Determination of Complex Magnetic Structures From Polarized Neutron Reflectivity Data by Flexible Modeling of Depth-Dependent Vector Magnetization

A. D. Mont[1], P. A. Kienzle[2], S. M. Watson[2], J. A. Borchers[2], J. Eckert[3], P. Sparks[3], S. Moyerman[3], and M. J. Carey[4]

[1]University of Maryland, College Park, MD 20742 USA
[2]National Institute of Standards and Technology, Gaithersburg, MD 20899-8563 USA
[3]Harvey Mudd College, Claremont, CA 91711 USA
[4]Hitachi Global Storage Technologies, San Jose CA 95135 USA

In multilayer systems with exchange-coupled layers such as exchange-spring magnets, interfacial pinning can give rise to spiral domain walls and other complex magnetic structures that are sensitive to temperature, relative layer thicknesses, etc. Though these spin structures develop in subsurface layers, the depth-dependent magnetic profile can be fully characterized using polarized neutron reflectivity (PNR). In order to obtain the profile of the vector magnetization as well as the chemical composition, these data are typically analyzed using software in which the sample is described by a series of flat layers. This approach is cumbersome for continuously varying depth profiles, such as magnetic spirals, since the magnetic layers must be artificially subdivided to mimic the smooth changes in the vector magnetization. Thus, we have developed a flexible PNR fitting program in which users can specify a formula for the model (e.g., flat, power law, or piecewise polynomials). The program can easily be extended to handle simultaneous fitting of multiple data sets from measurements made with different techniques (such as PNR and X-rays) with constraints between the models.

***Index Terms*—Depth-dependent magnetization, exchange springs, polarized neutron reflectometry, spiral domain wall.**

I. INTRODUCTION

THE DISCOVERY of giant magnetoresistance (GMR) [1] has prompted the development of spintronic devices for commercial applications such as hard-drive read heads and has led to the development of spin valves exploiting the exchange-bias phenomenon. An exchange-biased spin valve (EBSV) consists of a pinned ferromagnetic (FM) layer biased by an adjacent antiferromagnetic (AFM) layer and a free ferromagnetic layer separated by a nonmagnetic spacer layer. While GMR measurements provide information about the spin valve properties, such as the coercive and exchange-bias fields, they do not provide information about the individual magnetic layers or interfaces between layers, which drive the behavior of the composite system. Polarized neutron reflectometry (PNR) can probe the individual magnetic layers and measure the depth-dependent vector magnetization. PNR is thus a useful tool in building a microscopic model of the field-dependent magnetization and switching.

A previous PNR study of a spin valve with a 1.6 nm antiferromagnetic IrMn layer [2] reveals that a domain wall develops in the pinned $Co_{86}Fe_{14}$ layer parallel to the FM/AFM interface after magnetic training (i.e., field cycling). In an attempt to better understand the exchange-bias on a microscopic level, a study using a thin AFM layer was conducted. The sample was grown onto a 1.8 cm^2 Si (100) substrate using dc magnetron sputtering, as described elsewhere [2]. The spin valve consisted of $Si/Ta(5)/Ni_{80}Fe_{20}(3)/Co_{60}Fe_{40}(1)/Cu(3)/Co_{60}Fe_{40}(3)/Ir_{20}Mn_{80}(0.6)/Cu(1)/Ta(5\ nm)$ structure. Pinning in the top CoFe layer was achieved by cooling from 300 K in a saturating field of -700.0 mT. A GMR value of $\sim 20\%$ was measured at 5 K along with an exchange field of 28.0 mT and a coercive field of 11.5 mT as shown in the magnetoresistance data in Fig. 1. The first field cycle exhibits the typical square loop and a maximum resistivity present over a long plateau. The second field cycle shows a reduced maximum resistivity over a lesser range. In general, regions of low resistance are attributed to parallel alignment of the FM layers while regions of high resistance are attributed to antiparallel alignment, nominally observed to be square in shape. For this film, this is an incorrect assumption as the hysteresis curve exhibits a "training effect" with field cycling. The first field cycle exhibits the expected hysteresis for a film with decoupled FM layers, but the second field cycle exhibits a rounded edge

