Atomic Assembly of Giant Magnetoresistive Multilayers

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ABSTRACT

The emergence of metal multilayers that exhibit giant magnetoresistance (GMR) has led to new magnetic field sensors, and approaches for nonvolatile random access memories. Controlling the atomic scale structure across the many interfaces within these multilayers is central to improve the performance of these devices. However, the ability to manipulate atomic arrangements at this scale requires an understanding of the mechanisms that control heterometal film growth during vapor deposition. It is important to develop methods that enable prediction of the effects of deposition conditions upon this structure. Atomistic simulation approaches have been combined with deposition reactor models to achieve this. We have applied these approaches to analyze the atomic scale structure of sputter deposited CoFe/Cu/CoFe giant magnetoresistive multilayers similar to those used for magnetic field sensing. Significant intermixing is revealed at the CoFe-on-Cu interface, but not at the Cu-on-CoFe interface. Recent experiments verified these predictions. The insights provide a basis for the development of processes that inhibit thermally activated atomic diffusion while allowing the controlled use of the metal atom impact energy and inert gas ions to manipulate the structure of interfaces.

BACKGROUND

About 13 years ago Fe/Cr/Fe multilayers with nanoscale layer thickness were discovered to exhibit very large (5-25%) drops in their electrical resistance when a moderate magnetic field was applied [1]. These “giant” magnetoresistive (GMR) materials soon became the subject of very intensive research because they appeared well suited for sensing small magnetic fields. Many multilayer systems composed of ferromagnetic metal layers separated by nonferromagnetic conductive metal layers have since been found to possess this property. The most frequently studied systems include Co/Cu/Cu [2,3] and NiFe(Co)/Cu/NiFe(Co) [4,5]. Rapid progress has resulted in the development of sensors for the readheads of hard disk drives. This contributed to the very large recent increases in the hard drive storage capacity [6]. These sensor applications are driving the development of materials that possess larger magnetoresistance. Many groups are now also exploring the application of this technology to create magnetic random access memory (MRAM) [6]. This MRAM appears to offer nonvolatility in addition to a similar functionality to state of the art dynamic random access memory. Two device approaches are being explored. One utilizes all metal multilayers and exploits electron spin dependent scattering. The second seeks to exploit changes in the tunneling conductance across dielectric barrier sandwiched between magnetic multilayers [6].

Both the sensor and MRAM applications of GMR materials are greatly improved by increasing the magnitude of the resistance change when a magnetic field is applied. This property is quantified by the GMR ratio (defined as the maximum resistance change divided by the resistance at magnetic saturation). In the metal multilayers, it has now been established that
when pairs of ferromagnetic layers are separated by a non-magnetic spacer layer of an appropriate thickness, the RKKY interaction causes their magnetic moments to align antiferromagnetically in a zero magnetic field\[7\]. Spin dependent electron scattering then results in a relatively high electrical resistance\[8,9\]. If a sufficiently large magnetic field is applied to align the magnetic moments of the ferromagnetic layers, the spin-dependent electron scattering (and electrical resistance of the film) can be reduced. Experiments have also indicated an extreme sensitivity of the magnitude of the resistance change to average spacing between the ferromagnetic layers\[2\]. High quality GMR multilayers must therefore have a precisely controlled layer thickness. In addition, Neel coupling can occur and greatly increases the coercivity of the device. This becomes increasingly significant as the layer’s roughness becomes comparable in amplitude and wavelength to the spacer layer’s thickness. Alloying (mixing) of one layer with the metal atoms of another also causes an increase in spin independent scattering\[8\] and a loss of local magnetic alignment\[10\], which can also reduce the size of the GMR effect. Many of today’s vapor deposited films appear to possess significant roughness and mixing. The development of processes that result in relatively smooth, unmixed interfaces is critical if the GMR ratios of multilayers are to be increased\[4,11\].

The atomic scale structure and compositional gradient across a metal multilayer are controlled by the atomic assembly processes active during their synthesis by vapor deposition. These processes are sensitive to the interatomic potentials between the various atoms and the availability of a means to facilitate assembly. In vapor deposition, assembly can be mediated by controlling the temperature of the surface and the deposition rate which in effect control the time available for a thermal fluctuation to induce an atom to hop from one site to another\[12\]. Hops can also be induced by other (nonthermal) processes such as hyperthermal metal impacts\[13,14\], inert gas ion assistance\[15\], and the heat of reaction when atoms approach\[13\].

