of the Co/Cu/Co trilayer part of the spin valve multilayer. Assisting ion energies of around 10 eV resulted in structures with a 30% higher magnetoresistance ratio and significantly reduced coupling field compared to samples grown with no ion assistance or with ion energies above 15 eV. These results are consistent with the atomistic simulation predictions. Other promising ion assistance schemes identified by the simulations were then used to deposit the Ta, NiFe, FeMn, and the top copper layer. A near optimal strategy was identified that resulted in the further improvement of the GMR ratio.
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I. INTRODUCTION

Metal multilayers have many applications. For instance, some exhibit giant magnetoresistance (GMR) and can be used to make magnetic sensors and magnetic tunnel junction devices.[1,2] Others are high spectral reflected at suboptical wavelengths set by the film layer thickness and are used for extreme ultraviolet and x-ray mirrors.[3,4] Others exhibit high strength and hardness[5,6] and might eventually be used for erosion resistant coatings. The performance of the materials used in all these applications is critically affected by their interfacial structures. For most applications, materials with atomically flat and chemically sharp interfaces are preferred.[2,7,8]

Ion beam assistance during the vapor deposition of a monolithic thin film is often used to enhance its surface smoothness.[9] However, atomistic simulations using molecular dynamics techniques indicate that if this ion assistance strategy is used during the deposition of multilayers, it is essential that low energy (less than a few eV) atoms or ions are used to avoid intermixing at interfaces.[10,11] When interlayer mixing does occur, it can lead to a dead magnetic layer in GMR structures and increase electron scattering which both degrade the GMR effect.[8,12] An optimum, ion mass specific energy must exist because the use of too low an ion energy usually results in rough surfaces and interfaces which also degrade GMR properties.

Recent (molecular dynamics) simulations of the low energy ion assisted deposition of Co/Cu multilayers have identified near optimal ion assistance recipes for the growth of high quality interfacial structures with reduced roughness and intermixing.[10,13,14] The atomistic simulations reveal that the energy barriers for surface flattening and interlayer mixing mechanisms are both low.[10] However, the critical ion energy for flattening is usually less than that for mixing under oblique ion illumination conditions.[10] Ion assistance with energies above the flattening energy but below the mixing threshold therefore appear promising.[13] For ions with a similar atomic mass to that of the metal (e.g., argon ions for the Cu–Co system), the use of a fixed ion energy of 8 eV at an incidence angle of around 50° during ion assisted (simultaneous) deposition was predicted to give the best interfacial structures.[10,13] Other ion assistance approaches including modulated ion energy (where the ion energy was gradually increased as each layers' thickness increased) and sequential ion beam assistance (where the ion assistance was used after deposition of each layer) also appeared equally promising and enabled the use of higher energies once an interface was buried by several monolayers of the new metal.[14]

Here, this model predicted low energy ion assistance strategies which are experimentally investigated using a specially constructed biased target ion beam deposition (BTIBD) system designed to access the low energy ion assistance regime.[15,16] The vapor deposition tool used here enabled independent control of the ion energy, incident angle, and flux and facilitated the study of all three of the ion assistance schemes identified above. We find that the GMR ratio and coupling fields of a model spin valve structure can be significantly improved using these ion assistance concepts.

II. EXPERIMENTAL DEPOSITIONS

Giant magnetoresistive spin valves with a Ta/NiFe/Co/Cu/Co/FeMn/Cu structure were deposited by a biased target ion beam deposition approach using various

