Large and Local Magnetoresistance in a State-of-the-Art Perpendicular Magnetic Medium

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Morgan Williamson^{1,2}
Cheng Wang³
Pin-Wei Huang³
Ganping Ju³
Maxim Tsoi^{1,2}

¹Physics Department, University of Texas at Austin, Austin, TX 78712, USA; ²Texas Materials Institute, University of Texas at Austin, Austin, TX 78712, USA;

³Fremont Research Center, Seagate Technology, Fremont, CA 94538, USA

Purpose: Magnetotransport properties of granular oxide-segregated CoPtCr films were studied on both macroscopic and microscopic length scales by performing bulk and point-contact magnetoresistance measurements, respectively. Such a perpendicular magnetic medium is used in state-of-the-art hard disc drives and, when combined with magnetotransport phenomena for read/write operations, may lead to a novel concept for magnetic recording with high areal density.

Materials and Methods: The CoPtCr films were deposited by an epitaxy-like sputtering and contained several perpendicularly magnetized granular-media layers with different coercivities; they are very much like the state-of-the-art perpendicular magnetic medium, which can be found in today's hard disc drives. Magnetoresistive properties of bulk films were assessed by measuring the film resistance in the standard Van der Pauw geometry, while the local transport was probed by the point-contact technique.

Results: The bulk measurements showed only a negligible magnetoresistance of less than 0.02%. In contrast, the local point-contact measurements revealed giant-magnetoresistance-like changes ΔR in local resistance of the contact R with more than 10,000% ratio $\Delta R/R$.

Conclusion: The observed large and local magnetoresistive effect could be tentatively attributed to a tunnel magnetoresistance between oxide-segregated CoPtCr grains with different coercivities. The tunneling picture of electronic transport in our granular medium was confirmed by the observation of tunneling-like current-voltage characteristics of the contacts and bias dependence of the contact magnetoresistance – both the local point-contact resistance and magnetoresistance were found to decrease with the applied dc bias.

Keywords: magnetoresistance, granular magnetic medium, tunnel magnetoresistance, hard disc drives

Introduction

Magnetic data storage has been pushing scientific innovations and technology limits for more than half a century and has grown into a multi-billion-dollar industry today.¹ The recording of information in a magnetic medium, eg a thin magnetic film in a hard disk drive (HDD), is achieved by switching its magnetization locally between two different orientations, which represent “1” and “0” of digitally stored information. The way of storing bit information in a granular medium with high magnetic anisotropy (K_U) is by far the most cost-effective technology for high-density data storage and has a clear road map of extendibility up to 2 TB/in².² Such granular systems with high scalability and desired thermal stability can also be potentially leveraged into fabricating memory cells of non-volatile magnetic memory such as magnetic random access memory (MRAM). We thus envision

Correspondence: Maxim Tsoi
Email tsoi@physics.utexas.edu

a merging of hard drive granular medium and the MRAM concepts into a novel magnetic memory where reading and writing operations will be achieved by magnetoresistive transport means like in MRAM, while the thermally stable perpendicularly magnetized grains with diameters of below 10 nm provide extraordinary scaling potential for memory applications.

In this letter, we explore magnetoresistive effects in the state-of-the-art perpendicular magnetic medium of today's HDDs – granular oxide-segregated CoPtCr films deposited on glass substrates. We use point-contact technique³ to characterize the local transport in the film composed of granular layers with different coercivities. When the relative orientation of layers changes under the influence of an externally applied magnetic field, we observe large ($>10,000\%$) variations in the point-contact resistance. Interestingly, the magnetoresistance is essentially absent on the macroscopic scale – bulk resistivity measurements performed on the same films showed practically no magnetoresistance ($<0.02\%$). The observed local and large magnetoresistance was found to decrease with the value of dc bias applied to the contact. Such variations are consistent with a tunnel magnetoresistance between individual CoPtCr grains and support the potential of the granular medium for future magnetic memory technology.

Materials and Methods

In our experiments we have tested a number of thin-film samples deposited by the media development team at Fremont Research Center, Seagate Technology with various layer combinations, compositions, and individual layer thicknesses ranging from 0 to 100 Å. In what follows we focus mainly on the magnetoresistive behavior of

