Researchers Glance at Magnetic Surfaces with Synchrotron X Rays

Researchers using resonant x-ray scattering to study surface magnetism may be on the verge of technological advances and of a deeper understanding of magnetism.

The proliferation in recent years of the number of techniques for studying surface magnetism is partly attributable to the increasing importance of surface magnetism in the multibillion-dollar recording industry—in the design of ultrahigh-density storage devices and possible applications of giant and colossal magnetoresistance in metallic thin film multilayers, for example. To many physicists, however, surface magnetism has a more fundamental importance: Bulk magnetism is the quintessential example of long-range ordering, and crystalline symmetry—on which theories of bulk magnetism depend—is broken at the surface. As such, surface magnetism provides physicists with an opportunity to investigate the mechanisms of magnetic ordering between the well-understood limits of atomic and bulk-matter magnetism.

In the past, a broad variety of techniques—including spectroscopy, atomic force and scanning tunneling microscopy and low-energy electron diffraction (LEED)—have been applied with some success to studies of magnetic energetics and ordering on (but not below) surfaces. Neutron scattering has been a particularly powerful tool for studying magnetism in bulk matter and in magnetic multilayers.[1] At present, however, only synchrotron x-ray sources offer beams bright enough to resolve the detailed atomic structure underlying surface magnetism over length scales of micrometers and to penetrate below the surface to investigate the transition from surface to bulk magnetism.

At first glance, x rays would seem to be unsuitable for the job. For 10 keV x rays, conventional magnetic scattering is five to six orders of magnitude weaker than scattering by electric charges, and surface scattering is suppressed by a similar factor relative to bulk scattering. However, calculations by Martin Blume in 1985 (Brookhaven National Laboratory) suggested that tuning the energies of incident x rays to atomic absorption lines of elements in the sample could substantially enhance magnetic scattering cross sections. Experiments at the Cornell High-Energy Synchrotron Source (CHESS) and subsequent calculations found that resonances in actinide and rare-earth elements could be especially large. This discovery allowed researchers to take full advantage of the brilliant beams then available at synchrotron x-ray facilities. Further experiments on bulk matter and even thin films demonstrated the feasibility and utility of the technique, which eventually came to be called x-ray resonant magnetic scattering. XRMS had the additional advantage of mapping only the element to whose absorption line the x-ray source was tuned. However, surface magnetic scattering presented problems because of the weakness of surface scattering and because most surface magnetic scattering experiments require ultrahigh-vacuum chambers and, in many cases, cryogenic equipment.

Moreover, diffraction patterns contain contributions from a variety of sources, including the distributions of charge within the bulk and surface of the sample, as well as the sample's surface topography. Using XRMS to study aspects of surface magnetism requires isolating the magnetic signal from these other contributions so that one can extract the information therein. Recently, researchers at Brookhaven's National Synchrotron Light Source (NSLS),[2] the European Synchrotron Radiation Facility (ESRF)[3] in Grenoble, France, and the University of Wisconsin Synchrotron Radiation Center[4] (Stoughton, Wisconsin) did just that by combining XRMS with x-ray scattering techniques commonly used for surface studies.

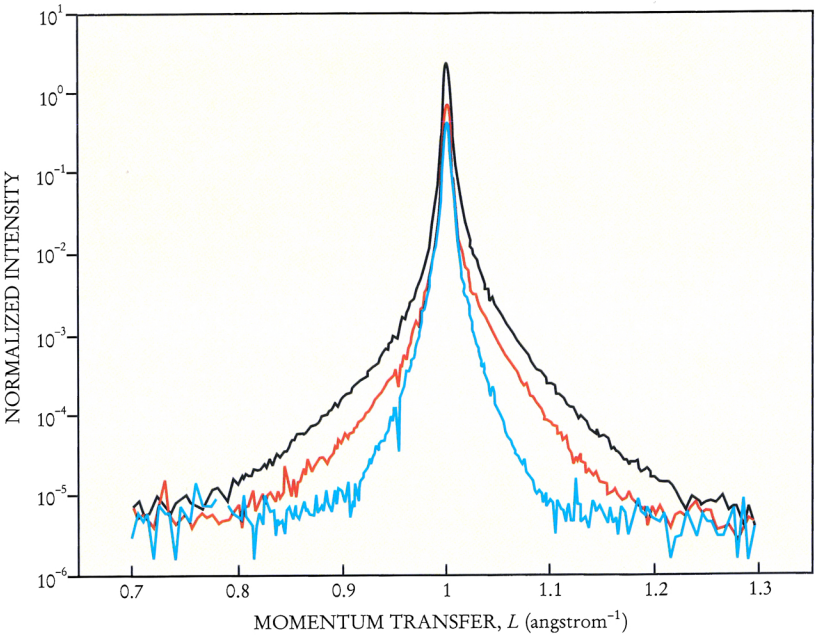
Scratching the surface, and below

At Brookhaven and Grenoble, researchers combined XRMS with grazing incidence diffraction (GID), in which x rays incident at near the critical angle for total external reflection scatter from a flat crystalline face of a sample. GID ensures that one probes only the surface and near-surface of the sample, and it has the additional advantage that one can vary the depth to which one probes the sample by varying the angle at which x rays are detected.