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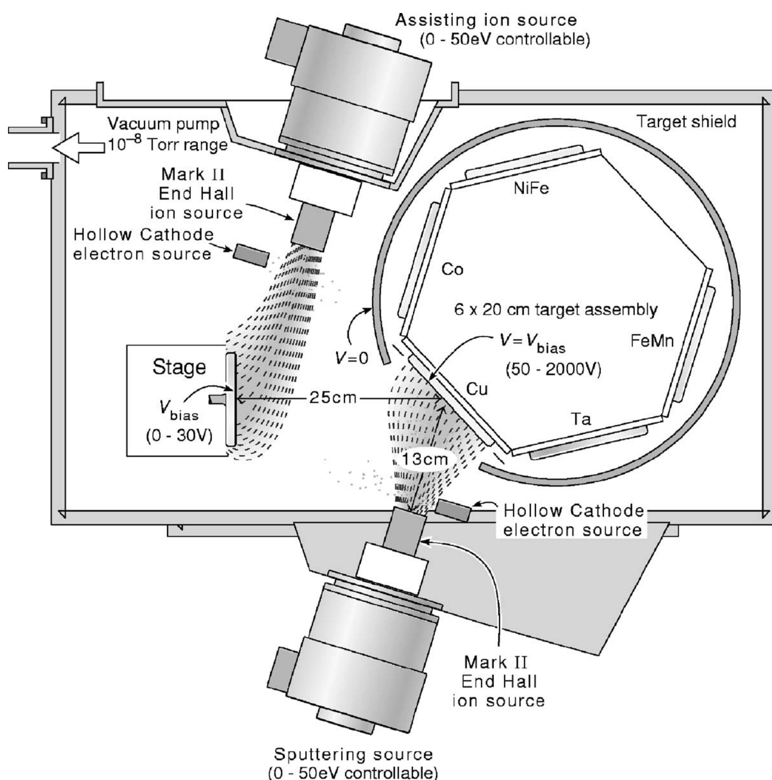

FIG. 1. Schematic illustration of a biased target ion beam deposition system. A high flux, low voltage ion source is used to soft sputter a metal target. A hollow cathode electron source is used to form a low energy ion flux for ion assistance during or after metal deposition. It enables the independent control of the ion energy, ion flux, and ion incident angle.

ion assisting strategies. The samples were grown at ambient temperature in the BTIBD system schematically illustrated in Fig. 1. All wafers were cut into 1×2 cm^2 sample size before deposition. Each of the metal layers of a prototypical spin valve structure was sequentially deposited without breaking vacuum. Magnetic field (around 60 Oe) was applied along with sample long edge while depositing layers of NiFe, Co, and FeMn. A rotatable target holder held the five metal sputtering targets. These were sequentially argon ion sputtered using a low voltage, high flux ion beam created using an end Hall ion source and secondary ionization by a low voltage electron beam. The ions were accelerated towards a target by applying a bias voltage to the target. This facilitated near normal incidence sputtering ion impacts, which greatly reduced the probability of high energy, reflected neutral fluxes[16,17] and the sample contamination that results from their impact with the interior surfaces of deposition equipment hardware.[15]

The ion source used for ion assistance was designed to obliquely illuminate the face of the substrate stage and enabled the production of low energy assisting ion flux in the 0–50 eV range. Details of the ion source characterization can be found in other papers.[16,18,19] For all of the depositions conducted here, the assisting ion flux obliquely impacted the sample surface at 45°. The assisting ion to metal atom flux ratio was 4 for all the experiments. The base pressure established prior to the deposition of all the films was about 3×10^{-8} Torr. The working pressure during growth was about 4.5×10^{-4} Torr. Sputtering was achieved using a target bias voltage of 300 V. The film thickness was controlled by deposition time based upon prior calibrations using x-ray fluorescence spectroscopy to determine film thickness (the deposition rate was 0.57 Å/s for Cu with a deviation of less than 3%). Magnetoresistance (MR) measurements of spin valves were carried out at room temperature with a standard four-point-probe method in an external magnetic field. A constant sensing current of 1 mA was used and the voltage drops were measured under the external field applied in the film plane. The MR measurements were repeated several times and all the results were reproducible for both GMR ratios (±3%) and the coupling fields (±2%).