GMR materials have been synthesized using numerous vapor deposition techniques including ambient temperature sputter\[4\], ion beam deposition\[16\], and molecular beam epitaxy (MBE)\[1\]. The experimental data indicate that multilayers deposited using sputter deposition and ion beam deposition (IBD) generally possess superior properties to those produced using MBE\[17\]. Figure 1 shows one contributing factor responsible for this observation. During the growth of an A/B/A multilayer stack, suppose the bond energy between A atoms is higher than that between B atoms. If atoms A and B are about the same size, then B can wet A because the more strongly bonded A atoms can pull B atoms onto their surface, Figure 1(a). However, A will then not wet B because the relatively weakly bonded B atoms cannot help reconstruct A clusters, Figure 1(b). This simple analysis indicates that during the deposition of an A/B/A multilayer system under thermal equilibrated conditions, the layer-by-layer growth mode can only be achieved for one of its interfaces. Even when the bond strength argument favors the layer-by-

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\[ \gamma_B + \gamma_{AB} < \gamma_A \]

\[ \theta \text{ defined by } \gamma_A \cos(\theta) + \gamma_{AB} = \gamma_B \]

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**Figure 1.** Thermodynamic analysis of multilayer growth modes.
layer growth, surface roughness could still occur to reduce the strain energy arising from the size difference between different elements. The experiments reviewed above suggest the use of growth conditions that constrain thermally assisted processes combined with “hot” metal atom and/or inert gas ion assistance is the preferred approach for creating GMR multilayers. The kinetically constrained conditions also limit interdiffusion across interfaces.

**DEPOSITION PROCESSES**

If the control of interfacial structures is achieved by energetic fluxes incident upon a growth surface, it is important that they be well understood. To explore this, RF diode sputtering and IBD processes are examined. Figures 2(a) and 2(b) schematically illustrate both processes. In a RF diode sputtering system, Figure 2(a), an inert gas (e.g., Xe or Ar) plasma is maintained between a target and a substrate by a RF power. A sheath potential (typically around 1000 eV) is established near the target. Inert gas ions created in the plasma are accelerated across the sheath, striking the target surface at a near normal incident angle. These ions can have energies between a hundred and a thousand electron volts. Our MD simulations show that such impacts result in the sputtering of metal atoms from the target. These sputtered atoms have a wide distribution of energies with an average in the 5-10 eV range and a tail extending to above 30 eV \[18\]. They emit with a near cosine angular distribution. The sputtered metal atoms are then transported to the substrate and are deposited on the substrate surface. Diode deposition occurs at relatively high pressure (~20 mTorr) and even though the distance of propagation is usually short (a few centimeters), numerous collisions with working gas atoms occur during metal atom transport. Due to these collisions, the energy of the metal atoms at the substrate is generally much lower than the energy of the sputtered atoms at the target. Reactor scale simulations indicate that the metal atom energy are in the 0.1-1.0 eV range \[19\]. In practice, the adatom energy can be reduced by reducing RF power, or by increasing the chamber pressure or target-substrate distance \[19\]. Magnetron sputtering results in higher energy metal fluxes because of the lower pressure.

A second sheath potential also exists near the substrate. This sheath’s potential can range between 50 and 200 eV. This results in a fraction of the plasma inert gas ions impacting the substrate at normal incidence with energies of 50-200 eV \[19\]. The energy is again controlled by the plasma power and the background pressure. Under some conditions, the relative flux of these ions can be high (an ion to metal flux ratio above 0.5) \[19\]. Recent experiments have linked these fluxes to strong variations in the magnetoresistance of metal multilayers \[20\].

*Figure 2.* Schematic illustrations of RF diode sputtering and ion beam deposition.
An ion beam deposition system, Figure 2(b), uses an ion beam gun to generate high energy (>600 eV) inert gas ions to sputter the target. The energy of sputtered atoms scales with the ion beam gun voltage and again with an average in the 5-10 eV range and a tail extending to above 30 eV \[18\]. However, because no plasma needs to be maintained in the deposition chamber, the process can occur at a much lower pressure. This reduces the background scattering as the sputtered atoms are transported to the substrate. The energy of the atoms that strike the substrate is therefore high. Some energy control is possible by changing the ion beam gun voltage.

