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STUDIES OF MAGNETISM AT THE NATIONAL SYNCHROTRON LIGHT SOURCE

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NATIONAL SYNCHROTRON LIGHT SOURCE

STUDIES OF MAGNETISM AT THE NSLS

Although magnetism was discovered more than 2,600 years ago, scientists have only begun to understand the phenomenon during the last 70 years. And, it is just within the last decade that synchrotron radiation has been used as a probe to explore magnetic materials on the atomic scale.

Today, research in magnetic materials is crucial for developing state-of-the-art magnetic recording devices for the \$40 billion recording industry. Magnetic thin films, just a few atomic layers thick, are needed in both the storage media and the recording heads of all popular recording devices, including tape recorders and comput-

At beam line X13 at Brookhaven's National Synchrotron Light Source. (from left) Jerome Hastings, Peter Siddons, Erik Johnson and Chri-Chang Kao align the high-vacuum reflectivity apparatus

The world's foremost facility for scientific research using x-rays, ultraviolet and infrared radiation is operated by the National Synchrotron Light Source Department. In a single year, a total of about 2,600 researchers from almost 450 institutions perform experiments at the world's largest source of synchrotron light. Guest researchers often work in collaboration with staff scientists at the Light Source, conducting a wide range of innovative experiments in physics, chemistry, biology, materials science and various technologies.

ers. They are also a crucial component in specialized recorders for collecting data in research, industrial and military applications, and they are used in devices for transmitting data from satellites and spacecraft.

Since the 1980s, several new techniques for probing magnetism have been developed at Brookhaven's National Synchrotron Light Source (NSLS). These innovative methods allow researchers to study the phenomenon in more detail and with better accuracy and precision than ever before. So far, researchers from approximately 25 institutions — from BNL and IBM to the University of Munich in Germany — have performed studies on numerous magnetic materials at the NSLS.

X-RAYS: POWERFUL MAGNETIC PROBES

Traditionally, neutron probes have been used to gain information on the atomic structure and interactions in magnetic materials. In the 1970s, researchers from AT&T Bell Laboratories were the first to theorize that magnetic structure also could be investigated using x-rays.

Few experiments were performed at that time because x-ray sources were not powerful enough to be used routinely as atomic-scale probes. But, in the 1980s, with the commissioning of such high-brightness synchrotron sources as the NSLS, researchers began to use x-rays to gain new data on magnetic materials.

On an atomic scale, a material's magnetism is attributed to the orbital motions and spins of electrons in its atoms. This is true of any magnetic material, including about one-fifth of the elements in the periodic table that are naturally magnetic, and the

thousands of magnetic alloys and compounds.

The nucleus of an atom is surrounded by electrons that both spin, or rotate, on their own axis, and travel in an orbit around the nucleus, much as the earth spins on its axis and travels in an orbit around the sun. A material's magnetic moment is the sum total of all its electrons' orbital motions and spins.

Certain properties of magnetic materials that are important to their functioning can best be investigated with x-rays. In addition, extremely small samples can be studied using x-rays, while studies that use neutrons as probes, such as those conducted at Brookhaven's High Flux Beam Reactor, require relatively large samples.

Other advantages of using x-rays to probe magnetic materials are that they provide high resolution and are therefore sensitive to even slight structural distortions. Further, the x-ray beam polarization and energy can be chosen to meet the requirements of a specific experiment.

Polarization involves the alignment of x-ray beam components, which can form a linearly or circularly polarized beam. In general, when x-rays are directed onto a sample, the beam is scattered. By measuring the intensity and polarization of the scattered x-rays, researchers can learn about the spin and orbital contributions to a sample's magnetic moment.

The ability to perform these measurements will be significantly enhanced when BNL and Argonne National Laboratory complete their design and construction of a polarized wiggler at NSLS beam line X13. Scheduled for completion in 1994, this combined permanent magnet/electromag-

netic device will provide circularly polarized x-rays, which are particularly sensitive to the spin of electrons in magnetic materials.

In 1987, Brookhaven, AT&T Bell Laboratories, Cornell University and ESRF researchers discovered another phenomenon that expanded the usefulness of x-rays for probing magnetic materials. While tuning a polarized beam to different energies in an x-ray scattering experiment, they noticed that there was a dramatic increase in the scattering signal when the incident x-ray energy was tuned to the absorption edges of the sample — the energies at which the absorption of x-rays markedly changes. Consequently, the magnetic properties of materials such as thin films, or those with small magnetic moments, can be probed at synchrotron sources.

VARIED TECHNIQUES, DIFFERENT DATA

At the NSLS, materials as diverse as the rare-earth metal holmium and multilayers of iron-chromium-iron are studied using seven different techniques (see chart). To investigate fully various structural and electronic properties of magnetic materials, often three or four techniques might be used on a single sample.