a particular multilayer/sample: Ru (180)/M1 (100)/Spacer (15)/M2 (100)/Spacer (15)/M3a (100)/M3b (10) where all numbers in brackets are thickness in Å; this multilayer sample has a 63 nm thick adhesion/seed layer underneath, and a 6 nm thick continuous CoPtX capping layer (X is combination of non-magnetic diluting elements such as Cr, B, and Ru). M1, M2, M3a, and M3b are perpendicularly magnetized granular-media layers with different coercivities and a columnar structure of the metallic grain cores. The columnar structure is achieved by an epitaxy-like sputtering deposition⁴ of the CoPtCr alloy⁵ on top of a Ru-rich columnar growth template. Spacer layers are Ru-rich materials with oxide contents and are inserted to break the interlayer exchange coupling between adjacent ferromagnetic layers. Individual magnetic grains are segregated by a mixture of TiO₂, SiO₂ and other oxides. **Figure 1A** shows a top-down planar-view transmission electron microscopy (TEM) image of the granular composite media. **Figure 1B** shows the magneto-optical Kerr effect (MOKE) hysteresis loop of a single magnetic layer of the granular medium. The Kerr signal confirms the perpendicular anisotropy of the material and displays the effects of the magnetic grain switching field distribution and demagnetizing field anisotropy as evidenced by the shearing of the major hysteresis loop. **Figure 1C** shows a result of micromagnetic modeling which simulates the hysteresis behavior of such a granular system. The modeling parameters were setup as follows: 656 Voronoi grains are generated, with the mean and coefficient of variation of grain-to-grain pitch being 7.8 nm and 16.7%, respectively. The 0 K saturation magnetization and anisotropy field of the medium are 650 emu/cc and 9.2 kOe. Demagnetization and thermal effects (300 K) were taken into account during

Figure 1 (A) Top-down bright field planar-view TEM image of the granular composite media. (B) Magnetization loop of the granular film obtained by the polar Kerr effect. Note that the external field is slightly tilted away from the normal to the film in order to highlight the thermal decay (see the slope at higher fields). (C) Micromagnetic modeling of the hysteresis (major) loop in our granular medium.

the simulation. The damping constant is set to be 0.2 in order to reach equilibrium state within a reasonable computational time. The modeling shows that there are various portions of grains that get reversed at low, median, and high fields, indicating nucleation, coercivity, and saturation, respectively.

Magnetoresistive properties of bulk films were assessed by measuring the film resistance in the Van der Pauw geometry: four-point probes (wire-bonds) were placed in the corners of a $10 \times 10 \text{ mm}^2$ square-shaped media film. A manual ultrasonic West-Bond wire bonder was used to make the probes; Keithley 2400 SourceMeter and Keithley 2000 Digital Multimeter were used to source current and measure voltage, respectively. An external magnetic field could be applied perpendicular to the film's plane (FPP-orientation) or in the plane of the film (FIP-orientation). Local magnetotransport properties of the same thin-film samples were assessed by point contacts used as a probe of small sample volumes (10^3 – 10^9 nm^3) typically associated with such contacts. The point contact was made by bringing a sharpened metallic (Cu or W) tip into contact with the multilayer using a differential screw mechanism; details of the point-contact setup can be found here.⁶ This mechanical point-contact technique enables us to produce electrical contacts of only a few nanometers in diameter (a) and probe electrical transport in very small sample volumes $\sim a^3$. In this work we focus on dc transport measurements of point contacts in magnetic fields B up to 0.7 T applied in the FPP-orientation and dc biases I up to 1.5 mA. All measurements were done at room temperature.

Results and Discussion

We began by investigating the bulk transport properties of our samples. All samples showed a very small magnetoresistance (MR) with MR ratio ($\Delta R/R$) of about 0.02% in the FIP geometry where the current flows primarily along the magnetic field direction. In this paper, the MR ratio is expressed as $\Delta R/R$ where ΔR is the change in the electrical resistance due to an applied magnetic field ($\Delta R = R_{\text{max}} - R_{\text{min}}$ is the difference between maximum and minimum resistance) and R is the minimum resistance ($R = R_{\text{min}}$). No MR was detected in the FPP orientation, and a continuous transition (decrease) in MR behavior was found between FIP and FPP orientations as a function of the field angle.

Contrary to the bulk measurements, point-contact measurements with Cu tips revealed a larger ($\sim 1\%$) MR in both the FPP and FIP orientations. Small variations in the

point-contact resistance under a varying magnetic field confirm that the magnetostrictive effect at the interface between the high anisotropy granular medium and point-contact tip is negligible. At the same time, the relative softness of the copper tip material compared to the hard media sample can be at the origin of a relatively small MR ratio. In order to make stable electrical contact with the sample, a high amount of pressure visibly deformed sharpened Cu tips thus resulting in large contact areas and, consequently, involving many individual grains into the contact region. Due to the complications associated with the relatively soft copper tips, we have switched to tungsten as the point-contact tip material for better mechanical robustness.