Typically the inert gas ions strike the target at an incident angle near 45°. At the ion incident energy of 600 eV and the incident angle of 45°, a significant fraction of ions can be reflected as neutrals from the target surface. The reflected neutrals have energies ranging between 50 and 200 eV, and are emitted in an angular distribution that enables many to reach the substrate surface \[18\]. A secondary (assisting) ion beam gun can be used to irradiate the growth surface with inert gas ions to help flattening the surface. This flux can be several times of that of the metals.

From the analyses above, it is clear that a wide range of atom/ion energies can be encountered during these deposition processes. We have used atomistic simulation approaches to understand the effects of adatom energy as well as low and high energy inert gas impacts on the interfacial roughness and interlayer mixing of multilayers.

**ATOMISTIC MODELING METHODS**

To realistically predict the effects of the many factors affecting the multilayer growth, a highly predictive atomistic simulation method must be used. In our work, a molecular dynamics (MD) approach is used to simulate the atom/ion interactions, including the transition of atoms from vapor to a solid surface and the subsequent surface reconstruction. Because Newton’s equations of motion are used to describe the evolution of atomic positions, the detailed atomic structures of a deposited film and many of the mechanisms active throughout the entire deposition process can be correctly described, but only when a high fidelity interatomic potential is used. We have adapted the embedded atom method (EAM) model initially developed by Daw and Baskes \[21\] which assumes that the crystal energy is the sum of a pairwise potential and an energy required to embed an atom into a local medium with a given electron density. Because this embedding energy depends on the net local environment, EAM models provide a good format for solving problems involving surfaces and other defect states.

EAM potentials developed for atoms of a single element cannot generally be applied for alloys. We have recently proposed a generalized EAM potential database that enables the calculations of alloys with any combination of sixteen metals (Cu, Ag, Au, Ni, Pd, Pt, Al, Pb, Fe, Mo, Ta, W, Mg, Co, Ti, and Zr). These potentials are well fitted to basic material properties such as lattice constants, elastic constants, bulk moduli, vacancy formation energies, sublimation energies, and heats of solution. The derivation of the integrated EAM potentials for a variety of elements and their alloys has been described previously \[22\], and only pertinent results are reported here. In EAM, the total energy \(E\) of the crystal can be expressed as

\[
E = \frac{1}{2} \sum_{i,j \neq j} \phi(r_{ij}) + \sum_i F(\rho_i) \quad (1)
\]
where $\phi(r_{ij})$ represents the pair energy between atoms i and j separated by $r_{ij}$, and $F(\rho_i)$ stands for the energy to embed an atom i into a local site with electron density $\rho_i$. The electron density $\rho_i$ is calculated using

$$\rho_i = \sum_{j, j \neq i} f(r_{ij})$$

with $f(r_{ij})$ the electron density at the site of atom i arising from atom j.

Alloy EAM potentials can be constructed from elemental EAM potentials if the potentials are normalized [23] and unified cutoff functions are used. To fit such an EAM potential set, the generalized pair potential for a given element was chosen to have the form

$$\phi(r) = \frac{A \cdot \exp \left[-\alpha \left(\frac{r}{r_e} - 1\right)\right]}{1 + \left(\frac{r}{r_e} - \kappa\right)^2} - \frac{B \cdot \exp \left[-\beta \left(\frac{r}{r_e} - 1\right)\right]}{1 + \left(\frac{r}{r_e} - \lambda\right)^2}$$

with $r_e$ the equilibrium spacing between nearest neighbors, $A$, $B$, $\alpha$, $\beta$ are four adjustable parameters, and $\kappa$, $\lambda$ are two additional parameters for the cut off.

The electron density function is taken with the same form as the attractive term in the pair potential with the same values of $\beta$, and $\lambda$:

$$f(r) = f_e \cdot \exp \left[-\beta \left(\frac{r}{r_e} - 1\right)\right]$$

$$1 + \left(\frac{r}{r_e} - \lambda\right)^2$$

Using the alloy model [23], the pair potential between different species a and b is constructed as

$$\phi^{ab}(r) = \frac{1}{2} \left(\frac{f^b(r)}{f^a(r)} \phi^{aa}(r) + \frac{f^a(r)}{f^b(r)} \phi^{bb}(r)\right)$$