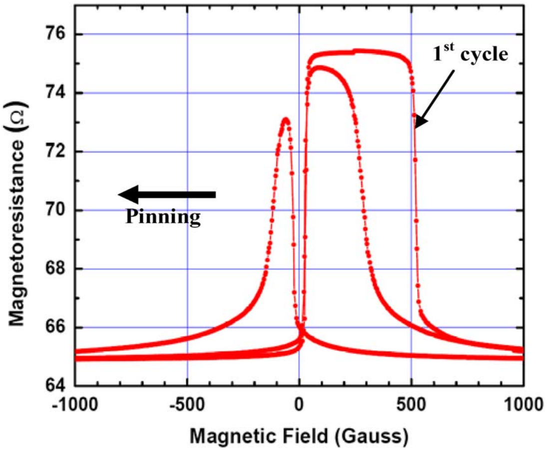

Fig. 1. Initial field cycle (Sweep #1) has a typical hysteresis with a long plateau at maximum resistivity. The second field cycle has a reduced maximum resistivity over a lesser range. (1 $G = 0.1$ mT).

Digital Object Identifier 10.1109/TMAG.2007.893870

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

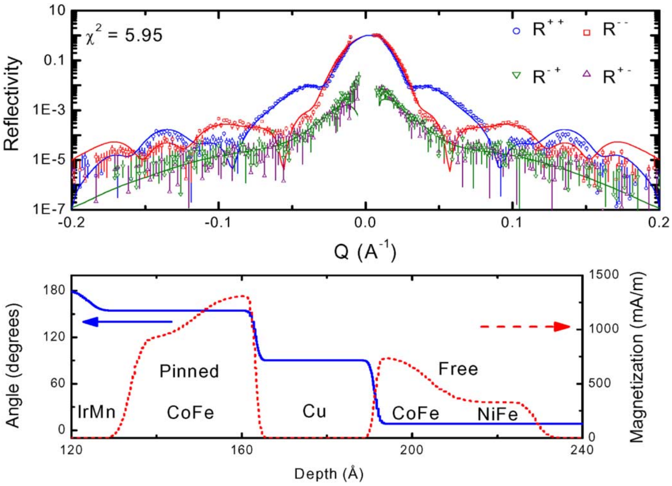

Fig. 2. Reflectivity data taken at -4.5 mT and 5 K. (a) Back (left)/front (right) Polarized neutron reflectivity data. (b) Magnetization as a function of depth for -4.5 mT as obtained from the fits to the reflectivity data. Magnetization angle was constrained to be uniform relative to the applied field within a layer. The pinned CoFe layer was artificially split, with the two halves free to take on independent values of magnetic magnitude and interfacial roughness creating the sloped profile.

corresponding to a decrease in the range of the maximum resistivity.

PNR measurements at the NIST center for Neutron Research were carried out to probe the spin structures of the individual layers. Measurements were performed using neutrons with a wavelength of 0.475 nm polarized parallel to the applied field using Fe/Si supermirrors. The data were corrected for efficiency of the polarizing elements (typically $>97\%$), background, and footprint of the beam. This procedure generates four reflectivity cross-sections: R^{++} and R^{-}, designated nonspin flip (NSF) as the neutron retains its original polarization, and R^{+-} and R^{-+}, designated spin flip (SF), where the neutron spin rotates 180°. SF reflectivity is sensitive only to the component of the magnetization which lies perpendicular to the field direction, while NSF reflectivity provides information concerning the chemical composition of the film and the component of the magnetization aligned along the field axis.

Fig. 2(a) shows the reflectivity data obtained at 5 K in a field of −4.5 mT. The reflectivity data from the back (substrate) surface are plotted versus the $-Q$ wave vector on the left while the data from the front (sample) surface are plotted against $+Q$ wave vector on the right. The NSF cross-sections are collapsed onto each other at low Q, which is indicative of an antiparallel alignment for the free and pinned FM layers.