Figure 2 shows the resistance of a tungsten point contact under a 0.7 mA bias as a function of an applied magnetic field in the FPP orientation. Starting down from high positive fields (red curve) the contact resistance gradually decreases from $\sim 25 \text{ } \Omega$ to $\sim 20 \text{ } \Omega$. The resistance of $20 \text{ } \Omega$ corresponds to about 5 nm size of a metallic point contact in the diffusive conduction regime.⁷ As the applied field is decreased below $\sim 100 \text{ mT}$ the resistance raises to a relatively high resistance value of $\sim 30 \text{ } \Omega$, demonstrating a 50% change in resistance. At negative fields, the contact resistance remains at this relatively high value down to $\sim -200 \text{ mT}$ and then decreases back to the low value of $\sim 20 \text{ } \Omega$. At high negative fields, the resistance increases again to about $25 \text{ } \Omega$. The up-sweep (black curve) shows a marked

Figure 2 Point-contact magnetoresistance at 0.7 mA bias for a $\pm 400 \text{ mT}$ magnetic-field sweep displaying 50% MR ratio. Red (black) curve depicts magnetic-field down-sweep (up-sweep). The inset shows schematically a point contact between a sharpened W tip (top light grey) and the sample with two magnetic layers (dark grey) separated by a nonmagnetic spacer (light grey).

symmetry to the down-sweep (red curve) replicating both low (R_L) and high (R_H) resistance states. This behavior was observed in a dozen of tungsten point contacts with different resistances and is similar to the switching between the parallel (R_L) and antiparallel (R_H) states in a GMR-like storage element, eg, spin valve. The hysteretic MR behavior is symmetric about the zero applied field but with a properly tuned exchange bias field of ~ 150 mT could potentially be utilized in a field-toggled device with 50% MR ratio.

Along with the major reproducible features, like the high and low resistance states, MR traces show a fine structure that may vary from sweep to sweep. We illustrate the reproducibility and variability of MR traces in Figure 3 which shows major (black/gray) and minor (colored) MR loops for a tungsten point contact under a 0.7 mA bias. Solid (dotted/dashed) curves depict magnetic field down-sweeps (up-sweeps). Each measurement starts at a positive field (just over +400 mT); the field is swept down to a turning-point field (between -400 and 100 mT) and then back up. Minor loops with different turning points

Figure 3 Major (black/gray) and minor (colored) magnetoresistance loops showing magnetic hysteresis of the tungsten point contact at 0.7 mA bias. Each measurement starts at a positive field just over +400 mT. Solid (dotted/dashed) curves depict magnetic field down-sweeps (up-sweeps). Minor loops with different turning points were shifted vertically for clarity. Turning point fields are -400 , -260 , -130 , -60 , 0 , 70 , and 100 mT. Lighter colored curves indicate down-sweep/up-sweep repetitions of the same measurement while thicker traces highlight representative sweeps.

were shifted vertically for clarity; turning-point fields are -400 mT (black/grey), -260 mT (red), -130 mT (green), -60 mT (blue), 0 mT (orange), 70 mT (cyan), and 100 mT (purple). Lighter colored curves indicate down-sweep/up-sweep repetitions of the same measurement while thicker traces highlight representative sweeps. The turning point of the first minor loop (red) coincides roughly with the resistance crossover between high and low resistance states in the major loop, effectively capturing the 50% MR behavior at the smallest negative value magnetic field. The second minor loop (green) with its turning point of -130 mT does not access the low resistance state associated with high negative magnetic fields. Furthermore, the third (blue) and fourth (orange) minor loops all show a similar behavior of remaining in the high resistance state near their turning points. The fifth minor loop (cyan) with its turning point at 70 mT shows a hysteretic opening with slightly less area than the previous three minor loops, while the last minor loop (purple) fails to produce an appreciable hysteretic opening and remains in the low resistance state. The observed behavior is fully consistent with the switching between the parallel (R_L) and antiparallel (R_H) states in a spin valve or magnetic tunneling junction.^{6,7}

Figure 4A shows an example of point-contact magnetoresistance with an MR ratio of $>10,000\%$. Here red and black curves show magnetic field down- and up-sweeps, respectively, for a high-resistance tungsten point contact under a relatively low applied bias of 0.1 mA. Compared with the contact from Figure 2 the principal modification is the resistance of the high resistance state (R_H), while the low resistance state (R_L) and the general form of the MR curve are mostly retained. At high fields, where all layers in the multilayer stack are presumably saturated, the resistance R_L is measured at 20Ω , while at modest fields there are large variations in resistance hovering between 2 k Ω and 6 k Ω . We observed a strong dependence of the MR on the bias current applied to the contact. Lighter colored curves in Figure 4A show MR of the same device under a different (higher) bias current of 1.5 mA that results in a 1500% MR ratio. Figure 4B shows that the MR ratio decreases with increasing bias independent of the bias polarity. We note that the main change in the magnitude of MR originates in the bias dependence of R_H while R_L remains relatively intact. Such a bias dependence of the contact magnetoresistance is very similar to the one well established in magnetic tunneling junctions (MTJs),^{6,8,9} where this behavior is usually associated with magnon-