To have embedding energy functions that can work well over a wide range of electron density, we have used three equations to separately fit to different electron density ranges. For a smooth variation of the embedding energy, these equations are required to match values and slopes at their junctions. These equations are listed in the following:

$$F(\rho) = \sum_{i=0}^{3} F_m \left(\frac{\rho - \rho_n}{\rho_n - 1}\right)^i, \quad \rho < \rho_n, \quad \rho_n = 0.85 \rho_e$$
\[
F(\rho) = \sum_{i=0}^{3} F_i \left( \frac{\rho}{\rho_e} - 1 \right)^i, \quad \rho_n \leq \rho < \rho_o, \quad \rho_o = 1.15\rho_e
\]  
(7)

\[
F(\rho) = F_e \left[ 1 - \ln \left( \frac{\rho}{\rho_e} \right)^\eta \right], \quad \rho_o \leq \rho
\]  
(8)

With this model, the parameters needed to define 16 metals are listed in Table I.

The interactions between inert gas ions and metal surfaces can be described by a pairwise universal potential [24]. The pairwise universal potential was fitted to experimental data for the ion bombardment of a solid surface, and hence well represents the interaction forces between metal atoms of a solid surface and the energetic inert gas atoms or ions that strike it. Details of our MD simulation can be found in ref. [25]. A Lagrangian form of Newton’s equation of motion was used in the MD model to account for relaxation of the crystal size under the influence of the lattice mismatch between different layers [26].

**ATOMIC SCALE STRUCTURES**

The simulation methodology has been used to simulate the growth of magnetron sputter deposited multilayers for which atomic scale structure data existed [27]. The growth of a 58ÅCo90Fe10/15ÅCu/40ÅCo90Fe10 multilayer unit on a 10ÅNi80Fe20 substrate was simulated using an adatom energy of 3.0 eV, a normal adatom incident angle, a substrate temperature of 300 K and a deposition rate of 1nm/ns. The detailed atomic structure of the simulated multilayers is shown in Figure 3 (the NiFe substrate is not shown). The Fe, Co and Cu atoms are marked black, gray, and white respectively. Interestingly, it can be seen that the Cu-on-CoFe interface is relatively sharp, whereas the CoFe-on-Cu interface is much more diffuse. Also, a high fraction of Cu atoms were seen to be mixed in the CoFe layer. This phenomenon is more significant across the CoFe-on-Cu interface than the Cu-on-CoFe interface, indicating that the most of these Cu atoms were incorporated in the subsequently deposited CoFe layer. A direct visualization of the atomic scale structures of this multilayer stack deposited using similar growth conditions by a three-dimensional atom probe approach have confirmed the mixing of Cu in the over deposited ferromagnetic layer and the sharp nature of the Cu-on-CoFe layer [27].

The composition at a given depth in the multilayer was calculated as the atomic fraction of Cu, Co and Fe atoms in a small box placed at that position. Composition vs. depth profiles was mapped out by continuously moving the box along the growth direction. The calculated composition profiles for Co, Cu and Fe are shown in Figure 4 with solid, dash and bold lines respectively. Figure 4 more graphically indicates that the Cu-on-CoFe interface is much sharper than the CoFe-on-Cu interface. To further explore the relative distribution of Co and Fe, the Co/Fe ratio was calculated along the multilayer film thickness. In this calculation, we ignored the regions where Fe concentration is too low to yield accurate Co/Fe ratios. The results of this calculation are included in Figure 4 as a shaded region. Compared to a nominal Co/Fe ratio of 0.9, Figure 4 appears to show a short-range region of abnormally higher Co/Fe ratio at the CoFe-on-Cu interface indicative of Fe depletion near the interface. Similar results have been found when we used typical IBD growth conditions [18].
Table I: EAM parameters

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O4.1.7
ATOMIC ASSEMBLY MECHANISMS

The simulations above use a metal atom kinetic energy of 3.0 eV. A wide range of energies can be accessed. To explore the effects of adatom energy, MD simulations were used to examine individual Cu adatom impacts with a surface asperity on a (111) Cu surface. The change of the asperity during a low (0.1 eV) and a high (5.0 eV) energy atom impact is shown in Figure 5, where the adatom is marked with “A”. It can be seen that at the low energy, the adatom promoted surface roughness by attaching to the sidewall of the asperity. The higher energy impact resulted in momentum transfer to the atoms in the asperity and subsequent rapid rearrangement on the surface. This resulted in a local flattening effect. This flattening occurred within a few picoseconds, which is generally much faster than the thermally activated processes. Hot (hyperthermal energy) metal atom deposition under thermally constrained conditions provide potent route for avoiding interfacial roughness.

