

Solving a Forefront Problem in Materials Science: The Magnetic and Chemical Structure of a Buried Interface

H. Ohldag, J. Lüning and J. Stöhr, SSRL/SLAC

Computer hard drives and other advanced electronic devices depend on layered stacks of magnetic and non-magnetic materials, but researchers don't fully understand why such layered materials exhibit new properties that cannot be predicted from the properties of the individual layers. In a recent publication a team working at SSRL and the ALS describes new methods, based on x-ray spectroscopy and xray microscopy that reveal the magnetic structures at the boundaries between these layers. Their data show that the boundaries are not as clean as previously assumed but a new ultrathin interface layer may be formed by a chemical reaction. The thickness of the interfacial layer is found to change with temperature and this change can be directly correlated with the magnetic properties of the multilayer stack. The work provides the first magnetic images of a buried interface and gives direct experimental evidence for the existence and long-assumed importance of interfacial magnetic spins.



Co (left column) and Ni (right column) high resolution L_3 -edge xray absorption spectra resolving the near edge absorption fine structure (NEXAFS) of the metal (top) and the oxide (bottom). The NEXAFS of Co and Ni atoms within the interface layer (middle) reveal a chemical reaction of Co-oxidation and Nireduction. By choosing an appropriate photon energy and polarization x-ray photoemission electron microscopy (XPEEM) images of the top metal and bottom oxide layer as well as the interface layer have been obtained (middle column)

Spectroscopic Identification and Direct Imaging of Interfacial Magnet Spins,

H. Ohldag, T. J. Regan, J. Stöhr, A. Scholl, F. Nolting, J. Lüning, C. Stamm, S. Anders and R. L. White Phys. Rev. Lett. 87, 247201 (December 10, 2001)

Uncovering a New Layer Tim Palucka Phys. Rev. Focus (December 5, 2001)

An der Grenze gerüttelt Martin Wieser wissenschaft-online (December 12, 2001) The determination of the crystallographic, electronic or magnetic structure of interfaces has remained one of the great challenges in all of materials science. The key reason is the difficulty to detect and isolate the weak interface signature from that of the dominant bulk. This is largely due to the lack of depth specificity of most techniques, impeding the detection of a signal from a well-defined depth, only. For lack of better capabilities scientists have tried to circumvent this problem, often studying the early stages of interface formation with surface science techniques or simply assuming "perfect" interfaces between "bulk" materials. Using advanced soft x-ray spectroscopy and microscopy techniques in conjunction with interface sensitive electron yield detection we are now able to look at buried interfaces and observe that, in reality, interfaces are quite different from model systems.

Modern magnetism is one area where interfaces play a crucial role. Today's high-tech magnetic devices are based on thin film multilayers whose magnetic properties depend on the magnetic coupling and spin transport across interfaces. Examples are giant magnetoresistance structures, spin tunnel junctions, as well as "spintronics" devices based on spin injection. A specific interfacial problem which is of considerable scientific interest and technological importance is the origin of "exchange bias", an effect utilized in many of today's magnetic sensors and memory cells.



Figure 1: Hysteresis loops of a pure ferromagnet (gray) are symmetric. Once in contact with an antiferromagnet of the loop widens (coercivity increase, blue) and when heated above the Néel temperature in a magnetic field shifts in one direction (exchange bias, red).

The exchange bias effect, empirically discovered nearly 50 years ago, is used today to create well-defined ferromagnetic а reference layer in a magnetic device. Natural ferromagnets have a preferred magnetization "easy axis", and an external field can align the spins into either of two equally stable directions along this axis the magnetization loop is symmetric as shown in Fig. 1. When a ferromagnet (FM) is grown on an antiferromagnet (AFM) the exchange coupling between the leads two systems to an increased coercivity of the which ferromagnet, can be viewed as an increased "friction"

to turn the spins around. The ferromagnetic hysteresis loop is still symmetric, indicating two equivalent easy directions. If, on the other hand, the AFM-FM system is grown in a magnetic field or, after growth, is annealed in a magnetic field to temperatures above the AFM Néel temperature, the hysteresis loop becomes asymmetric and is shifted from zero, as shown in Fig. 1. This unidirectional shift is called "exchange bias". There is now a preferred magnetization **direction** for the FM along which it is most easily aligned. The easy alignment direction can serve as a reference direction in a device. It is clear that exchange bias has to originate from the coupling of the spins in the AFM to those in the FM but, because of the magnetic neutrality of the AFM, the coupling has to involve uncompensated spins at the AFM-FM interface. The key to the exchange bias puzzle lies in the determination of the origin of these interfacial spins and their role in coercivity increases and bias.

It is the very difficulty associated with the determination of the magnetic interfacial structure mentioned above, that has impeded the solution of the exchange bias puzzle for more than forty years.



Figure 2: High resolution x-ray absorption spectra (red) for a MgO(100)/NiO(1 nm)/Co(1 nm)/Ru(2 nm) sample, recorded at the Ni L_2 and Co L_3 edges, in comparison to reference spectra taken for the pure metals (blue) and monoxides (green). The black curve is a weighted superposition of the blue and green spectra.

