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Authors

Scholl, A.
Nolting, F.
Seo, J.W.
et al.

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Domain-size-dependent exchange bias in Co/LaFeO₃

A. Scholl,^{a)} F. Nolting,^{b)} J.W. Seo,^{c)} H. Ohldag,^{d)} J. Stöhr,^{d)} S. Raoux,^{e)} J.-P. Locquet,^{f)}
and J. Fompeyrine^{f)}

a) Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

b) Swiss Light Source, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

c) Swiss Federal Institute of Technology EPFL, CH-1015 Lausanne, Switzerland

d) Stanford Synchrotron Radiation Laboratory, Stanford, CA 94309, USA

e) IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, CA 95120,
USA

f) IBM Research Division, Zürich Research Laboratory, CH-8803 Rüschlikon, Switzerland

(Received

ABSTRACT

X-ray microscopy using magnetic linear dichroism of a zero-field-grown, multi-domain Co/LaFeO₃ ferromagnet/antiferromagnet sample shows a local exchange bias of random direction and magnitude. A statistical analysis of the local bias of individual, micron-size magnetic domains demonstrates an increasing bias field with decreasing domain size as expected for a random distribution of pinned, uncompensated spins, which are believed to mediate the interface coupling. A linear dependence with the inverse domain diameter is found.

Exchange bias, the unidirectional pinning of the magnetization of a ferromagnet (FM) by an antiferromagnet (AFM), is the result of magnetic interface exchange coupling [1]. Since the macroscopic magnetization of an antiferromagnet is compensated by the balance of moment in all spin sub-lattices, it is widely accepted that breaking of the in-plane translational symmetry is required to explain bias, in particular on compensated AFM surfaces. This symmetry breaking can either be the result of interface roughness, of magnetic domains [2], of defects [3], of the grain structure [4], or of a combination of these four. Elaborate models have been developed describing many phenomena related to exchange bias, which consider one or more of these possible sources of bias [2–6]. Common to most models is a randomness of the local bias due to the stochastic distribution of pinned, uncompensated spins in the AFM that mediate the coupling. This randomness can lead to a strong lateral variation of the bias field [7]. The existence of pinned, uncompensated spins was experimentally observed by magnetometry [8, 9], by x-ray dichroism [10, 11], by magnetic force microscopy [12], and by nonlinear optical spectroscopy [13]. It was also shown that the coupling strength scales with the number of pinned uncompensated spins [11, 13] and that it is inversely correlated with the grain size of a polycrystalline AFM [4]. Such a scaling law was first discussed by Malozemoff [2] in a random-field model. The underlying idea is that for a large statistical sample of domains with diameter d and a total number of surface spins of $N \sim d^2$, the statistical deviation of the uncompensated magnetization from exact compensation is proportional to the standard deviation or width of a normal distribution: $\sqrt{N} \sim d$. The width of the bias distribution in such a model is proportional to the uncompensated moment per area d^2 and therefore $\sim 1/d$. In this Letter we will demonstrate that the local bias field of a large sample of domains is indeed normal distributed and that a $1/d$ scaling law describes the width of the bias distribution as function of domain diameter.

We studied an MBE-grown 1.2 nm Co/40 nm LaFeO₃ epitaxial thin film on a SrTiO₃(001) substrate. The sample preparation was described in Ref. [14]. The experiments were conducted at the PEEM-2 microscope of the Advanced Light Source. The sample was neither field-cooled nor field-grown and therefore did not show macroscopic exchange bias. This allows us to study the undisturbed microscopic distribution of the unidirectional coupling. It was shown before that similar samples exhibit local bias [15]. The (001) surface of antiferromagnetic LaFeO₃ is completely compensated with antiferromagnetic axes along out-of-plane $\langle 110 \rangle$ directions [16]. The directions refer to the cubic lattice of the substrate. Interface cou-

pling to the AFM and the magnetostatic field force the Co magnetization parallel to the in-plane projection of the local LaFeO₃ antiferromagnetic axis, parallel to $[\pm 100]$ or $[\pm 010]$. Magnetic domain images of Co using Photoemission Electron Microscopy (PEEM) and X-ray Magnetic Circular Dichroism (XMCD) show 2 classes of FM domains, Fig. 1. The FM domains are coupled 1:1 to AFM domains (compare LaFeO₃ XMLD image and 223 Oe Co XMCD image). A field along $[100]$ switches those Co domains from white (-223 Oe) to black (+223 Oe), which possess a uniaxial anisotropy parallel to $[100]$. The orthogonal $[010]$ domains remain unchanged (gray). Remanent hysteresis loops recorded from single domains show a bias field of random size and random direction. The loops were calculated from a sequence of Co XMCD images, acquired after applying field pulses of increasing magnitude (step 5 Oe) [15]. Two typical loops of spatially close domains, which show opposite bias, are displayed in Fig. 1, bottom left.