However, energetic atom impacts can cause other phenomena. Figure 6 shows MD results for a high-energy Ni adatom (marked with “A”) impact with a flat (111) Cu surface. It can be seen that during this impact, the Ni atom exchanged with a surface copper atom. This occurred when the Ni adatom penetrated the Cu lattice sufficiently that it was embedded in the Cu lattice and ejected a Cu atom on to the top of the surface. The atom exchange probability has been found to depend on many factors including the adatom and surface composition, the defect structure of the surface and the energy and angle of the impact [25,28]. These results have shown that impact atom induced exchange is the cause of significant mixing during the low temperature

Figure 5. Impact induced flattening mechanism occurs at 5.0 eV but not 0.1 eV.
deposition condition used for GMR multilayer growth. The exchange probability is lower on a perfect surface than on a defective (rough) surface, and increases with increasing impact energy. At a given energy and surface roughness, the exchange probability is much higher when a Co or an Fe atom impacts a Cu surface than when a Cu atom impacts a CoFe surface. This arises because surface segregation of Cu in a CoFe film is energetically favored. This finding accounts well for the observations in Figures 3 and 4 that in sputter deposited CoFe/Cu/CoFe multilayers, the Cu-on-CoFe is relatively sharp while the CoFe-on-Cu interface is more diffuse, and Cu mixing occurred preferentially in the subsequently deposited CoFe layer.

Simulations of the type described above also revealed why the Fe concentration is lower than the nominal value at the CoFe-on-Cu interface. Detailed analyses have indicated that during the deposition of the CoFe layer on top of a given Cu plane, a deposited Co atom more frequently exchanged with a Cu atom in that plane than a deposited Fe atom. As a result, a larger fraction of deposited Co atoms than deposited Fe atoms will occupy that plane. A short-range Fe depletion (with respect to Co) region is then created at the Cu side near the CoFe-on-Cu interface.

**METAL ATOM ENERGY CONTROL**

The MD simulations have provided compelling evidence that increasing the metal atom kinetic energy helps flatten interfaces but also induces interlayer mixing at interfaces, especially when the underlying material has a strong tendency to segregate to the surface of the material being deposited. While lowering the adatom energy can sharpen the chemical boundary of the interface, it is often accompanied by an increase of the interfacial roughness. As a result, better GMR properties are obtained from materials grown at an intermediate adatom energy \[^{[4,16]}\]. The identification of the asymmetric interfacial structures and their formation mechanisms suggests that the use of different adatom energies to deposit the Cu and the CoFe layers may be more beneficial than the use of a single “optimum” energy for deposition of all layers. In particular, the use of a lower adatom energy for deposition of the CoFe layer on Cu than for the Cu on CoFe (interlayer energy modulation) can effectively sharpen the CoFe-on-Cu interface. This strategy has been experimentally implemented and found to improve the GMR ratio \[^{[29]}\]. Even better films are anticipated by modulating the energy during deposition of each layer (intralayer energy modulation). To investigate this idea, a simulation was conducted where the first few monolayers of Cu (or CoFe) were deposited on CoFe (or Cu) using a low adatom energy to avoid impact induced mixing. After the interface was well buried below the surface, higher adatom energy was used to flatten the surface. This modulated energy scheme and the simulated result for the 58ÅCo\(_{90}\)Fe\(_{10}\)/15ÅCu/40ÅCo\(_{90}\)Fe\(_{10}\) multilayer on a 10ÅNi\(_{80}\)Fe\(_{20}\) substrate is shown in Figure 7. A
composition profile was also calculated and is shown in Figure 8. Compared to Figures 3 and 4, it can be seen that the interlayer mixing is greatly reduced while the interfacial roughness remains almost unchanged. The interfaces are seen to be very sharp, and the Fe depletion at the CoFe-on-Cu interface due to the exchange mechanism is also removed.

**ENERGETIC ION EFFECTS**

Low energy ion assisted deposition may provide additional ways to control the morphology of growth surface and the interfacial structures of multilayers. In sputtering, this can be achieved by applying a substrate bias voltage to induce low energy inert gas ion impacts with the substrate surface. In IBD, a secondary ion beam gun can be used to inject low energy inert gas ions directly to the substrate surface. Here, MD was used to analyze the effects of individual low energy inert gas ion impacts with a pre-designated rough surface. To exemplify, we show the impact of Xe ions with a model Ni/Cu/Ni multilayer surface.