Scattering and magnetic reflectivity are x-ray techniques that reveal magnetic structure — the orientation of the magnetic moments of the atoms in a material. In ferromagnetic materials, the magnetic moments spontaneously organize in a parallel direction, while in antiferromagnetic materials, magnetic moments are aligned antiparallel to each other. This structural information is important for determining possible applications.

Each magnetic material has its own critical temperature below which the

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In magnetic reflectivity, also known as soft x-ray magneto-optical Kerr effect, x-rays hit the multilayer sample and are reflected onto the detector, which measures the reflected intensity of light as a function of photon energy, polarization, incident angle and direction of the magnetic field. Element-specific magnetic moments and their distribution can be determined using this technique.

Two circular polarized reflectivity curves measured from a cobalt containing multilayer near the Co $L_{2,3}$ absorption edges, using the experimental configuration shown below, left. The curves were measured with the spin of the incident photons parallel (solid) and anti-parallel (dashed) to the sample magnetization. The incident angle was 25° . Asymmetry ratios (circles) are defined as the difference divided by the sum of the two reflectivity curves.

magnetic moments are ordered, either ferro- or antiferromagnetically. Scattering is also the best x-ray technique for studying the details of this

ordering as the temperature is varied near the critical temperature.

Just above the critical temperature so-called critical scattering is

observed. This results from coherent fluctuations in time of the local order of the magnetic moments, and the average size of the fluctuations di-

Gavin Watson (left), Tom Thurston, John Hill and Doon Gibbs prepare to study the spin-density-wave state of chromium on beam line X22C at the NSLS. This is part of a larger program to investigate the magnetic properties of materials by x-ray scattering.

verges as the critical temperature is approached. The rate of divergence can depend on whether the magnetic moments are near to the surface of a crystal rather than well inside the crystal. Studies of the critical scattering provide important tests of our understanding of the onset of magnetism.

Below the critical temperature the rate at which all of the moments become fully aligned or at which the moment direction can be reversed under the influence of an external magnetic field are of technological as well as of fundamental importance. Magnetic x-ray scattering is developing as an important new tool to study this alignment as a function of temperature and magnetic field.

Except for nuclear resonance scattering, a technique that includes the excitation of nuclei, the remaining techniques — spin polarized photoemission, magnetic circular dichroism, the Faraday effect, and spin-resolved x-ray absorption fine structure — are all forms of x-ray spectroscopies, which involve the excitation of electrons. Depending on the magnetic properties of the sample, this excitation is manifested as x-ray absorption or emission of radiant energy. These techniques give insight into the spin dependence of the energy spectrum of magnetic materials.

X-ray spectroscopies enable researchers to look in detail at the separate contributions made by the electron orbit and electron spin to a material's magnetic moment — data

that are important for understanding how a magnetic material functions. These techniques are complementary to structural determinations, since they allow scientists to delve into the underlying reasons why magnetic structures evolve in particular ways.

Over the last decade, the NSLS has been a focal point for developing pioneering techniques to probe magnetic materials. Most likely, new techniques will emerge that provide more insight into magnetism — the industrially valuable but not-completely-understood phenomenon discovered over two-and-a-half millennia ago.

Spin-resolved x-ray absorption fine-structure measurements show the difference in photon absorption between spin-up and spin-down states in manganese fluoride. This technique allows researchers to determine the distribution of magnetic states in a material.

SPIN POLARIZED PHOTOEMISSION — Probes thin films of magnetic materials, just a few atoms thick, and multilayers, which are thin films sandwiched together. An intense flux of photons excites electrons in the sample, and they come out spinning, either up or down. When the electrons hit a gold atom target, the direction of their spin can be determined to reveal information about the magnetic properties of the sample.

05

MAGNETIC CIRCULAR DICHROISM (MCD) — Used to determine, with element and site specificities, the electron orbital and spin contribution to the magnetic moment of transition metals and rare earths. MCD is the difference in the absorption of light by magnetic systems using circularly polarized light in which an electric field rotates in either a left or right direction. The absorption of soft x-rays depends on the polarization of light and the relative direction of the sample's magnetic moment.

048

FARADAY EFFECT — Describes the rotation of the plane of polarization of an x-ray beam as it passes through a magnetic sample. Measures essentially the same quantities as MCD (see above), but this technique does not need circularly polarized x-rays, and, without requiring correction factors, is capable of providing absolute measurements. The technique is limited to measurements in the 6 to 12 kilo-electron-volt range, but this covers most magnetic elements of interest.

X120, X270

MAGNETIC REFLECTIVITY — Similar to MCD (see above); measures the magnetic moment of solids, alloys, thin films or multilayers. It also measures the distribution of magnetic moments in the sample. Photons hit the sample and are reflected onto a detector, which measures the reflected intensity of light as the photons' energy and angle are changed. The reflected intensity allows researchers to determine the orientation and distribution of the sample's magnetic moments.