At NSLS, Gavin Watson (University of Maryland at Baltimore), Gibbs (BNL), Gerard Lander, Hans-Joachim Matzke (Institute for Transuranic Elements in Karlsruhe, Germany), Bruce Gaulin (McMaster University in Hamilton, Ontario) and Walter Ellis (Los Alamos National Laboratory) used grazing-incidence XRMS to examine surface effects in the magnetic order–disorder transition of the antiferromagnet, UO_2. They chose UO_2 because uranium has large resonances and because UO_2's relative chemical inertness allowed them to run without a vacuum chamber. To first order, the NSLS group controlled the contribution of surface topography to the diffraction by obtaining the flattest, smoothest sample possible. Their UO_2 sample was cut and polished along a particular crystal face and then annealed at 1400 °C to repair surface damage. In fitting their diffraction data, the group further compensated for surface topography by allowing for a slight miscut along the crystal face (0.25°) and for a thin layer of impurities at the sample's surface.

Because neighboring magnetic moments in an antiferromagnet (such as UO_2) are aligned antiparallel to one another, the sample's magnetic structure differs from its atomic structure, and portions of the x-ray diffraction pattern result entirely from magnetic scattering. By measuring the x-ray intensities scattered in different directions in the purely magnetic portions of the pattern as the sample was heated toward the antiferromagnetic order–disorder–transition, or Néel, temperature, T_N, Watson and his collaborators could observe the effect of temperature on the magnetic ordering at surface (5 nm), near-surface (12 nm) and bulk (85 nm) depths. The researchers found that, unlike bulk UO_2, which exhibits a discontinuity in the magnetization at T_N, the surface layers begin to become disordered at lower temperatures but exhibit no discontinuous transition to magnetic disorder at T_N.

Watson and his collaborators point out that order–disorder transitions in metallic alloys and other systems exhibit similar suppression of discontinuities near the surface and that this suppression might be understood in terms of surface-induced disorder (SID)—a concept introduced by Reinhard Lipowsky in 1982, in which surface layers become disordered below the normal transition temperature. Although the NSLS results suggest that magnetic disordering in UO_2 is more complex than previous examples or theoretical treatments of SID, additional measurements much closer to T_N will be needed to establish whether the behavior of UO_2 can be understood in terms of SID, according to David Landau (University of Georgia, Athens, Georgia), who, along with Kurt Binder (University of Mainz, Germany) and



APPROACHING MAGNETIC DISORDER. When an x-ray beam is magnetically reflected from a magnetically ordered UO_2 sample, the x rays emerge from the surface in a narrow cone, the width of which gives a measure of the range of momentum transfers that occur in the scattering process. As the sample is heated toward its bulk antiferromagnetic order–disorder transition temperature (T_N = 30.2 K), the cone broadens, indicating an increase in the width, ξ, of the interface between the disordered surface and the ordered bulk. In so-called reciprocal space—where transferred x-ray momentum is proportional to an inverse distance, L—the widening of the interface causes successive intensity curves to narrow as the sample is warmed from 10.2 K (black) to 26.9 K (red) to 29.6 K (blue). Fits to the magnetic reflectivity show that ξ increases from one to four spacings of the crystalline lattice as the temperature increases from 10.2 K to 26.9 K. (Figure courtesy of Gavin Watson, University of Maryland, Baltimore County.)

Werner Schweika (Institute for Condensed Matter Research, Jülich, Germany), has recently published Monte Carlo studies[5] of SID and related phenomena.

The ESRF group, which consisted of Salvador Ferrer, Pablo Fajardo, Jesus Alvarez, Xavier Torrelles (all of ESRF), Francois de Bergevin (Crystallography Laboratory, also in Grenoble), Erik van der Vegt (FOM Institute in Amsterdam) and Victor Etgens (University of Paris), prepared the surface of their Co_3Pt sample in a manner similar to that used by the NSLS team (although the Co_3Pt sample was etched as well as annealed). The ESRF experiment was conducted in ultrahigh vacuum, and surface cleanliness was monitored by Auger electron spectroscopy.

