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The role of boron on exchange coupling in NiFe/Ru$_{1-x}$B$_x$/FeCo trilayer structures

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ABSTRACT
In this work, we study the interlayer exchange coupling, J, between two NiFe and FeCo layers in a series of NiFe/Ru$_{1-x}$B$_x$(d)/FeCo synthetic antiferromagnet (SAF) samples, where the thickness of the spacer layer, d, is varied from 0.4 nm to 0.9 nm, and the boron concentration, x, is varied from 0 to 15 at. %. The samples are studied as deposited and after being annealed at 250°C. B is deposited into the Ru spacer layer to investigate what occurs after annealing a FeCoB/Ru/FeCoB SAF structure, which is commonly used in modern nanoscale magnetic devices, in which the FeCoB layer crystallizes to FeCo and B diffuses to adjacent layers. We find that J in as-deposited samples is relatively unaffected by adding up to 15% B into the Ru spacer layer. However, after annealing at 250°C, J changes the sign from antiferromagnetic coupling for spacer layers thinner than 0.45 nm for 5% and 10% B and thinner than 0.525 nm for 15% B. We used transmission electron microscopy energy-dispersive x-ray spectroscopy in order to investigate the diffusion of atoms within a similar Ta(2.5 nm)/NiFe(0.8 nm)/Ru$_{1-x}$B$_x$(23 nm) layer structure. We find that after annealing at 250°C, the sample containing 15% B within the Ru$_{85}$B$_{15}$ layer had significantly more diffusion of Fe into the Ru$_{85}$B$_{15}$ layer, from the NiFe layer, as compared to the sample with 0% B. Thus, the presence of B within the spacer layer enhances diffusion of Fe into the spacer layer.

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I. INTRODUCTION
Spin transfer torque magnetic random access memory (STT-MRAM) has attracted a large amount of research interest recently due to the promise that it will be far superior to the current generation nonvolatile solid state memory, which is found in a large number of electronic devices. Specifically, STT-MRAM is expected to be faster, having 2–30 ns write time latency¹ compared to 200 μs for flash,² have close to infinite endurance, exceeding 10¹⁵ write cycles³ compared with 10⁵ for flash,⁴ and have high memory density.⁵ All of this while being relatively easily integrated into the CMOS process.⁶,⁷

The memory element of an STT-MRAM device commonly contains an FeCoB/MgO/FeCoB tunnel junction. This layer structure is preferred due to the high perpendicular magnetic anisotropy (PMA) at the FeCoB/MgO interfaces and high tunneling magnetoresistance (TMR) through this junction.⁸–¹⁰ Binary data are stored in the direction of magnetization of one of the FeCoB layers, known as the free layer. The other FeCoB layer is used as a reference, known as the fixed layer. Data are read using a change of resistance caused by TMR¹⁰ and written using spin transfer torque.¹¹ This functionality requires that the direction of magnetization within the fixed layer remains constant under all circumstances, which is typically achieved by coupling two magnetic layers antiferromagnetically (AFC) across a nonmagnetic spacer layer to create a synthetic antiferromagnet (SAF). In comparison to a single ferromagnetic film, a SAF requires a larger external field or polarized currents for magnetization reversal, has higher thermal stability, and has reduced stray fields.¹²–¹⁷

However, the nature of coupling between FeCoB layers across a Mo or Ru spacer layer less than 0.6 nm in thickness, for example, changes from AFC to ferromagnetic coupling (FC) when annealed above 200°C.¹⁸ Annealing is a required step to achieve the high PMA and TMR of the tunnel junction, which cannot be avoided, making it difficult to create a stable fixed layer.¹⁸,¹⁹,²⁰ This change of coupling from AFC to FC upon annealing was not observed in structures having ferromagnetic layers without the presence of boron.

In this article, we investigate why coupling within the related NiFe/Ru/FeCoB SAF fixed layer changes from AFC to FC after...
annaling above 200 °C, and we will study the role of boron in causing this change. The first possibility is that annealing the sample causes boron to diffuse from the magnetic layer into the Ru spacer layer, which creates a RuB alloy, and that this alloy exhibits FC. In order to test whether this is the case, we have created samples where boron is only contained within the spacer layer. This allows us to isolate and study the scenario where boron has diffused into the spacer layer after annealing.

