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Magnetization reversal mechanisms in Heusler alloy spin valves

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Ferromagnetic layers composed of Heusler alloys, which are predicted to be 100% spin polarized in bulk, have been incorporated into spin-valve sensors to improve performance. Transport studies of spin valves containing Co_2MnGe (CMG) in the free and pinned layers show an increase in field-dependent magnetoresistance that is lower than expected. When 0.5 nm CoFe insert layers are added to the top and bottom surfaces of the 8 nm CMG free layer, the magnetoresistance increases by almost 25% relative to that measured in spin valves with CMG alone. Magnetometry data reveal that the transition between the nominal parallel and antiparallel states is sharp for samples with CoFe/CMG/CoFe, but it is sheared for samples with only CMG. To understand this difference, polarized neutron reflectivity (PNR) was used to probe the interfacial magnetic structure of spin valves with and without CoFe. Near the transition, PNR measurements for the CMG-only samples show spin-flip scattering. Fits to the data revealed that the free layer magnetization is canted relative to the field, and the orientation of the magnetization changes as the field is varied. The free layer reversal thus proceeds via coherent rotation rather than domain formation. In contrast, the absence of spin-flip scattering for the CoFe/CMG/CoFe sample in comparable fields indicates that the mechanism for the free layer reversal is domain formation. Structural analysis revealed that the interface between the free and Cu layers is less distinct in the spin valve with CMG alone relative to the CoFe/CMG/CoFe sample. Enhanced roughness may alter the coupling between the free and pinned layers and thus be responsible for both the undesirable reversal behavior and the reduced magnetoresistance. © *2011 American Institute of Physics*. [doi:10.1063/1.3551592]

Future high-density magnetic recording applications may use current perpendicular to the plane (CPP) spin valves (SV) that exhibit giant magnetoresistance (GMR). The signal from CPP-GMR SV's is limited, in part, by the intrinsic spin polarization of the ferromagnetic layers used to form the structure. The primary components of a typical SV include an antiferromagnetic layer that is exchange coupled to a pinned ferromagnetic layer (PL), a nonmagnetic spacer layer, and a free ferromagnetic layer (FL) with magnetization that is free to align with the applied field. The resistance is greatest in these structures when the magnetic moments of the FL and PL align antiparallel. Furthermore, the magnitude of the change in resistance between the parallel and antiparallel state is directly related to the spin polarization of these layers. Heusler alloys, such as Co_2MnGe (CMG), have been predicted to be half-metallic ferromagnets which have a high spin polarization. If experimentally achievable, 100% spin polarization would theoretically lead to infinite GMR in Heusler-based spin valves. While substantial improvements have been realized using Heusler-based read-heads, extremely high GMR values have not yet been achieved.[1]

To increase the GMR in Heusler SV's, we have added thin $Co_{50}Fe_{50}$ layers to the sides of a CMG FL.[2] For this investigation, a series of spin valves were fabricated on 18 × 18 mm glass substrates at Hitachi Global Storage Technologies using DC magnetron sputtering and then annealed at 245 °C. The samples, used for both magnetometry and neutron reflectivity studies, had the following structure: glass/Ru 3 nm/Cu 3 nm/FL/Cu 5 nm/CoFe 0.5 nm/CMG 4 nm/CoFe 1 nm/IrMn 6 nm/Ru 12 nm in which the FL composition was CMG 8 nm (Sample 1) or CoFe 0.5 nm/CMG 8 nm/CoFe 0.5 nm (Sample 2). To determine the relative importance of the two interfaces of the FL, Samples 1 and 2 were compared to SV's in which the FL composition was CMG 8 nm/CoFe 0.5 nm (Sample 3) and CoFe 0.5 nm/CMG 8nm (Sample 4). The same four structures were grown on conductive NiFe bottom leads and fabricated into CPP-GMR SV's for transport measurements. Details of film preparation are provided elsewhere, including evidence that CoFe nanolayers adjacent to CMG improve GMR.[2] Our current studies indicate that the CoFe nanolayers increase the field range over which the antiparallel state is stable. Using polarized neutron reflectivity (PNR), depth-dependent measurements of the magnetization reveal that the reversal process in SV's with a bare CMG FL is accompanied by a canting of the magnetization relative to the field. This canting is clearly responsible for at least some of the reduction of the MR in this SV as it inhibits antiparallel alignment and it is not present in the sample with a CoFe/CMG/CoFe FL. Structural characterization via neutron reflectivity and x-ray reflectivity suggests that the interface between the free and nonmagnetic layers is somewhat sharper in SV's with CoFe. Thus, the

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maximum achievable spin polarization in Heusler spin valves may be limited by the growth characteristics of the ferromagnetic free layer.

