

Structural and magnetic roughness in a Co/Ru multilayer patterned into a large scale hexagonal array

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(Presented on 7 November 2007; received 11 September 2007; accepted 3 October 2007; published online 11 February 2008)

Self-assembled arrays of nanospheres have been used to pattern a Co/Pt multilayer into nanopillars. On top of this has been deposited a Co/Ru multilayer, which is antiferromagnetically coupled. The nanopillars introduce a known structural and magnetic lateral modulation into the multilayer. Soft x-ray magnetic scattering was used to observe the interference patterned from the patterned substrate. This has allowed us to show how the magnetic roughness correlates with the structural roughness and obtain selective magnetometry of the various magnetic elements. © 2008 American Institute of Physics. [DOI: [10.1063/1.2829394](https://doi.org/10.1063/1.2829394)]

There are great difficulties in obtaining information on buried interfaces in multilayer nanostructures. This is a crucial problem in magnetic information storage and spintronics research, where the magnetic properties are strongly coupled to the structure. Internal characterization of nanostructures is possible using neutron and x-ray scattering, particularly off-specular scattering techniques to investigate lateral disorder.^{1,2} There has been great interest in lateral structures recently partly because the behavior of such systems, when patterned into large scale arrays, provides model systems that can be utilized to study scientifically interesting areas such as digital computation³ and artificial frustrated magnetism,⁴ as well as applications where surface magnetism is important like giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR) devices and patterned media.⁵ In this case, large scale arrays are an ideal system to study the effects of small local deviations in the magnetic structure. These are of interest since these magnetic deviations can affect properties such as polarized electron transport. Scattering techniques allow averaged statistical quantities to be measured from arrays of devices more easily than from local probes on single devices. Past studies have concentrated on patterned samples with nonmagnetic structural modulations using both polarized neutron reflectivity and soft x-ray resonant magnetic scattering (SXRMS).^{6,7} We have made use of

off-specular SXRMS in a previous study on structurally flat Permalloy, which has a domain pattern imprinted upon it by a perpendicularly magnetized Co/Pt multilayer.⁸ In this report, self-assembled arrays of nanospheres were used to pattern perpendicularly a magnetized Co/Pt multilayer into nanopillars. On top of this has been deposited a Co/Ru multilayer, which is antiferromagnetically coupled,⁹ introducing a known structural and magnetic modulation.

The samples were prepared using a dc magnetron sputtering system with a base pressure of $\sim 5 \times 10^{-8}$ Torr. A partial pressure of 2.8 mTorr of Ar was used during deposition. The nominal structure of the samples was [Co(5 Å)/Pt(10 Å)] \times 20(patterned)/Ru(400 Å)/[Co(31 Å)/Ru(31 Å)] \times 20, shown in the inset of Fig. 1. Co/Pt was deposited to obtain a perpendicularly magnetized film.^{10,11} This Co/Pt multilayer was then patterned into a hexagonal dot array using commercially available suspensions of monodisperse polystyrene nanospheres with diameters of approximately 780 nm. These formed a hexagonal template via self-assembly, allowing arrays with ordering over the range of ~ 1 cm². A combination of reactive ion etching and Ar ion milling was used to produce nanosphere capped Co/Pt pillars. The spheres were then removed via abrasion. Further details on this process can be found in the following by Weekes *et al.*^{12,13} A scanning electron microscope (SEM) image of the patterned Co/Pt multilayer is shown in Fig. 1. To ensure that there was

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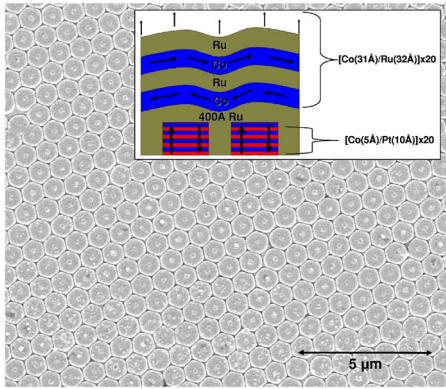


FIG. 1. (Color online) $16 \times 16 \mu\text{m}^2$ SEM image of Co/Pt dots after milling. Inset: schematic of the complete sample structure.

no form of coupling other than magnetostatic between the Co/Pt pillars and the following $[\text{Co}(31 \text{ \AA})/\text{Ru}(32 \text{ \AA})] \times 20$ multilayer, 400 Å of Ru was then deposited to form a large nonmagnetic spacer layer. The Co/Ru multilayer had thicknesses tailored to be on the third antiferromagnetically (AF) coupling peak.⁹