A map of the local bias field was generated by analyzing the local loops of all domains i with area A_i above $0.1 \mu\text{m}^2$, inset of Fig. 2. The local bias field h_i was determined as the difference of the switching field measured with increasing positive and negative magnetic field. Domains along $[010]$, which didn't switch, were masked out. The area of each domain was determined by an image analysis program. The bias map shows a wide variation of the local bias field of up to ± 30 Oe. Both negative and positive values are present because no bias direction was set.

Three switching field cycles were measured on the same area to obtain switching data from a total of more than 1200 domains with minimum area above $0.1 \mu\text{m}^2$. A cross-correlation of 0.7 between bias maps of consecutive cycles demonstrated a generally reproducible local bias of individual domains. A domain was defined as a connected area that switched at the same field. The resolution limit of the microscope of $0.1 \mu\text{m} \times 0.1 \mu\text{m} \approx 0.01 \mu\text{m}^2$ was well below the minimum domain size that was considered in the analysis. The switching data as function of field and domain area is summarized in Fig. 2. The majority of the domains are below $2 \mu\text{m}^2$ in area and a widening of the bias distribution for smaller domains is apparent. The data is normal distributed and is symmetrical to zero bias, in agreement with the prediction of the random-field approach. The standard deviation of the bias distribution can be written as $\sigma = \sqrt{\sum h_i^2}$ (the average bias vanishes), and it is a measure for the average absolute value of the bias. In order to statistically test or reject the hypothesis of a widening of the bias distribution we performed Bartlett's test [18], which tests the hypothesis that

two or more normal distributed samples have the same or different variance. We chose 400 domains each for two statistical samples, one containing the largest, the other the smallest domains. Bias field histograms and Gauss fits of these two statistical samples are shown at the top of Fig. 2 [17]. An increase of the standard deviation from 9.4 Oe for the largest to 14.1 Oe for the smallest domains was found. The test result of 64.7, compared to a much smaller χ^2 critical value of 10.8 at a 99.9% confidence level, means that the hypothesis of equal variance has to be rejected. The increase in variance or width of the bias distribution with decreasing domain size is statistically significant according to Bartlett's test.

The widening of the bias distribution as function of decreasing domain area is shown in Fig. 3. The total distribution was divided into 11 equal-sized groups of 115 domains, of which the standard deviation σ of the bias field is plotted. The bunching of the data points at low area is a result of the dominance of small domains in the distribution. The standard deviation starts at 7.9 Oe for, on average, $3.9 \mu\text{m}^2$ domains and reaches 14.0 Oe for $0.11 \mu\text{m}^2$ domains. Note, that because of the equal group size all data points have approximately equal errors, the value of which can be discerned from the scatter of the bias data at small domain area. More instructive is a plot of the standard deviation of the bias distribution as function of $1/\sqrt{A}$ or the inverse diameter (Fig. 3, inset). The dependence is clearly linear over about a decade in inverse diameter, with a slope of $2.4 \text{ Oe}/\mu\text{m}^{-1}$. The offset at infinite domain area is the result of the coarseness of our bias field measurement ($\pm 5 \text{ Oe}$) and the stochastic nature of the switching of single magnetic domains. The randomness of switching also explains the imperfect cross correlation of 0.7 between bias maps of consecutive loops.

The inverse dependence of the standard deviation of the local bias field with domain diameter is in agreement with the prediction of the random-field model, put forward by Malozemoff [2] and with experimental data and their interpretation by Takano *et al.* on polycrystalline CoO [4]. In contrast to the latter experiment, the origin of the size-dependent bias in this epitaxial film is primarily the magnetic domain structure. Smaller domains statistically have a larger ratio of uncompensated to compensated spins, leading to a widening of the bias field distribution towards smaller domain sizes. This widening appears in our measurement as an increased standard deviation of the bias field distribution for small domains. Our result supports the conjecture of a statistical origin of exchange bias due to uncompensated spins located at magnetic or structural defects, which break the translational symmetry of the crystal. Steps, domain walls, and grain boundaries can produce

such uncompensated spins. Depending on their crystal coordination some spins are strongly anchored in the antiferromagnet and appear as pinned [11, 13]. Over large areas these pinned spins average to zero pinned moment in an unbiased sample and no or little bias is measurable. Small domains, however, have a higher chance of containing a significant surplus of pinned spins pointing in the same direction, which leads to local bias. When a sample is explicitly biased, e.g., by field-cooling, then the center of the bias distribution is displaced from zero bias. A larger bias will be found on a sample containing predominantly small domains, which have, on average, a larger ratio between uncompensated spins and compensated spins.