III. RESULTS

A. Unassisted deposition

The effect of Cu spacer layer thickness (t) in a Ta (40 Å)/$Ni_{80}Fe_{20}$ (40 Å)/Co (15 Å)/Cu (t)/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin valve structure deposited without ion assistance was explored first. The GMR ratio is plotted as a function of copper spacer layer thickness in Fig. 2. It can be seen that the GMR ratio of devices grown in this manner is sensitive to the spacer layer thickness and is consistent with results for films grown by other techniques.[20,21] The best GMR ratio of 6.97% was obtained using a Cu layer thickness of 33 Å. It is also consistent with measurements of other groups who have shown that the maximum GMR ratio for this class of spin valve usually lies in the range of 6%–7%.[22–27] We note that the use of Ar for the sputtering gas rather than higher atomic mass sputtering ions such as Xe is thought to produce this good GMR ratio because of the reduced reflected neutral energy of near normal ion impacts and the low ion energy.[17,28] This BTIBD method therefore appears to provide a means for creating high quality spin valves using lighter sputtering gases.[17]

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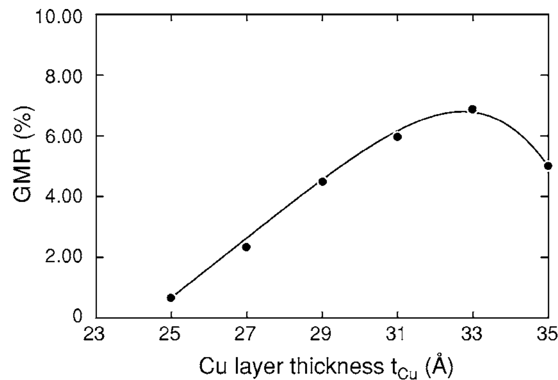

FIG. 2. GMR ratio as a function of Cu spacer layer thickness for Ta (40 Å)/$Ni_{80}Fe_{20}$ (40 Å)/Co (15 Å)/Cu (t)/Co (45 Å)/FeMn (100 Å)/Cu (20 Å) spin valve deposited without ion assistance.

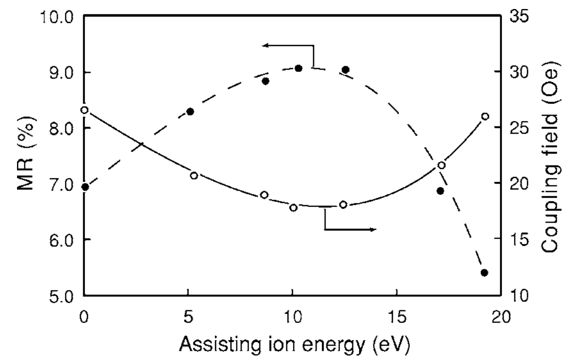

FIG. 3. GMR ratio and coupling field as a function of assisting argon ion energy. Simultaneous ion assistance was only applied during the deposition of the Co/Cu/Co part of the Ta/NiFe/Co/Cu/Co/FeMn/Cu spin valve.

B. Simultaneous assistance

Spin valves with a 33 Å spacer layer thickness were then deposited using argon ion assistance with ion energies that were varied up to 20 eV. This series of experiments utilized simultaneous ion assistance during deposition of the basic Co/Cu/Cu part of the spin valve structure described earlier. The other layers were deposited without ion assistance. The GMR ratio and coupling fields are plotted as a function of ion energy in Fig. 3. It can be seen that the use of an assisting ion energy of approximately 10 eV resulted in the highest GMR ratio and the lowest coupling field. This ion energy for best performance agrees well with the optimum energy predicted by earlier atomistic simulations for creating flat, unmixed interfaces and is consistent with some experiments.[10,14,19] The highest GMR measured ratio (9.06%) represented a significant increase over that of structures made without assistance (6.97%). It is also interesting to note that higher ion assistance energies reduced the GMR effect presumably due to the mixing that has been predicted to then occur at these interfaces.

C. Other strategies

In the simultaneous ion assistance scheme used above, both the ion and adatom (metal) fluxes arrive at the film growth surface simultaneously with a fixed ion/adatom flux ratio. The best ion energy for depositing the Co/Cu/Co trilayer was around 10 eV. Simulations have indicated that higher ion assistance energies give slightly better results if used once an interface is well covered by several monolayers of a new metal.[14] This overlay thickness is critical in both sequential and modulated energy ion assistance schemes because their use of high energy assisting ions can otherwise induce interfacial mixing.