Figure 4 (A) Magnetoresistance (MR) trace of tungsten point-contact device displaying vast variation in resistance from 20 Ω to 4 k Ω corresponding to >10,000% MR. Red (black) curve depicts magnetic field down-sweep (up-sweep) recorded at low applied bias of 0.1 mA. Lighter colored curves show MR of the same device under a different (higher) bias current of 1.5 mA (1500% MR ratio). **(B)** The bias dependence of MR ratio. **(C)** The current–voltage curves at 0 mT (open circles) and 700 mT (solid squares). The inset shows a zoom into the 700 mT curve.

assisted and intrinsic band-structure mechanisms of bias dependence as well as phonon- and impurity-assisted contributions. Figure 4C shows the current–voltage curves at 0 mT (open circles) and 700 mT (solid squares); the inset of Figure 4C zooms into the 700 mT curve. Here the low-field (0 mT) curve indicates an increase in the contact R_H resistance with decreasing bias, which is consistent with the MTJ behavior. In contrast, the high-field (700 mT) curve displays a metallic-like behavior that correlates with an almost bias-independent R_L .

A point contact between two metallic electrodes at room temperature usually assumes a diffusive metallic conduction,⁷ as ballistic resistance can be ruled out due to the short mean free path of electrons at room temperature.^{10,11} However, neither diffusive nor ballistic regime can explain the very high resistance R_H (almost 10 k Ω) of our point contact in Figure 4; such a high resistance value is usually associated with tunneling contacts. We speculate that the large resistance changes demonstrated in Figure 4 may be caused by tunneling effects, which we would expect to be relevant in our composite sample material. The Co-Pt-Cr granular media under study has a hexagonal columnar structure that is perpendicular to the plane and would presumably support high current densities in the perpendicular direction. The perpendicularly magnetized columns in adjacent magnetic layers of our multilayered samples would serve as channels for these perpendicular currents. However, an

antiparallel orientation of magnetizations in the adjacent layers (layers with different coercivities separated by spacer layers/oxide barriers) may hinder the perpendicular current flow, perhaps bringing it to a point on par with the small in-plane currents between channels, ie, across the oxide segregation barriers. If perpendicular currents and in-plane currents have similar path resistances the total perpendicular current will be funneled to the relatively few magnetic domain interfaces that exhibit aligned orientations. This process, active under low (non-saturating) applied fields, would effectively create a network of tunneling junctions composed of both intralayer and interlayer tunneling junctions leading to possibly very large resistance (R_H). In contrast, a magnetic saturation at higher fields would promote a parallel orientation of magnetizations in the adjacent layers and opening a low-resistance (R_L) perpendicular channel. Observing such behavior would require a probe with a similar contact area as the magnetic grain size, namely, 7–9 nm diameter since probes with larger areas would average over many grains overlooking the nanoscale behavior. Note that recent measurements of tunneling magnetoresistance (TMR) from larger areas of granular magnetic media (with device geometry of ~100 nm or larger) have reported a small TMR ratio of 0.23%, which is possibly attributed to inter-granular averaging effects.¹² Achieving a nanoscale probe area has motivated the introduction of a tungsten point-contact tip instead of copper as discussed above. Tungsten tip is

expected to deform less when contacting the sample surface due to its hardness and will be more mechanically stable so as to minimize the impact of sample's magnetostriuctive effect.¹³ Tungsten point contacts would therefore possess a smaller contact area more suitable for investigating the nanoscale behavior of the granular composite media and associated large resistance changes caused by the proposed tunneling junction network.

Conclusions

In conclusion, we investigated the bulk and local transport properties of CoPtCr granular composite media films. The bulk shows essentially no magnetoresistance. In contrast, local (point-contact) measurements revealed giant-magnetoresistance-like changes in resistance with $>10,000\%$ ratios. The observed magnetoresistive effect is tentatively attributed to a tunneling magnetoresistance between CoPtCr grains with different coercivities. The observed effects support the feasibility of a novel concept of magnetic memory based on granular composites where reading and writing operations would be achieved by magnetoresistive transport means.

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Disclosure

The authors report no conflicts of interest in conducting this work.

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