In summary, we have studied the dependence of the standard deviation of the local bias field distribution with the domain area for Co/LaFeO₃(001). Using x-ray microscopy we have determined microscopic maps of the bias field, domain-by-domain and have statistically analyzed the bias field distribution. The analysis shows a statistically significant increase of the width of the bias field distribution with decreasing domain area and an inverse dependence with the domain diameter. This functional dependence is the result of the stochastic nature of exchange bias.

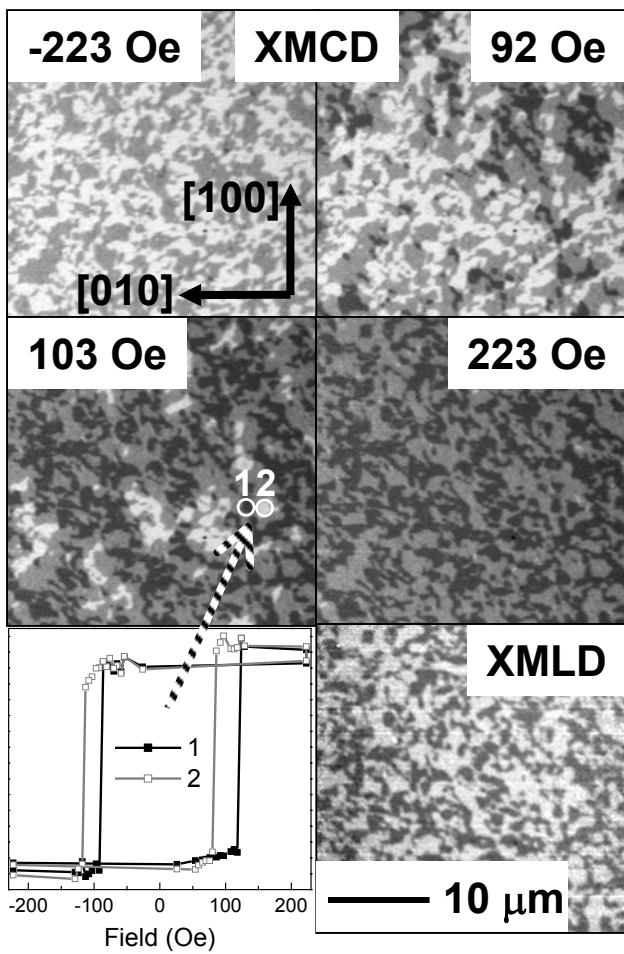
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- [17] Both distributions have a mean bias not significantly different from zero, tested using a t-test, and are fitted well by Gaussians, which are not constrained in width and position.

[18] NIST/SEMATECH e-Handbook of Statistical Methods.