The data were initially fitted to the reflectivity theoretical formalism [3]–[5] with the REFLPAK software [6] using least squares optimization. Further fits were obtained using genetic algorithm optimization, which permits simultaneous fitting of multiple data sets with both correlated and uncorrelated parameters. The fit shown in Fig. 2(a) is excellent, though it overshoots the data at higher values of Q. The magnetization as a function of depth [Fig. 2(b)] indicates that the NiFe/CoFe free layer is tilted at an angle of 10° relative to the applied field. Though the pinned CoFe layer is expected to be antiparallel to the field, it is uniformly aligned at an angle of 155° rather than 180°, giving rise to a significant amount of SF scattering. It is surprising that the moment in the pinned layer is reduced near the top of the layer. This result could be explained by the formation of in-plane magnetic domains that cannot be detected by PNR, but this effect is more pronounced than would be expected.

REFLPAK was able to generate reasonable fits at this field and at other field values. However, we cannot say with certainty whether this is the only model that will fit the data. This is true in general for all PNR fits because of a loss of phase information about the reflected neutron wave [4], [5] that is intrinsic to the measurement technique. Strictly speaking, we can only say that domains appear to be present in the pinned CoFe layer. We cannot be sure of the specific characteristics of these domains.

II. Analysis

A desirable tool for reflectivity analysis would allow one to convert the measured reflectivity directly into the unique depth profile which generates it. Since the neutron phase information cannot be determined from the measurement, we are limited to solving the inverse problem, where parameter values are adjusted until a model matches the data.

Magnetic systems can be modeled using a series of slabs described by four parameters: scattering length density, neutron absorption, magnetic scattering density, and magnetic field angle. Following the work on parametric B-spline fitting of nuclear structures [7], we attempted to improve the flexibility of magnetic modeling using a free-form curve in each profile dimension. The fits we obtained using this technique were excellent, easily matching the measured data; however, many alternative free-form curves fit the data equally well. To reduce this uncertainty, we include information from other sources using a hybrid approach that combines the flexibility of free-form fitting with the strength of the traditional model-based approach.

The resulting program, KsRefl, is a curve-fitting software utility designed to fit neutron reflectivity data. It freely mixes specific models for nuclear and magnetic properties of the material for a given layer with free-form segments. To use the program, the user sets up a series of layers, each representing the materials at different depths in the sample. In programs such as RELFPAK, the layers are typically described as having uniform density in the bulk, with Gaussian smearing between layers to describe interfacial roughness or interdiffusion. In KsRefl, the layers describe more complex structures, such as a sinusoid parameterized by amplitude, offset, wavelength, and phase. For each layer and property, the user selects initial values for the parameters and decides which parameters to vary. Layer types supported by KsRefl include flat, power-law, B-spline, sinusoidal, as well as specific models for systems such as tethered polymers.

Fig. 3(b) shows the magnetization profile generated from the KsRefl fit in Fig. 3(a). Note that χ^2 is one half that of the fit to the slab model in Fig. 2(a). The magnitudes of the magnetization in the CoFe and NiFe layers were constrained to be uniform through the depth (though still subject to interfacial broadening), but the angle was free to vary. Layer thickness and interfacial roughness were allowed to vary by 50% from nominal

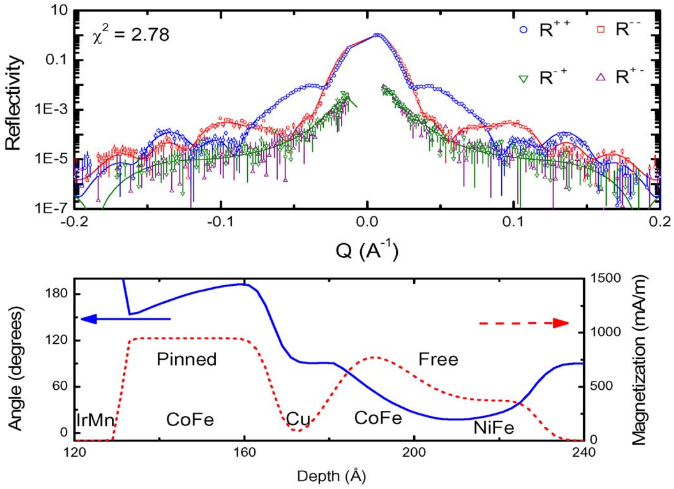

Fig. 3. Reflectivity data taken at -4.5 mT and 5 K. (a) Back (left)/front (right) Polarized neutron reflectivity data. (b) Magnetization as a function of depth for -4.5 mT as obtained from the fits to the reflectivity data. Magnetization angle was constrained to be a cubic spline with three control points.

values. The small thickness for the Cu spacer and the broad interfacial roughness were observed in all fits. The results are consistent with small vectorial spin domain walls in *both* the pinned CoFe and free CoFe layers. Because of the weakness of the spin flip scattering, this measurement was mainly sensitive to the cosine of the angle relative to the applied field. While the profile in Fig. 3(b) differs from our initial expectations (including a Cu spacer thickness 1/2 the nominal value), these features consistently appeared in a series of profiles produced by KsRefl, all of which fit the data equally well. The KsRefl fits indicate that domain structure may have nuances that were previously overlooked.