048, 0130A, 015

SPIN RESOLVED X-RAY ABSORPTION FINE STRUCTURE — Measures energy of unoccupied electronic orbits of a magnetic material. When photons hit the sample, electrons are knocked out of occupied orbits into unoccupied ones. By looking at different x-ray emission energies, researchers can separate spin-up and spin-down electronic states. Magnetic materials show energy differences between these states.

X25

SCATTERING AND RESONANT SCATTERING — Used to determine magnetic structure of surfaces, multilayers and bulk solids. X-rays hit the sample, scatter, and an analysis of the intensity of scattered x-rays gives the positions of magnetic moments in the material. When the x-ray energy is tuned near the excitation energy — the energy at which electrons move from one orbit to another — the magnetic scattering signal becomes much larger. This effect is called resonant scattering.

X14, X16, X20,
X22, X23, X25

NUCLEAR RESONANT SCATTERING (MÖSSBAUER EFFECT) — Measures the splitting of nuclear transitions, or changes in the energy states of a nucleus, in certain isotopes. Through the size of this splitting, the strength of the magnetic field at the nucleus of the isotope — for example, iron-57 — is measured. Earlier Mössbauer experiments traditionally used radioactive sources. The recent use of synchrotron radiation has many advantages: Samples in confined environments, such as high-pressure cells, can be used; field directions as well as magnitudes can be determined.

X12, X14, X25, X27

EXAMPLES

A large collaboration is studying the surface magnetism of thin films, such as cobalt-copper-cobalt multilayers. These films may be used in magnetic memory technologies, including cassette tapes, compact discs and computer storage discs.

The technique is used to probe the magnetic structure of a wide variety of systems, including surfaces, thin films, alloys, ferrites and proteins. For example, AT&T Bell Laboratories researchers found that a film less than one-atom thick of chromium and iron is magnetic and its magnetic moment is antiparallel with that of iron.

BNL and Ames Laboratory researchers measured the Faraday effect of holmium in a holmium-iron compound in order to understand the nature of the electronic transitions responsible for magnetism to occur in it.

BNL, together with AT&T and the Naval Research Laboratory, is studying various cobalt thin-film multilayers to understand the alignment of the magnetic moments and their relative orientation within the various layers of the sample. These multilayers are potential candidates for future recording heads in computers.

BNL researchers used a manganese fluoride sample to develop this technique. At room temperature, they made spin-resolved measurements of the material. As shown in the figure on page 37, there is a clear difference between the states with spin-up and spin-down.

Many groups have studied the evolution of magnetic order as the ordering temperature is varied. Above the ordering temperature (T_C), critical fluctuations are observed. Below T_C , the rate at which the average magnetic moment increases with decreasing temperature gives insight into the ordering process.

BNL, Cornell University and SUNY Stony Brook researchers found changes in the magnetic field of iron as its temperature changed. As the temperature increased, the intensity of the detector's signal oscillated, and the oscillations became slower as the field became weaker.

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Argonne National Laboratory; AT&T Bell Laboratories; BNL; IBM; National Institute of Standards and Technology (NIST); SUNY Stony Brook; U. of Texas

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BNL; Cornell U.; SUNY Stony Brook; U. of Hamburg, Germany; U. of Munich, Germany; Old Dominion U.

*Cover Figure Top:
Resonant Magnetic X-ray Scattering
Study of Uranium Arsenide.*

The large resonant enhancement of the intensity of an anti-ferromagnetic reflection in UAs at the M_V , M_{IV} and M_{III} absorption edges was measured on beamline X14A by a team from AT&T, Institut Laue Langevin, ORNL and ETH (Zürich). The open diamonds are the observed intensity (corrected for detector efficiency and absorption in the flight path) times the absorption coefficient. The solid curve through the data is a fit to the coherent sum of three quantum-mechanical dipole oscillators.

Inset: expanded view of the M_{III} edge.

*Cover Figure Bottom:
Experimental Verification of X-ray
Magnetic Circular Dichroism
Sum-Rules.*

Magnetic circular dichroism (MCD) spectra $\sigma^+ - \sigma^-$ of Fe, Co and Ni thin films at the $L_{2,3}$ - absorption edges have been recorded by an AT&T Bell Labs and NRL group using transmission x-ray absorption spectroscopy (XAS) on beamline U4B. σ^+ and σ^- are the circular polarizations with positive and negative helicity, respectively. By eliminating the experimental uncertainties suffered by other XAS techniques, these data provide a stringent test for the X-ray MCD sum-rules. The orbital magnetic moments derived from the sum-rules were found to be in good agreement with those obtained from Einstein-de-Haas gyromagnetic ratio measurements, but in poor agreement with values deduced from neutron scattering experiments.

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