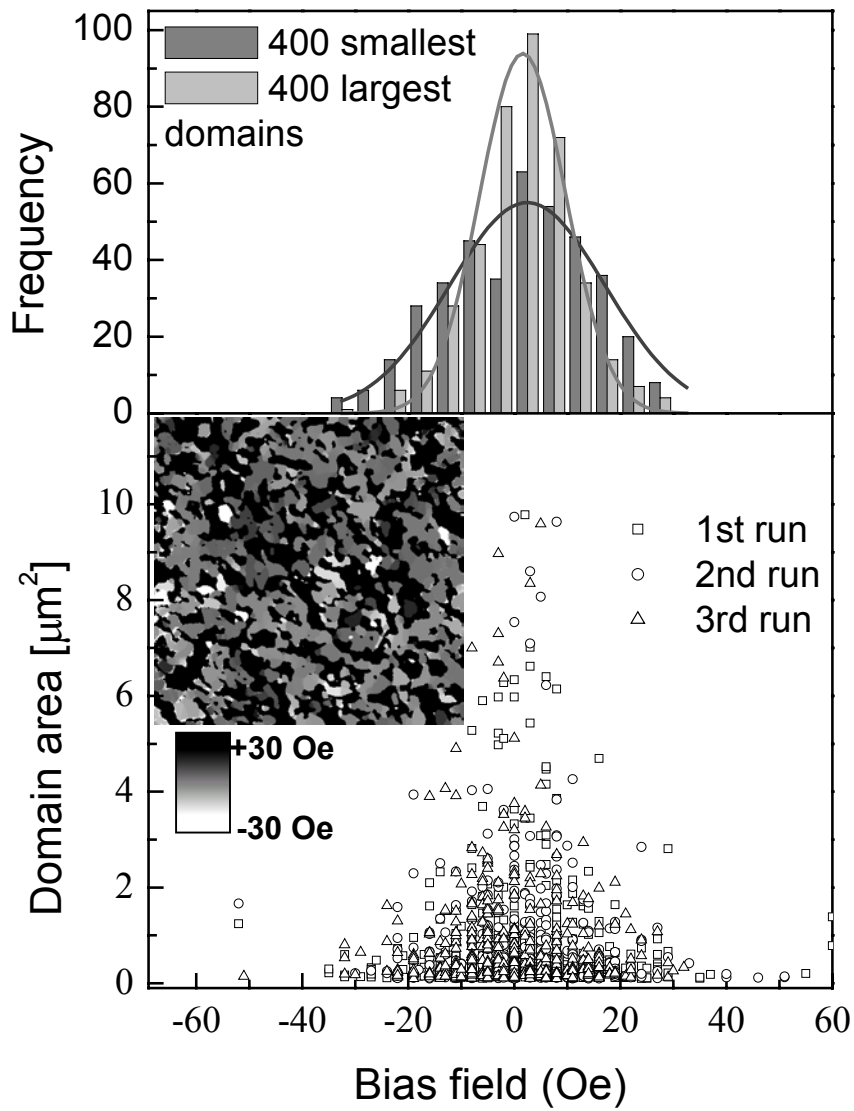
FIG. 1: Top: Co XMCD domain images in remanence, sensitive to the $[100]$ magnetization direction, after applying fields between ± 223 Oe along $[100]$. Bottom right: LaFeO_3 XMLD image using linear polarization along $[010]$. Bottom left: Local remanent hysteresis loops of two domains, showing opposite bias directions. The domains are marked by circles in the domain image.

FIG. 2: Bottom: Domain area as function of bias field of 1266 Co domains from 3 magnetic loops. Inset: Map of the local bias field extracted from local loops. Top: Histogram of domain number as function of bias field for the 400 smallest and the 400 largest domains. Lines: Gauss fits showing larger width of the bias distribution of smaller domains.

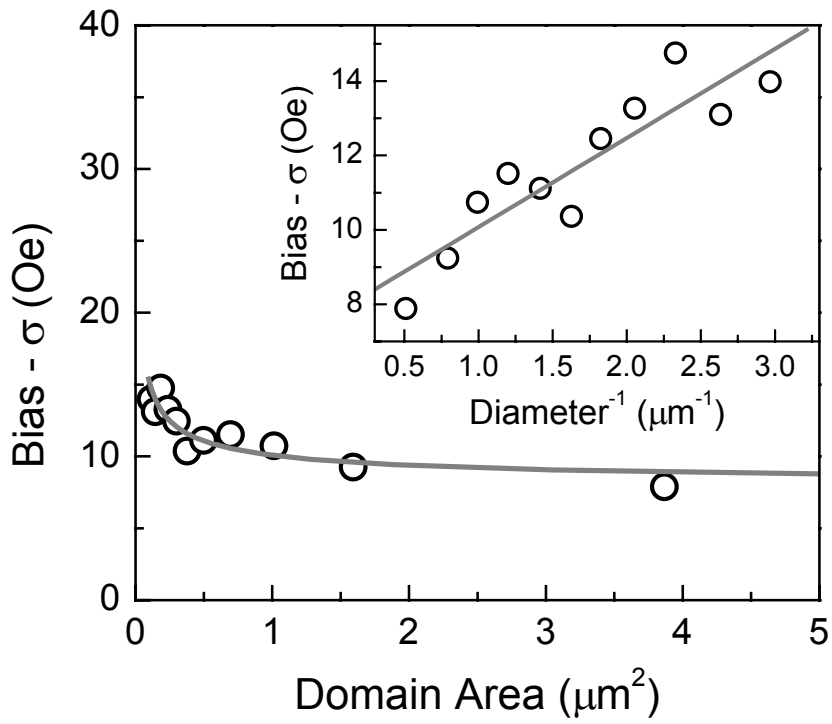
FIG. 3: Bias field distribution as function of domain area. Circles show the standard deviation of the bias field distribution in groups of 115 domains as function of average area. Inset: Same data shown as function of inverse domain diameter. Thick lines (both plots): Fit with a linear function of the inverse domain diameter.



A. Scholl, Figure 1



A. Scholl, Figure 2



A. Scholl, Figure 3