Another possible cause of the change of coupling from AFC to FC after annealing above 200 °C is that the presence of boron enhances diffusion of magnetic materials into the Ru spacer during the annealing step. It has been shown previously that a high enough concentration of ferromagnetic atoms within the Ru spacer layer can result in FC.21

II. EXPERIMENTAL

We created a series of samples (SERIES-A) with different concentrations of boron within the Ru spacer layer to see what effect it has on the coupling. The exact structure that was sputtered is Ta(2.5 nm)/NiFe(6 nm)/Ru1−xBx(d)/FeCo(4 nm)/Ta(4 nm), where the spacer layer thickness d is varied from 0.4 to 0.9 nm, and the B concentration within the spacer layer x is varied from 0 to 0.15 in increments of 0.05, as is shown in Fig. 1. The elemental compositions of the two ferromagnetic alloys are Fe75Co25 and Ni80Fe20. We have chosen NiFe and FeCo because their vastly different saturation magnetization (Ms) causes their ferromagnetic resonance (FMR) absorption lines to be well separated, which is required in order to measure the coupling strength using our FMR technique.18

The samples are deposited by means of RF magnetron sputtering on oxidized Si substrates. The deposition conditions, sample preparation, and rapid thermal annealing (RTA) procedure are explained in detail in our previous work.22 The demagnetizing dipolar fields in the two magnetic layers are much larger than any fields perpendicular to the film that arise from the surface and magnetocrystalline anisotropies, forcing the magnetization to lie in-plane. The samples are polycrystalline and rotated during deposition, resulting in the in-plane magnetocrystalline anisotropy being averaged out causing the easy-plane of the magnetic anisotropy being in the plane of the sample. The samples are also capped with 4 nm of Ta to protect the top FeCo layer from further oxidation or water absorption from the atmosphere. After deposition, the samples are cut into two pieces. One piece is left as is and the other is rapidly thermally annealed (RTA) in vacuum at 250 °C and 450 °C.

In order to investigate whether having boron within the Ru spacer layer causes increased diffusion of atoms from the adjacent magnetic layers into the Ru layer, we sputtered a second series of samples (SERIES-B) to be measured with energy-dispersive x-ray spectroscopy (EDXS). The structures of these samples are Ta(2.5 nm)/NiFe(0.8 nm)/Ru1−xBx(23 nm), where x is varied from 0 to 0.2 in increments of 0.05. These SERIES-B samples were then cut into three different pieces. One of them was annealed at 250 °C, the second was annealed at 450 °C, and the third one was left as is.

These samples are sputtered using the same deposition conditions and annealing procedure as SERIES-A samples.

Spectrum imaging analysis based on EDXS was performed in scanning transmission electron microscopy (STEM) mode. Prior to EDXS analysis, TEM lamellae were prepared using a Ga-focused ion beam (FIB). To minimize sidewall damage, Ga ions with only 5 keV energy were used for final thinning of the TEM lamella to electron transparency.

FMR was used to measure the coupling strength in the SERIES-A samples. FMR measurements were carried out at room temperature with a coplanar waveguide in an in-plane field-swept, field modulated configuration, as detailed in Ref. 23 over a frequency range of 8–36 GHz. The directions of the DC magnetic field, μ0H, where μ0 = 4π × 10−7 H/m, and RF magnetic field, μ0hf, used in FMR are both in the plane of the sample and orthogonal with respect to each other as shown in Fig. 1. In all measurements, μ0H is large enough to saturate magnetization in the samples. Typical raw FMR data are shown in Fig. 2, along with the fit using our model.

FIG. 1. The thin film structure of the studied samples. The numbers in parentheses indicate the layer thicknesses in nm, where d is varied from 0.4 nm to 0.9 nm, and x represents the fraction of boron in the ruthenium alloy, which is varied from 0 to 0.15.

FIG. 2. Example of typical raw FMR data, orange solid circles, along with the fit, blue solid line, using the model detailed in Ref. 22. These data are for an as-deposited SERIES-A sample with 15% boron in the 0.55 nm thick spacer layer.
The model that we use to analyze the FMR data is explained in detail in our previous work. However, this FMR technique can only measure weak coupling strengths of approximately $\pm 0.8 \text{ mJ/m}^2$. For samples with AFC strengths beyond those limits, we characterized using a vibrating sample magnetometer (VSM). In this case, magnetization as a function of field $M(H)$ is acquired in a field sweep from 7 to $-7 \text{ T}$ with a magnetic field applied parallel to the film surface. In the presence of the magnetic field, AFC coupled layers form spin spirals that are simulated by the micromagnetic model proposed by Eyrich et al.