By fitting nonresonant x-ray scattering data, the ESRF group modeled the structure of their sample, allowing the ratio of platinum to cobalt in their model to vary in the top three layers. In results that were consistent with previous LEED studies of the same alloy, the best fit found the cobalt to be depleted in the top layer by a factor of two, to be enhanced in the second layer by a factor of three and to be nearly consistent with the ideal stoichiometric ratio in the third layer. In the topmost layer, the Co and Pt atoms were also found to be displaced by a few hundredths of a lattice spacing from the positions they would have occupied in bulk. Because the ferromagnetic ordering of Co_3Pt has the same spatial dependence as the alloy's atomic structure, the magnetic diffraction patterns coincide with those resulting from the distribution of charges in the sample. However, reversing the magnetization in the sample reverses the phase of the magnetic contribution to the diffraction pattern. Therefore, one can extract the magnetic contribution by taking the difference between the x-ray intensities for opposing field polarizations. For convenience, this difference is divided by the sum of the two intensities, resulting in an "asymmetry ratio."

To investigate how the surface affected the magnetic moment of the Pt atoms there, the experimenters assumed their sample had the structure they had determined previously with nonresonant x rays and fit the asymmetry-ratio data to determine the most probable value of the Pt magnetization. Because the ESRF x-ray beam is more brilliant than that at NSLS, the ESRF researchers were able to look at x-ray scattering from a single layer of Pt atoms (about 0.3 nm). The best fit to these data found the Pt magnetization in the top layer was less than that in bulk by a factor of two. The fact that the Co content in the top layer was also depleted by a factor of two suggests that, as expected, the magnetization of Pt atoms is induced by neighboring Co atoms.

Taking it nice 'n rough

Although the success of the measurements at NSLS and ESRF depended critically on controlling and modeling surface roughness, at Wisconsin the surface roughness was the signal. There James MacKay, Christian Teichert, Don Savage and Max Lagally combined XRMS with x-ray reflection. Just as one can tell whether a metal plate has been polished by whether reflections in it are sharp or diffuse, the ratio of the intensities of reflected x rays in the diffuse tail and peak of the reflected distribution provides a measure of surface roughness. MacKay and his collaborators compared this ratio for resonant and nonresonant x rays to obtain a comparison of magnetic roughness—the distribution of magnetic moments over a surface or interface—to the actual topographic roughness for surfaces and interfaces in very smooth, sputter-deposited cobalt films and cobalt–copper multilayers. Somewhat paradoxically, they found that the magnetic moments were distributed over the interfaces more smoothly than the atoms that contained them.

The Wisconsin researchers suggest that interface roughness causes magnetic moments at the interface to be less strongly coupled to the film's bulk magnetization, especially where the roughness is sharpest and the magnetic atoms are relatively isolated. If this explanation turns out to be true, then surface magnetic moments should be more easily decoupled at higher temperatures, and XRMS measurements as a function of temperature should reveal such decoupling.

Latest results

There is every reason to suspect that the results outlined above will be only the first in a long line of studies of surface magnetism using resonant x rays. At NSLS, Watson and his collaborators are continuing their studies

on UO_2 to try to determine how rapidly disorder descends from the surface into the sample as a function of temperature. Lipowsky's theory of surface-induced disorder predicts that the depth to which disorder extends will diverge logarithmically as one approaches T_N from below. Monte Carlo simulations of SID by Landau and his collaborators suggest a similar dependence. The NSLS team also hopes to see whether different crystal faces of UO_2 behave similarly to that already investigated.

Ferrer and his collaborators at ESRF have switched their attention from Co_3Pt alloy to cobalt thin films with from 2 to 12 atomic layers deposited on a particular crystal face of platinum. Using XRMS, they have found that the magnitude of the induced magnetic moment in the platinum atoms depends on both the thickness and the perfection of the cobalt film. They also have found that the thickness dependence of the moment resembles the thickness dependence of the ferromagnetic order–disorder–transition, or Curie, temperature of the film. Moreover, magnetization is induced in only the topmost layer of platinum atoms.

According to Lagally, Monte Carlo simulations of how surface roughness affects surface magnetism (recently completed by the Wisconsin team) may help to explain his group's roughness studies. Moreover, because even polished surfaces have some roughness, the Monte Carlo Studies may also be relevant to the surface-induced magnetic disordering seen at NSLS. The Wisconsin group is also trying to determine whether temperature-dependent measurements of diffuse XRMS reveal evidence that surface roughness weakens the magnetic coupling of surface atoms to the bulk. Future projects include studies of how surface roughness and film thickness affect the fraction of magnetic moments that participate in giant magnetoresistance, and studies of the magnetic interactions of magnetic thin films and multilayers grown on arrays of quantum dots and quantum wires of very well-defined, periodic roughness. **RAY LADBURY**

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