In general it is awkward to fit continuously changing profiles using only flat layers, such as done with the REFLPAK software. For the fits in [2], for example, we had to create several "pseudolayers" with complicated constraints to generate the depth profile of the vector magnetization. KsRefl eliminates the need for artificial layers, and makes it easier to model directly a situation in which there is theoretical justification for a profile with a specific functional form, such as a tethered polymer, but the values of the relevant parameters are unknown.

Using a hybrid model of a layered nuclear structure and a free-form magnetic structure we have more robust, unique fits that support the choice of a magnetic twist model in our spin valve samples and provide information about the spiral characteristics that are consistent with physical expectations. The fit, in this case, is assisted with an *a priori* knowledge of aspects of the structural depth profile. Specifically, the structural scattering length densities of the bulk materials are well known and tightly constrained, and some layer thicknesses were determined from X-ray reflectivity.

In KsRefl, an optimization routine is used to find a set of values of the fit parameters that result in a reflectivity calculation matching the observed data. Fits can be constrained to physically meaningful states by adding bounds to the fit parameters. KsRefl generates a covariance matrix on the fit parameters. It has the ability to generate a large number of profiles by using the covariance matrix to perturb the fit parameters, thus generating a family of profiles from which we can estimate a confidence interval for each portion of the profile. We are also adding features such as refinement of multiple datasets with constraints between models. All these improvements promise to improve further the quality of the fits produced. Even at the current stage of development, the software is already useful for identifying and characterizing complex, continuously varying spin structures (such as spirals) in magnetically coupled layers.

ACKNOWLEDGMENT

This work was supported in part by the U.S. National Science Foundation under Grant DMR-0412074. This paper was presented at the Tenth Joint Magnetism and Magnetic Materials–International Magnetics (MMM–INTERMAG) Conference. See IEEE TRANSACTIONS ON MAGNETICS, vol. 43, no. 6, June 2007.

REFERENCES

[1] M. N. Baibich, "Giant magnetoresistance of (001)Fe/(001)Cr magnetic superlattices," *Phys. Rev. Lett.*, vol. 61, pp. 2472–2475, Aug. 1988.
[2] S. M. Moyerman, J. C. Eckert, J. A. Borchers, K. L. Perdue, M. Doucet, P. D. Sparks, and M. J. Carey, "Magnetic structure variations during giant magnetoresistance training in spin valves with picoscale antiferromagnetic layers," *J. Appl. Phys.*, vol. 99, p. 08R505, Apr. 2006.
[3] L. G. Parratt, "Surface studies of solids by total reflection of X-rays," *Phys. Rev.*, vol. 95, p. 359, 1954.
[4] C. F. Majkrzak, "Neutron scattering studies of magnetic thin films and multilayers," *Physica B*, vol. 221, pp. 342–356, Apr. 1996.
[5] G. P. Felcher, "Neutron reflectometry as a tool to study magnetism," *J. Appl. Phys.*, vol. 87, p. 5431, 2000.
[6] P. A. Kienzle, M. Doucet, D. J. McGillivray, K. V. O'Donovan, J. F. Ankner, N. F. Berk, and C. F. Majkrzak, "REFLPAK reduction and analysis software suite." [Online]. Available: http://www.ncnr.nist.gov/reflpak 2000–2006
[7] N. F. Berk and C. F. Majkrzak, "Using parametric B-splines to fit specular reflectivities," *Phys. Rev. B*, vol. 51, p. 11296, 1995.

Manuscript received November 1, 2006; revised February 5, 2007. Corresponding author: P. Kienzle (e-mail: paul.kienzle@nist.gov).