Figure 2(a) shows a Cu $K\alpha$ rocking curve taken through the first order Co/Pt Bragg peak for just the patterned Co/Pt multilayer. The in-plane structure is clearly evident from the satellite peaks about $Q_x=0$. Vibrating sample magnetometry (VSM) loops are shown in Figs. 3(a) and 3(b) for the completed sample structure. Panel (a) shows the full VSM loop, while panel (b) shows a minor loop in the range of ± 400 Oe. VSM is sensitive to the bulk magnetization of the sample. However, the Co/Ru moment is four times the size of the Co/Pt moment, so it is evident that the majority of the minor loops in panel (b) comes from the Co/Ru.

In order to perform the SXRMS measurements, the samples were taken to the U4B beamline at the National Synchrotron Light Source.¹⁴ U4B is equipped with an electromagnet able to apply a field of up to ± 300 Oe in both the plane of the sample and the scattering plane. We define the coordinate system for scattering such that the z direction lies normal to the sample, while the x direction lies in both the sample and scattering planes. The sample geometry is such

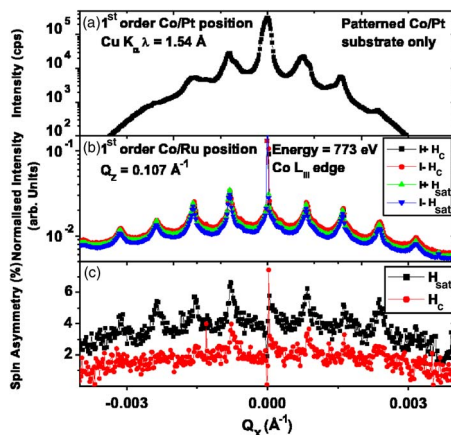


FIG. 2. (Color online) (a) Cu $K\alpha$ x rays on the Co/Pt patterned substrate. (b) Q_x scans at the first order Co/Ru Bragg position ($Q_z=0.108 \text{ \AA}^{-1}$) at saturation and coercivity. (c) Spin asymmetry at saturation and coercivity.

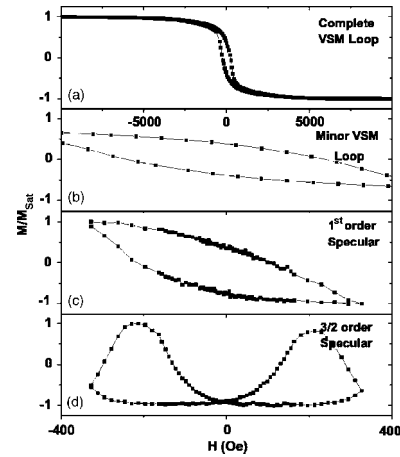


FIG. 3. (a) Full in-plane VSM loop. (b) Minor in-plane VSM loop. (c) SXRMS hysteresis loops at the first order specular position. (d) SXRMS hysteresis loops at the 3/2 order specular position.

that the sample and detector angles allow the components of the elastic wavevector transfer \mathbf{Q} to be selected in x and z . X rays with a polarization of 90% were used, with the reflected intensity being normalized via a Au grid monitor upstream of the entrance slits to the scattering chamber.

In accordance with earlier studies^{15,16} at each point in reciprocal space, the scattered intensity was measured for both field directions (I^+ and I^-). The sum ($I^+ + I^-$) and difference ($I^+ - I^-$) are calculated, with the sum representative of the structure and magnetism self-correlations, and the difference the cross-correlations between the two.¹⁷ We refer to the ratio $(I^+ - I^-)/(I^+ + I^-)$ as the spin asymmetry (SA).

Energy scans (not shown) were recorded across the Co absorption edges, with the maximum in the SA being found close to the L_{III} edge at 772.5 eV and the slight shift from the tabulated value of 778.1 eV (Ref. 18) being due to our reflection geometry. A specular scan (not shown) was taken to determine the position of the Co/Ru Bragg peak at $Q_z = 0.107 \text{ \AA}^{-1}$. No 1/2 order AF peak was observed in the specular scatter, which is in keeping with other studies using SXRMS.¹⁹ In order to probe the lateral magnetic structure, transverse Q_x scans were taken at the 1/2 order ($Q_z = 0.054 \text{ \AA}^{-1}$) and first order ($Q_z = 0.107 \text{ \AA}^{-1}$) Bragg positions.