For the set of samples considered in our current investigation, transport measurements reveal that the GMR increases by almost 25% when CoFe layers are added to the CMG. Specifically, the GMR measurements for sample 1 (CMG only) and sample 2 (CoFe/CMG/CoFe) were 2.56 ± 0.34 mΩ/μ^2 and 3.18 ± 0.23 mΩ/μ^2, respectively.[3] A similar difference is evident when comparing the GMR measurements for sample 4 (CoFe/CMG) and sample 3 (CMG/CoFe), which were 2.39 ± 0.21 mΩ/μ^2 and 3.05 ± 0.24 mΩ/μ^2, respectively. Enhancement of the GMR thus occurs only when CoFe is added to the side of the CMG FL adjacent to the Cu spacer layer.

Magnetic hysteresis measurements performed at room temperature in a vibrating sample magnetometer provide further insight into the origin of the variation of the GMR among the samples. Upon decreasing the field, the relative alignment of the free and pinned layers in sample 2 (with FL = CoFe/CMG/CoFe) changes abruptly from parallel to antiparallel at -1.2 mT [Fig. 1(a)]. The antiparallel state appears to be stable over a field range of approximately 70 mT. In contrast, the reversal of the free layer (FL = CMG) in sample 1 is sheared, and the moment has a pronounced slope in the field region (approximately -10 mT to -60 mT) through which the antiparallel state is nominally stable [Fig. 1(b)]. The reversal mechanism for the FL clearly differs among these samples. It is notable that the shape of the hysteresis curves for both samples 3 and 4 (CMG/CoFe and CoFe/CMG FL's, respectively) more closely resemble those shown in Fig. 1(a) rather than Fig. 1(b).

To investigate the magnetic reversal process, PNR measurements were carried out at the NIST Center for Neutron Research at room temperature on samples 1 and 2. PNR is sensitive to the depth-dependent structure and magnetization on a nanometer length scale and has proven to be effective in determining the relative orientation of free and pinned magnetic layers in analogous metallic spin valves.[4,5] Using Fe-Si supermirrors, incident and scattered neutrons with a wavelength of 0.475 nm were polarized parallel to the applied field, as detailed elsewhere.[6] Al-coil flippers before and after the sample enabled the measurement of the two nonspin-flip (NSF) cross sections, R^{++} and R^{--}, in which neutrons preserve their polarization, and the two spin-flip (SF) cross sections, R^{+-} and R^{-+}, in which the neutrons' polarization rotates 180°. SF scattering is entirely magnetic in nature and arises because of the net in-plane magnetization perpendicular to the applied field. The difference between the R^{++} and R^{--} data is related to the component of the net in-plane magnetization parallel to the field. The NSF data are also sensitive to the nuclear composition of the sample as a function of depth. Before the data were analyzed, all four cross sections were corrected[6] for the footprint of the beam, the instrument polarization efficiency ($> 97\%$), and instrumental background. To create depth-dependent nuclear and magnetic profiles of the sample, the reduced data were fitted using least squared optimization in the reflectivity software REFLPAK.[7] X-ray reflectivity[8] was also measured and analyzed on all four samples, and the x-ray structural parameters, including roughness and layer thickness, were iteratively reconciled with those obtained from analysis of the PNR data to ensure consistency.