Using insights gained from the atomistic modeling of the three ion assistance schemes,[14] a multiassistance strategy shown in Table I was developed for the deposition of the spin valve structure. In this proposed approach, the simultaneous ion assistance scheme was used for the deposition of the very thin Co and Cu layers (those with less than ten atomic layers as shown in Table I). This was chosen to avoid high ion energy induced interlayer mixing. A sequential ion assistance approach was used during the deposition of the relatively thick Ta, NiFe, and Cu layers (these are 10–20 atomic layers thick), and the modulated ion energy ion assistance scheme was used for the deposition of the thickest (FeMn) layer. The modulated ion energy ion assistance is preferred over the sequential ion assistance for the thickest layer because the maintenance of an assisting flux during the deposition of thick film can avoid island buildups from flux shadowing effect. Using this near optimal ion assistance scheme, Ta/NiFe/Co/Cu/Co/FeMn/Cu spin valve structures were grown and their measured GMR ratio was found to increase from 9.06% to 10.51%.

IV. DISCUSSION

Recent atomistic simulations have shown that the impact of assisting ions with a sample surface transfers energy from the assisting ions to surface adatoms. This enables athermal adatom hopping to more coordinated (local minimum en-

TABLE I. Ion assistance parameters for spin valve with 10.51% GMR ratio.

Layer	Thickness (Å)	Ion assistance scheme	Ion energy (eV)	Ion/adatom flux ratio	Ion flux (ions/Å^2 s)	Overlayer thickness (Å)
Ta	40	Sequential	20	⋯	0.18	40
NiFe	40	Sequential	20	⋯	0.18	40
Co	15	Simultaneous	10	4	⋯	0
Cu	33	Simultaneous	10	4	⋯	0
Co	45	Sequential	20	⋯	0.8	45
FeMn	100	Mdulated	20	4	⋯	30
Cu	20	Simultaneous	10	4	⋯	0

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ergy) sites on the surface and the growth of more atomically perfect structures. If ions are applied simultaneously with adatom condensation, a step flow mode of growth can be induced and the vertical growth of islands suppressed.[10,13] This minimizes the development of rough surfaces and ensuing flux shadowing for oblique components of the condensing atom flux. Intermixing occurs by atom exchange which is activated by the vertical component of the ions' momentum during an impact. The use of oblique assisting ion fluxes can enhance the surface flattening processes and reduces the probability of intermixing at interfaces formed as a new metal is deposited on an earlier grown surface. The most recent simulations indicate that argon ions with energies of around 8 eV and incidence angles near 50° result in near optimal interfaces for GMR applications.[13] The results obtained here appear to corroborate this hypothesis.

The simultaneous ion assistance scheme gradually becomes less effective as the layer thickness and thus average size of islands grows. More ion energy then needs to be transferred to the structure for the continuous value of flatness to be maintained. This becomes less problematic for interlayer mixing as the interface between the two layers becomes more deeply buried and therefore isolated from an ion impact. As a result, modulated ion energy and sequential ion assistance schemes are superior for thick films.[14,19]

The study conducted here indicates that the use of a low energy, simultaneous ion assistance scheme for very thin layers combined with higher ion energy modulated and sequential ion assistance schemes for the thicker ones can improve the interfacial structures of a model spin valve and lead to significant increases in GMR ratio. Biased target ion beam deposition appears to provide an effective means for the implementation of this strategy.

V. CONCLUSION

The effects of low energy, inert gas ion assistance during the deposition of a Ta/NiFe/Co/Cu/Co/FeMn/Cu spin valve structure have been experimentally explored. It was found that oblique, low energy, argon ion assistance with an energy around 10 eV during the deposition of the Co/Cu/Co trilayer part of the spin valve significantly improves spin valve properties. Atomistic simulations indicate that this results from improvements to the interfacial structures of these multilayer systems. By using modulated energy and sequential ion assistance for the other (thicker) layers of the spin valve structure, devices with more than 50% higher GMR ratios have been fabricated.

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