Figure 2(b) shows the Q_x scan at the first order Bragg peak for applied fields corresponding to saturation and coercivity. The positions of the in-plane Bragg peaks are in excellent agreement with the Cu $K\alpha$ x-ray data shown in Fig. 2(a). The satellite peak positions are also in agreement with what would be expected for their positions via $Q_x \approx 2\pi/d$, which for a center to center nanosphere separation gives a spacing in Q_x of $\approx 8 \times 10^{-4} \text{ \AA}^{-1}$. It is also clear that the Q_x scans at coercivity are slightly more intense than the Q_x scans at saturation due to there being more lateral ferromagnetic (F) disorder at coercivity. There is a good agreement between both the I^+ and I^- curves at both saturation and coercivity, indicating that the in-plane F domain disorder is strongly correlated to the structural modulation. This becomes more evident when looking at the SA shown in Fig. 2(c). The SA shows that at saturation, there is a definite magnetic correlation with the structural modulation. At coer-

civity, any correlations of the ferromagnetic domains and the structure are very weak as there is no net magnetization, but peaks are still just about visible, probably due to the fact that the sample was not quite at coercivity during the measurements. Q_x scans were also taken at the 1/2 order position (not shown) in order to be sensitive to the nominal lateral AF disorder between the Co layers in the Co/Ru multilayer. The curves displayed the same general features and behavior as the first order Q_x scan, indicating some crossover between the AF and F order parameters in both cases. A Gaussian fit to the left hand side in-plane Bragg peak gives a coherence length of ≈ 5000 nm for the dots, comparing well the SEM data, while a fit to the diffuse background (with in-plane features removed) gives an in-plane random roughness of ≈ 420 nm, which is of the same order as the size of the Co/Pt dots upon which the Co/Ru was deposited.

An advantage of the SXRMS technique is that by measuring the intensity variation as a function of applied field, it is possible to measure hysteresis loops.^{2,20} Loops were taken at both the first order and 3/2 order specular positions in Q_z and are shown in Fig. 3 along with a VSM measurement over the same field range for comparison. The first Co/Pt feature is to be found at $Q_z = 0.42 \text{ \AA}^{-1}$; hence, the hysteresis measurements are sensitive to the Co/Ru multilayer stack only.

The first order specular loop compares well to the corresponding minor VSM loop, implying that it is largely the macroscopic magnetization of the structure that is being probed. Hysteresis loops were also taken at the positions of the first trough- and in-plane bragg peaks in Q_x (not shown) as these off specular loops are sensitive to in-plane structures such as domains.² These were found to be of the same shape, indicating that the magnetic disorder is similar over a range of length scales as far out as $15 \mu\text{m}$. The loop taken at the 3/2 position in Q_z is sensitive to the AF magnetic structure between the Co layers in the Co/Ru multilayer. The unconventional loop shape is due to the intensity peaking at the coercive fields of the Co/Ru, where the greatest amount of AF order is present. This then dies off as the AF order is swept out of the sample. These loops are similar to the loops observed in GMR measurements, which are known to be sensitive to the AF order.

Magnetic roughness can have multiple forms such as lateral domain patterns, a nonuniform height distribution of the moments, and vectorial differences in the directions to which the moments point. It has been shown by Kinane *et al.*⁸ that it is possible to measure the domain disorder only, with minimal contributions from the chemical disorder, and that the structure and magnetism are discernible from each other using SXRMS. Langridge *et al.*⁶ have shown that the domain disorder does not necessarily correlate with the structural disorder in the active multilayer, while the magnetic roughness is driven by the structural feature size. In this case, we pattern both the structural modulation and the domain modulation. It is clear that the magnetic roughness locks in on the structure at saturation, while the domain disorder has little influence since it is locked to the domain structure of the dots below the active Co/Ru multilayer. This is backed up by the SXRMS hysteresis loops, which do not vary in

shape as a function of Q_x , which probe the different length scales. It seems a reasonable that different forms of magnetic roughness interact with the structure to different extents.

In conclusion, the magnetic roughness has been shown to follow the structural modulation at saturation, with little sensitivity to the magnetism at coercivity due to the low net magnetization. It is possible to obtain a variety of hysteresis loops as a function of position in reciprocal space. By selecting the correct position in reciprocal space, it is possible to select which part of the sample is measured. In this case, the Co/Ru multilayer rather than the Co/Pt dots, while moving into the diffuse scatter, allows different lateral length scales to be probed. We saw that the magnetic disorder was similar over a range of length scales as far out as $15 \mu\text{m}$.