SERIES-B samples were also characterized using x-ray diffraction (XRD) in order to determine the effect that adding B has on the Ru crystal lattice. XRD measurements were performed using Cu K$_\alpha$ radiation with the scattering wave vector both normal to the film surface (perpendicular) and in the plane of the sample (in-plane).

III. RESULTS AND DISCUSSION

XRD patterns for two SERIES-B samples, one with 0% B, and one with 15% B, both annealed at 250°C are shown in Fig. 3. The XRD measurements are much more sensitive to the top Ru and RuB layers rather than the lower Ta and NiFe layers due to the order of magnitude difference in thickness between them. Thus, the dominant peaks in the XRD patterns are that of Ru and RuB.

With 0% B, the Ru layer is very well textured along the [0002] direction. As we add more B to the Ru layer, it becomes less and less textured. In samples with 15% B, a small in-plane (1-101) peak is visible in the in-plane XRD pattern, as shown in Fig. 3. The small size of this peak indicates that the RuB layer is still mostly textured along the [0002] direction, but not as well textured as the Ru layer in the 0% B sample was. This [0002] texture for the RuB layer is desired to achieve a [110] textured top FeCo layer, resulting in narrow FMR absorption lines, as shown in Fig. 2, allowing us to more easily apply our FMR characterization technique.

Measurements of the magnetic coupling strength $J$ of the as-deposited SERIES-A samples, with B concentrations within the RuB alloy of 0%, 5%, 10%, and 15%, are shown in Fig. 4. It can also be seen that as the percentage of boron is increased, there is a slight decrease in coupling strength and a shift of the coupling strength oscillations. These coupling strength oscillations are a characteristic indicator of Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling, which has been shown to be dependent on the topological properties of the spacer layer Fermi surface. Thus, these changes in oscillatory behavior are likely the result of changes to the Fermi surface of the spacer layer caused by the increased B concentration. Similar effects have been seen when adding different concentrations of Ni into a Cu spacer layer.

In Fig. 4, it can be seen that the presence of B within the spacer layer does not cause the sign of coupling to change from AFC to FC for spacer layer thicknesses between 0.4 and 0.9 nm. This indicates that for FeCoB/Ru/FeCoB SAF structures annealed above 200°C, the change in sign of coupling from FC to AFC for spacer layer thicknesses less than 0.6 nm is not the result of B diffusing into the Ru spacer layer alone. Instead, the annealing step must have some effect other than B diffusion into the Ru spacer layer that causes the FC region for spacer layer thicknesses less than 0.6 nm.

Measurements of the magnetic coupling strength $J$ of the SERIES-A samples annealed at 250°C are shown in Fig. 5. Some samples had coupling strengths that were too strong to be measured by our FMR technique. For such samples, we determined if they were AFC or FC coupled using VSM. The connecting lines with arrows indicate that the coupling is FC and that the coupling strength is too large to be measured by our FMR technique.

The coupling strength for the samples without boron in the spacer layer has not changed significantly after annealing, while all of the samples containing boron within the spacer layer have a strong FC region for thin spacer layer thicknesses. The samples...
with 5% and 10% boron have a strong FC region for spacer layers thinner than 0.45 nm, while the samples with 15% boron have a strong FC region for spacer layers thinner than 0.525 nm. These strong FC regions have replaced what was previously an AFC region before annealing. This is the same behavior as seen in FeCoB/Ru/FeCoB samples annealed above 200 °C with spacer layers less than 0.6 nm in our previous work.18

These results indicate that having boron in the sample causes the same effect after annealing regardless of whether boron is in the magnetic FeCo layer or in the Ru spacer layer. This is likely due to a phenomenon known as transient enhanced diffusion (TED), which is well known in the semiconductor industry to cause boron to diffuse faster than is expected from normal thermal diffusion.24 This would cause the boron atoms to diffuse rapidly into virtually all layers upon annealing, making them relatively uniformly distributed regardless of which layer they started in.