Full PNR spectra for sample 1 (CMG only) were obtained at fields of 850 mT, 300 mT, 1.7 mT, -5.0 mT, -25 mT, and -52 mT along the hysteresis loop, as indicated by the points in Fig. 1(b) (for fields < 100 mT). Figure 2(a) shows a plot of the reflectivity measured in a field of -25 mT, in which the FL and PL are expected to align antiparallel along the field direction. Surprisingly, the spin-flip scattering is pronounced, indicating that a component of the magnetization is oriented perpendicular to the field. The spin-flip scattering is absent in large fields (>300 mT) and only appears below -3 mT upon reducing the field after positive saturation. The SF scattering then gradually decreases as the field is reduced from -25 mT to -52 mT. Fits to the -25 mT data reveal that the FL magnetization is oriented at an average angle of 25–30° relative to the applied field, while the PL is canted toward the free layer at an angle of $175 \pm 2°$ relative to the field. The orientations of the FL and PL magnetization do not vary substantially upon decreasing the field to -52 mT, though the net magnetization of the PL decreases by approximately 25% due to in-plane domain formation. This unusual magnetic configuration is reminiscent of noncollinear coupling reported in Fe/Cr multilayers[9] and related systems that exhibit GMR.

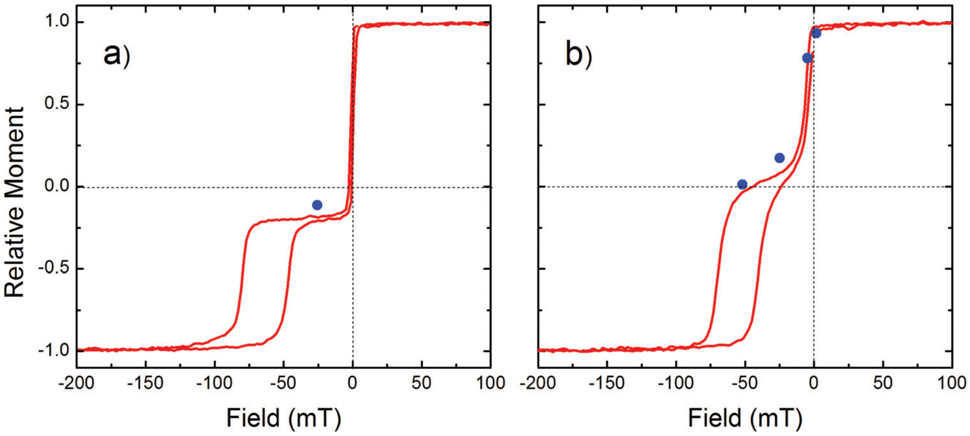

FIG. 1. (Color online) (a) Hysteresis loop for sample 2 with a CoFe/CMG/CoFe FL. (b) Hysteresis loop for sample 1 with a CMG FL. The dots on the hysteresis loops represent the relative net moment determined from fits to PNR data. The agreement between the dots and the line confirm the consistency between PNR and magnetization measurements.

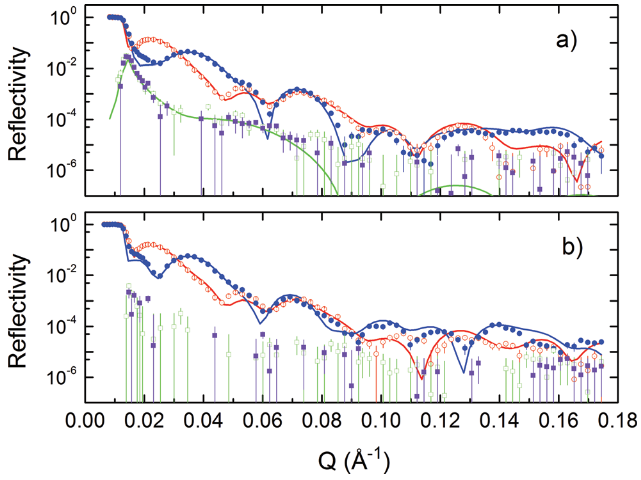

FIG. 2. (Color online) Polarized neutron reflectivity in −25 mT for (a) sample 1 (CMG FL) and (b) for sample 2 (CoFe/CMG/CoFe FL). The blue closed circles and red open circles represent NSF data. The purple closed squares and green open squares represent SF data. The lines correspond to fits to the data.