During the growth of Ni/Cu/Ni multilayers, the deposition of Cu on Cu, Cu on Ni, Ni on Ni, and Ni on Cu are all encountered. To explore the effects of inert ion impact on the reconstruction of a “rough” surface, the depositing Cu or Ni atoms were assumed to form arrays of hillocks on the underlying surface. An example of the Ni-on-Cu surface can be seen in Figure 9, where Ni and Cu atoms are marked with dark and light balls respectively. The inert ion impact was simulated by injecting a Xe ion toward the surface from random locations far above. The change of surface structures following the impact was calculated. After the thermal spike induced by the ion impact was dissipated, another ion was injected to repeat the same process until a total ion fluence of about 0.5 ions/Å² was reached. All simulations were carried out at a fixed substrate temperature of 300 K, and a normal ion incident angle [15].

When Cu clusters existed on either Ni or Cu crystals, Xe ion impacts at a low ion incident energy of 3.0 eV caused the Cu asperities to achieve single monolayer high configurations. Clearly, the flattening of Cu surface due to inert gas ion impacts is significant even at a very low ion energy of 3.0 eV. However, no complete flattening of the Ni asperities on either Ni or Cu crystals was observed during 3.0 eV Xe ion impacts. An increase in the Xe ion energy (to 9.0 eV) was found to flatten the Ni-on-Ni surface. However, the exchange induced by high-energy

![Figure 7. CoFe/Cu/CoFe multilayers deposited by energy modulation.](image)

![Figure 8. Composition profiles for the CoFe/Cu/CoFe multilayers deposited by energy modulation.](image)
Xe impacts with the Ni asperities on a Cu surface resulted in significant interlayer chemical mixing. To illustrate, the effects of the Xe ion impacts on the Ni-on-Cu are shown in Figure 9. It can be seen that no Ni asperities were reduced to single monolayers high islands after about 0.5 ions/Å² impacts of Xe at an ion energy of 3.0 eV. Increasing the Xe ion energy to 12.0 eV caused a significant flattening of the Ni-on-Cu surface, but extensive mixing between Cu and Ni atoms occurred. Rupp and Schuster \[30\] applied a substrate bias voltage to induce low energy inert gas ion impacts with a growth surface in their sputtering system. They deposited [10ÅCo/10ÅCu]₄₇ multilayers at various bias voltages, and measured the GMR ratios. Their results indicate that a substrate bias voltage as low as 20 eV significantly reduced the GMR ratio of the material. It is clear that an ion energy as low as about 20 eV is detrimental.

The observation that the ion energy to flatten the Ni surface asperities was higher than that to flatten the Cu surface asperities can be understood in terms of the mobility or bond strength of surface atoms. The local surface asperities formed during random deposition of atoms on the surface are less likely to be reconstructed into local flat single monolayer high island configurations if these atoms are strongly bonded and are less mobile. Because Ni has a higher cohesive energy and higher activation energies of diffusion than Cu, it requires higher energy impacts to break the Ni-Ni bonds. The ion energy required to flatten the Ni surface asperities is therefore higher than to flatten the Cu surface asperities. Generally, higher surface roughness is usually observed during deposition of materials with higher cohesive energy or higher activation energies of diffusion.

The results discussed above indicated that the most difficult scenario during low energy ion assisted deposition of Ni/Cu/Ni multilayers is the growth of the Ni-on-Cu surface. On this surface, the assisting ion energy required for the flattening is well above the threshold energy of mixing. The mixing preferentially occurs when the underlying metal is Cu because Cu tends to segregate to the surface and the relatively weakly bonded Cu can be easily penetrated during energetic impacts to cause the exchange. Another way to improve the deposition is to use modulated ion assistance in which the first few monolayers of a new material layer is deposited without ion assistance and the remainder of that layer is deposited with ion assistance. The results reviewed above provide guidelines for implementing a modulated ion assistance scheme. During depositing the Cu-on-Ni surface, for instance, the maintenance of a low ion energy and the modulation of ion assistance are not necessary because the probability for the mixing between surface Cu and underlying Ni is low for a wide range of ion energies. However, when depositing the Ni-on-Cu surface, the ion energy must be kept at very low values, and often it is necessary to use modulated ion assistance to achieve surface flattening without causing mixing between Ni and the underlying Cu.