EDXS measurements of the elemental composition of the SERIES-B samples with 15% boron within the spacer layer, as deposited, annealed at 250 °C, and annealed at 450 °C, are shown in Fig. 6. These results show the effect that annealing has on diffusion of the key elements, except boron, which was not accessible in the present analyses due to a strong overlap of the B-K line with the Ru-M line.

Before annealing, there is a slight magenta background signal within the top Ru region. This may be partially due to a small amount of Fe diffusion resulting from the energy involved during the sputtering process, and the negative enthalpy of formation of RuFe.27 Ni, on the other hand, has a positive enthalpy of formation with Ru, so it has not diffused into the RuB layer. Additionally, Fe fluorescence radiation caused by high-energy Ru x rays is likely partially responsible for the magenta Fe signal within the top Ru region. A similar effect is observed for the Ta layer region near the bottom.

After annealing at 250 °C, we now see that the magenta Fe signal of the NiFe film is no longer superimposing with the cyan Ni signal but has moved upwards toward the surface of the sample. This indicates that the bulk of the Ni and Fe atoms is beginning to separate within the NiFe layer. The Fe atoms have begun to diffuse into the RuB layer, while the Ni atom positions have been relatively unaffected. This differs from the as-deposited sample, in which Ni and Fe atoms are relatively uniformly mixed within the NiFe layer. For this sample annealed at 250 °C, we can see that the center of the Fe signal has moved slightly into the RuB layer (Fig. 6).

After annealing at 450 °C, we see that a large fraction of the Fe atoms have diffused into the RuB layer. Again, this is caused by the negative enthalpy of formation of Fe with Ru and is in agreement with the results from Schmalhorst et al.,27 who found that in FeCo/Ru structures, Fe and Co, which also have a negative enthalpy of formation with Ru, diffused into the Ru spacer when annealed above 325 °C.

Figure 7 shows the elemental composition line profiles, which have been extracted from EDXS measurements, for two SERIES-B samples, one with 15% B and one with 0% B in the Ru layer. Both samples were annealed at 250 °C, just like the SERIES-A samples shown in Fig. 5. These results show the effect that adding B to the Ru layer has on diffusion of the Ta, Ni, Fe, and Ru atoms. It can be seen that the addition of boron has caused the Fe signal to shift 0.4 nm further into the top Ru layer than it did in the sample without boron. The Ta, Ni, and Ru signals were relatively unaffected by the addition of B into the Ru layer. These results indicate that the presence of boron enhances the diffusion of Fe into the adjacent spacer layer.

The observed diffusion of Fe into Ru is likely responsible for the change in the sign of coupling in the 250 °C annealed
emphasis was on the spacer layer thickness between 0.525 nm for 15% B and thinner than 0.525 nm for 15% B.

SERIES-A samples with 15% B and for spacer layer thicknesses less than 0.525 nm as shown in Fig. 5. This diffusion increases the concentration of Fe atoms in the region of the Ru spacer layer close to the interface, to the point where thin Ru spacer layers become ferromagnetic. Nunn and Girt have demonstrated the same effect by showing that in Co/Ru/Co films, a large concentration of Co or Fe in the Ru spacer layer causes a change in the sign of coupling from AFC to FC. Furthermore, it is likely that the presence of boron would have a similar effect on the diffusion of Co into the spacer layer because it also has a negative enthalpy of formation with Ru. Therefore, SERIES-A samples containing B within the spacer layer likely have increased Fe and Co diffusion from both the adjacent NiFe and FeCo ferromagnetic layers after annealing, causing the observed change in the interlayer coupling.

IV. CONCLUSION

In summary, we studied the exchange coupling between NiFe and FeCo in NiFe/Ru1−xBx/FeCo SAF layer structures before and after annealing, with B concentrations from 0 to 15 at. %.

as-deposited samples, the presence of B in the spacer layer was found to not affect the sign of coupling. However, after annealing, the addition of B in the spacer layer causes increased diffusion of Fe, and most likely Co, into the Ru spacer layer. This increased diffusion results in a change in sign of coupling from AFC to FC in samples with Ru spacer layers thinner than 0.45 nm for 5% and 10% B and thinner than 0.525 nm for 15% B.

REFERENCES

2. Flash Memory (Samsung Electronics, 2007), rev. 1.