The authors thank Brookhaven National Laboratory for NSLS beamtime. We are grateful to EPSRC, and the STFC Centre for Materials Physics and Chemistry for funding. We acknowledge Lara San Emeterio Alvarez for the SEM image.

¹S. Langridge, J. Schmalian, C. H. Marrows, D. T. Dekadjevi, and B. J. Hickey, *Phys. Rev. Lett.* **85**, 4964 (2000).

²C. H. Marrows, P. Steadman, A. C. Hampson, L.-A. Michez, B. J. Hickey, N. D. Telling, D. A. Dvorak, and S. Langridge, *Phys. Rev. B* **72**, 024421 (2004).

³J. I. Martin, J. Nogués, K. Liu, J. L. Vicentand, and I. K. Schuller, *J. Magn. Magn. Mater.* **256**, 449 (2003).

⁴A. Imre, G. Csaba, L. Ji, A. Orlov, G. Bernstein, and W. Porod, *Science* **311**, 205 (2006).

⁵R. F. Wang, C. Nisoli, R. S. Freitas, J. Li, W. McConville, B. J. Cooley, M. S. Lund, N. Samarth, C. Leighton, V. H. Crespi, and P. Schiffer, *Nature (London)* **439**, 303 (2006).

⁶S. Langridge, L. A. Michez, M. Ali, C. H. Marrows, B. J. Hickey, T. R. Charlton, R. M. Dalgliesh, M. Toohey, E. W. Hill, S. McFadzean, and J. N. Chapman, *Phys. Rev. B* **74**, 014417 (2006).

⁷L.-A. Michez, C. H. Marrows, P. Steadman, B. J. Hickey, D. A. Arena, H.-L. Zhang, D. G. Bucknall, and S. Langridge, *Appl. Phys. Lett.* **86**, 112502 (2005).

⁸C. J. Kinane, A. K. Suszka, C. H. Marrows, B. J. Hickey, D. A. Arena, J. Dvorak, T. R. Charlton, and S. Langridge, *Appl. Phys. Lett.* **89**, 092507 (2006).

⁹S. S. P. Parkin, N. More, and K. P. Roche, *Phys. Rev. Lett.* **64**, 2304 (1990).

¹⁰L. S. E. Alvarez, G. Burnell, C. H. Marrows, K.-Y. Wang, A. M. Blackburn, and D. A. Williams, *J. Appl. Phys.* **101**, 09F508 (2007).

¹¹S. Landis, B. Rodmacq, and B. Dieny, *Phys. Rev. B* **62**, 12271 (2000).

¹²S. M. Weekes, F. Y. Ogrin, and W. A. Murray, *Langmuir* **20**, 11208 (2004).

¹³S. M. Weekes, F. Y. Ogrin, W. A. Murray, and P. S. Keatley, *Langmuir* **23**, 1057 (2007).

¹⁴See <http://www.nsls.bnl.gov/beamlines/beamline.asp?bliduu4b> for more information.

¹⁵J. F. MacKay, C. Teichert, D. E. Savage, and M. G. Lagally, *Phys. Rev. Lett.* **77**, 3925 (1996).

¹⁶J. W. Freeland, K. Bussmann, P. Lubitz, Y. U. Idzerda, and C.-C. Kao, *Appl. Phys. Lett.* **73**, 2206 (1998).

¹⁷R. M. Osgood III, S. K. Sinha, J. W. Freeland, Y. U. Idzerda, and S. D. Bader, *J. Appl. Phys.* **85**, 4619 (1999).

¹⁸A. Thompson, D. Attwood, E. Gullikson, M. Howells, K. Kim, J. Kirz, J. Kortright, I. Lindau, P. Pianetta, A. Robinson, J. Scofield, J. Underwood, D. Vaughan, G. Williams, and H. Winick, *X-Ray Data Booklet* (University of California, Berkeley, CA, 2001).

¹⁹T. P. A. Hase, J. D. R. Buchanan, B. K. Tanner, S. Langridge, R. M. Dalgliesh, S. Foster, C. H. Marrows, and B. J. Hickey, *J. Appl. Phys.* **93**, 6510 (2003).

²⁰C. Spezzani, P. Torelli, M. Sacchi, R. Delaunay, C. F. Hague, V. Cros, and F. Petroff, *Appl. Phys. Lett.* **81**, 3425 (2002).