Along come two powerful new magnetism techniques based on soft x-rays, the x-ray magnetic circular (XMCD) and linear (XMLD) dichroism techniques [1]. In conjunction with spectromicroscopy these x-rav techniques have allowed a unique fresh look at the old exchange bias problem and hold the promise to finally solving it. A series of XMCD/XMLD imaging experiments using the Photoemission Electron Microscope (PEEM2) at the ALS previously established the link of the AFM and FM domain structure, including the reorientation of the AFM spins in the vicinity of the interface [2,3]. The latest, just published [4,5], results home in on the allimportant interface. They were obtained by two complementary experiments, high energy resolution (150 meV) soft x-ray absorption spectroscopy in total electron yield mode performed at Beam Line 10-1 at SSRL and high spatial resolution (50 nm) soft x-ray absorption microscopy using the PEEM2 microscope at the ALS.

The SSRL spectroscopy results shown in Fig. 2 demonstrate that a thin Co layer deposited on top of bulk NiO (Co/NiO) contains Ni atoms that are in an environment somewhere between NiO and Ni metal, and Co atoms that are in an environment somewhere between Co metal and CoO. This is explained by an interfacial reaction in which the original NiO is reduced and the original Co is oxidized. A new interfacial layer is formed that we shall call NiCoO_x.

Magnetic images of the Co/NiO sandwich, obtained with the PEEM2 microscope at the ALS, are shown in Fig. 3. The Figure is an artist's rendition, with the original images put together

layer by layer to illustrate the magnetic structures in the NiO and Co layers and the image of the $NiCoO_x$. The antiferromagnetic NiO domain image (bottom: green) was obtained with XMLD spectromicroscopy, tuning the x-rays to the Ni L₂ peaks in NiO (see Fig. 2 top). The ferromagnetic Co domain image (top: blue, white, black) was obtained with XMCD spectromicroscopy, tuning the x-ray energy to the Co metal L_3 peak (see Fig. 2 bottom). The image of the domain structure of the NiCoO_x interface layer (middle: golden, white, black) was obtained with XMCD spectromicroscopy, tuning the x-rays to the Ni L₂ peak energy of 870.5eV (see Fig. 2 top). Since XMCD yields ferromagnetic contrast, the observed domain structure of the interface has to arise from uncompensated Ni spins formed by reduction of NiO. Close inspection reveals that the domains mimic the antiferromagnetic NiO domains below and the ferromagnetic Co domains above and therefore form the bridge between the two. The results indicate that the coercivity and increase in the bias shift in antiferromagnet/ferromagnet sandwiches, illustrated in Fig. 1, originate from the interfacial spins created by a chemical reaction. Further experiments show that in an



Figure 3: Magnetic structure, assembled layer by layer from original PEEM2 images, for a magnetic NiO (green). For the first time the magnetic structure of the interfacial CoNiO₂-like layer (golden), formed by chemical reaction, could be imaged by combining chemical specificity

external magnetic field most of the interfacial spins rotate with the ferromagnet, but they provide an increased rotational drag that leads to the widened magnetization loop. A small fraction of the interfacial spins that has not yet been isolated, is believed not to rotate at all in an external field because the spins are tightly coupled to the antiferromagnetic NiO lattice underneath. These spins are believed to give rise to the exchange bias phenomenon illustrated in Fig. 1. Because of their small abundance, corresponding to a fraction of a monolayer, only, the isolation of their magnetic signal and the determination of their spatial location remain a great challenge for future experiments.

References:

- 1. For more information see http://www-ssrl.slac.stanford.edu/stohr/index.htm
- F. Nolting, A. Scholl, J. Stöhr, J. W. Seo, J. Fompeyrine, H. Siegwart, J.-P. Locquet, S. Anders, J. Lüning, E. E. Fullerton, M. F. Toney, M. R. Scheinfein, and H. A. Padmore, Direct Observation of the Alignment of Ferromagnetic Spins by Antiferromagnetic Spins, *Nature* 405, 767 (2000)
- 3. H. Ohldag, A. Scholl, F. Nolting, S. Anders, F.U. Hillebrecht, and J. Stöhr, Spin Reorientation at the Antiferromagnetic NiO(001), Surface in Response to an Adjacent Ferromagnet, *Phys. Rev. Lett.* **87**, 247201 (2001)
- 4. T. J. Regan, H. Ohldag, C. Stamm, F. Nolting, J. Lüning, J. Stöhr and R. L. White, Chemical effects at metal/oxide interfaces studied by x-ray-absorption spectroscopy, *Phys. Rev.* **B 64**, 214422 (2001).
- 5. H. Ohldag, T. J. Regan, J. Stöhr, A. Scholl, F. Nolting, J. Lüning, C. Stamm, S. Anders and R. L. White, Spectroscopic identification and direct imaging of interfacial magnetic spins, *Phys. Rev. Lett.* **87**, 247201 (2001).

SSRL is supported by the Department of Energy, Office of Basic Energy Sciences. The SSRL Structural Molecular Biology Program is supported by the Department of Energy, Office of Biological and Environmental Research, and by the National Institutes of Health, National Center for Research Resources, Biomedical Technology Program, and the National Institute of General Medical Sciences.