![Figure 9](image-url). Effects of low energy assisting inert gas ion impacts.
One significant finding from MD simulation of ion assisted deposition of GMR multilayers is that only a very low ion assisting energy (several eV to about 10 eV) can be used to improve the flatness of a growth surface without causing mixing between surface and subsurface atoms. This energy sensitively depends on the surface and subsurface materials. The ion energy needed to flatten the surface increases with an increasing cohesive energy for the surface element. On the other hand, the ion energy that can cause significant mixing decreases as the cohesive energy of the underlying element decreases.

As described earlier, inert gas ions or neutrals with energies between 50 and 200 eV can impact the growth surface in RF diode sputtering or IBD deposition systems. High-energy impacts may cause atomic exchanges, resulting interlayer chemical mixing. The MD technique has been applied to the Ni/Cu/Ni multilayer system to calculate the exchange probability of atoms between any two adjacent atomic layers below the surface on which impacts occur \cite{31}. The results indicated that exchange probability rapidly decreases as the impact energy is decreased and the depth of the two atomic layers from the surface is increased. At 200 eV impact energy, however, significant exchange probabilities exist even when the interface is buried seven monolayers below the surface. The exchange probability on the Ni-on-Cu surface is higher than that on the Cu-on-Ni surface- a result to be anticipated because of the high tendency for surface segregation of the underlying Cu.

A reduced order model has been developed to simulate Xe impact induced exchanges between all adjacent atomic layers during deposition of Ni/Cu/Ni multilayers \cite{31}. Results for Cu composition profiles at two Xe energies and a fixed Xe:metal ratio of 0.8 are shown in Figure 10(a). The Cu composition profiles at two Xe:metal ratios and a fixed Xe energy of 200 eV are shown in Figure 10(b). It can be seen that as the Xe energy or Xe:metal ratio is increased, the interfaces become diffused. Because the exchange on the Ni-on-Cu surface is more significant than on the Cu-on-Ni surface, the Ni-on-Cu interface is more diffuse. At high Xe energy or Xe:metal ratio, Cu is seen to distribute into the entire Ni layers as no region with zero Cu content exists. On the other hand, Ni does not distribute into the entire Cu layers since there are still regions with a Cu concentration of 1. Another interesting finding in Figure 10 is that at high Xe energy or Xe:metal ratio, it is possible to build up a Cu surface segregation during the deposition of Ni.

**Figure 10.** Composition files across the multilayer thickness.
NEW PROCESSES FOR CONTROLLING ATOMIC ASSEMBLY

The results above indicate that to reduce the interlayer mixing and to sharpen the interfaces, it is essential to reduce both energy and flux of the high-energy inert gas ions or neutrals during the deposition of GMR multilayers. Processes that allow metal energies of a few (1-2 eV) are also desired. The ability to ultimately modulate the metal atom energy during the deposition may also provide a better opportunity to control atomic assembly in metal systems. A step in this direction has been proposed by Hylton et al \cite{32,33}. They have used a target bias voltage in combination with low voltage electron ionization of a dense inert gas beam to create a high flux of low energy ions for target sputtering. The resulting normal ion incidence reduces the flux and energy of reflected neutrals. It also reduces overshoot contamination, and facilitates the control of adatom energy. Preliminary results from this biased target ion beam deposition (BTIBD) process have resulted in an increase in the GMR ratio by at least 12.5% (compared to conventional IBD) for a NiFe/CoFe/Cu/CoFe/NiFe multilayer system. Other approaches may also be possible. For example, the use of low voltage electrons or strong RF fields to ionize metal atoms that have been thermally evaporated when combined with electrostatic biasing might also provide an improved means for energy control. Finally, we note all these efforts aim to control atomic hopping. Recently, it has become increasingly evident that the activation barriers themselves are amenable to some manipulation through the use of surfactants \cite{34}.

CONCLUSIONS

It has been shown that sophisticated atomistic simulation approaches can greatly help the design of new deposition tools that facilitate the control of atomic assembly of metal multilayers. They enable the interplay between thermal activation of mechanisms of assembly and those induced by physical (i.e., energetic atom/ion) or chemical processes to be quantifiably assessed. The emergence of this multiscale modeling capability promises to open up many opportunities for designing processes for growing other similarly exacting atomic scale structures.

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