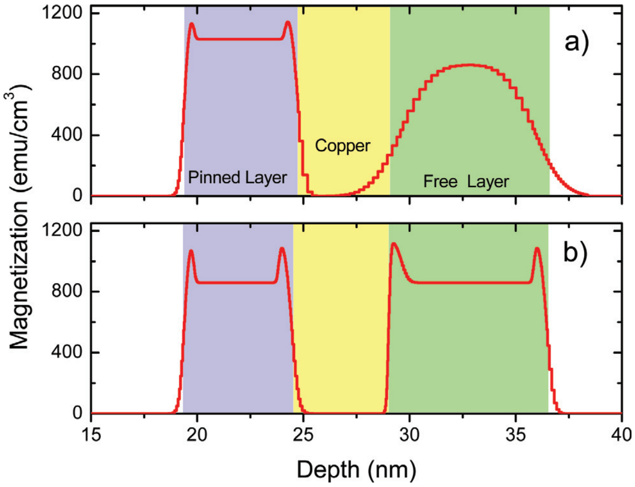

FIG. 3. (Color online) Magnetization depth profile obtained from fits to the polarized neutron reflectivity at 300 mT for (a) sample 1 (CMG FL) and (b) for sample 2 (CoFe/CMG/CoFe FL).

Judging by the magnetization data shown in Fig. 1(b), this canted state forms gradually upon decreasing the field from positive saturation. Fits to PNR data obtained in −5.0 mT indicate that the PL magnetization is again oriented at an angle of 175° ± 2° relative to the field, and the FL is nearby at an average angle of 136° ± 2°. These data suggest that the reversal of the FL magnetization in spin valves with only CMG proceeds via coherent rotation[10] rather than via domain formation, as anticipated.

In contrast, magnetization reversal in the CoFe/CMG/CoFe SV (sample 2) is more conventional. Figure 2(b) shows PNR data for this sample measured in a field of −25 mT, corresponding to the flat plateau in the magnetization data [Fig. 1(a)]. The absence of spin-flip scattering indicates that the moments are not canted, and fits reveal that the free and pinned layers are aligned antiparallel with net magnetizations that are only slightly reduced (approximately 15%) from their saturation values.

The origin of the physical differences between samples with and without CoFe surrounding the CMG FL is apparent upon inspection of the depth-dependent magnetic profiles (Fig. 3) for samples 1 and 2, obtained from fits to the PNR data at 300 mT (i.e., saturation). While the PNR fits proved to be quite sensitive to the magnetization of the individual magnetic layers, the structural layer thicknesses and structural interfacial widths were determined from complementary x-ray reflectivity analysis.[8] In both samples, the CoFe layers in the composite CoFe/CMG/CoFe free and/or pinned layers are distinct, with interfacial full-widths that match the approximate width of the CoFe layers (i.e., 0.5–1.0 nm). In contrast, the full-width of the interfaces of the CMG FL in sample 1 [Fig. 3(b)] is 2.5 ± 0.5 nm, and the layer is somewhat intermixed (via roughness or interdiffusion) with the neighboring Cu.

We speculate that the presence of Cu throughout the broad CMG interfacial region is responsible for suppression of the GMR. Cu impurities are predicted to dramatically reduce spin polarization in CMG.[11] In addition, interfacial roughness may change the nature of the magnetic coupling between the free and pinned layers (i.e., via pinholes) and give rise to the canting of the magnetization and to the undesirable reversal behavior of the FL.

The addition of CoFe between the CMG free layer and the Cu layers, however, facilitates the growth of smoother interfaces in these spin valves, and subsequently leads to a sharper transition between the parallel and antiparallel magnetic states as the field is varied. It is notable that a substantial gain in the GMR was achieved when only a single, thin CoFe layer was grown between the CMG and Cu. The characteristics of the interface between the ferromagnetic Heusler and nonmagnetic layer are key parameters in the performance of these SV sensors, and future work will focus on exploring other means to perfect the structure.

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[8] See supplementary material at http://dx.doi.org/10.1063/1.3551592 that includes x-ray reflectivity data, fits, and depth profiles for Samples 1 and 2.
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