Using Synchrotron Light for Magnetic Scattering

Although x-rays and neutrons provide equally valuable ways to probe the structure of materials, neutrons have traditionally had a leg up in investigations of magnetic materials because of the neutron's magnetic moment. Nonetheless, x-rays do weakly interact with magnetic moments in solids, and a very patient researcher would eventually collect useful data with a conventional x-ray source.

The newest sources of synchrotron radiation generate such intense fluxes of x-rays, however, that it is possible to compensate for the low magnetic scattering power with a high-intensity source. At the March Meeting of the American Physical Society (APS),* David Moncton and Doon Gibbs of the Brookhaven National Laboratory demonstrated that, not only is it possible to compensate, but x-rays have some advantages in the higher resolution possible and in being sensitive to small lattice distortions.

X-rays are also able to deal with small samples, and eventually it may be possible to look at surfaces and other two-dimensional systems, which provide the focus for the study of phase transitions, a primary thrust of condensed matter physics.

Gibbs, Moncton, and their Brookhaven colleagues Kevin D'Amico and Jakob Bohr did their synchrotron experiments at the Stanford Synchrotron Radiation Laboratory, which provides exceptionally intense x-ray beams by means of so-called wiggler magnets. The object of the study was holmium, chosen because its large magnetic moment was expected to help in maximizing the signal. Holmium's magnetic structure arises from electron spin and had been examined earlier in neutron scattering studies by groups at Oak Ridge National Laboratory and at Argonne National Laboratory. But the x-ray experiment, owing to the higher resolution, turned up some new findings.

Holmium has a hexagonal close packed crystal structure, which gives the usual x-ray diffraction pattern comprising intense spots (Bragg peaks) with a less intense halo that arises because of lattice vibrations (thermal diffuse scattering). The magnetic scattering in holmium shows up as sharp satellite peaks in the diffuse scattering halo.

Above 131K, holmium is not magnetic. Below that temperature, according to the earlier neutron scattering results, it is what physicists call a spiral antiferromagnet. Within each basal plane of the hexagonal structure, the spins are all aligned, but the orientation from plane to plane rotates by an angle that depends on the temperature.

The magnetic structure is said to be commensurate with the crystal structure if the spin alignment in a basal plane rotates through an angle so that the alignment returns exactly to its starting orientation in an integer number of planes. With a rotation of 60 degrees, for example, the alignment would repeat every six basal planes. Otherwise, the structure is said to be incommensurate.

In the last few years, researchers have found in different physical systems that nominally incommensurate structures sometimes actually comprise regions or domains having a commensurate structure that are separated by narrow regions that are highly incommensurate called "discommensurations."

In brief, the increased resolution of x-ray scattering due to synchrotron radiation enabled the Brookhaven group to show the existence in certain temperature ranges of discommensurations in the magnetic structure of holmium. Bohr, who was visiting from the Risø National Laboratory in Roskilde, Denmark, came up with the key idea that the group used to devise a model for the origin of the discommensurations.

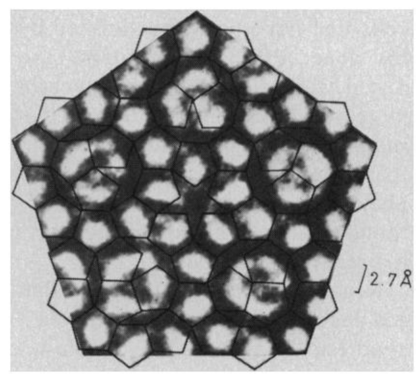
Using this model, it was possible to understand the origin of another satellite peak, which appeared only over a limited temperature range and was not seen by neutron scattering. It turned out that the peak is due to a distortion of the holmium crystal structure itself by the spin discommensurations. The Brookhaven group verified that this satellite was not due to magnetic scattering by analyzing the polarization of the scattered radiation, which is different for magnetic and normal scattering. This was possible because synchrotron light is naturally highly polarized.

*American Physical Society 1985 March Meeting, Baltimore, Maryland, 25–29 March 1985

Doing Crystallography in Six Dimensions

Even the most precocious child cannot array regular pentagons of the same size on a table in such a way that they cover the surface with no gaps showing through. The same problem with objects having fivefold symmetry persists in three dimensions. As crystallographers discovered long ago, one cannot fill space with icosahedrons. Hence there are no crystals with this symmetry.

Or at least there were none until Dan Shechtman and Ilan Blech of the Israel Institute of Technology (Technion) in Haifa, Denis Gratias of the Center for Metallurgical Chemistry (CECM) in Vitry, France, and John Cahn of the National Bureau of Standards in Gaithersburg, Maryland, astounded quite a few scientists with their publication last November describing a metal alloy with just that symmetry, which has been dubbed the icosahedral phase.*



Penrose tiling

High-resolution transmission electron micrograph of the Al_6Mn icosahedral phase showing fivefold symmetry and an overlay of a Penrose tiling pattern. [*Source: L. Bursill, Arizona State University*]

The material of composition 86 percent aluminum and 14 percent manganese differed in another striking way from convention. While it clearly displayed long-range five-fold orientational order in electron diffraction patterns, it could have none of the translational symmetry or periodicity ordinarily required for sharp diffraction spots in the pattern.

Diffraction patterns exhibiting fivefold symmetry have been seen before but have been explained in terms of

*D. Shechtman, I. Blech, D. Gratias, J. W. Cahn, *Phys. Rev. Lett.* **53**, 1951 (1984).

crystallographic defects. What is true in two and three dimensions, however, need not be so in higher dimensional spaces. Of the models that take the diffraction data at face value and try to explain it, one of the most intriguing is that the observed structure is a projection in three dimensions of a six-dimensional object that exhibits both fivefold rotational symmetry and translational peroidicity. At the APS meeting, Gratias explained how this might work.

The simplest case is that of modulated structures, which have a property called quasi-periodicity. In one dimension, the positions of the points in the structure may appear to occur randomly, but they actually occur in a predictable way determined by two characteristic lengths. The two lengths suggest recourse to a two-dimensional space. In particular, consider a two-dimensional cubic lattice. Draw a line through the lattice at some arbitrary angle, and retain only those points within a certain distance of the line. The projections of these lattice points on the line comprise a quasi-periodic sequence.

Generalizing this procedure to higher dimensions may stretch one's powers of visualization but is a straightforward application of linear algebra. To obtain a structure in three dimensions, one simply slices an n-dimensional object with a three-dimensional hyperplane.

Such a procedure never preserves all the rotational symmetry of the higher dimensional object, but it can retain a few symmetries that depend on the direction of the slicing. Recently, three groups (Michel Duneau and André Katz of the Ecole Polytechnique, Palaiseau, France; Veit Elser of AT&T Bell Laboratories; and Shechtman, Gratias, Richard Portier of the CECM, and Cahn) have shown that the icosahedral phase can be recovered from a simple cubic lattice in six dimensions by cutting the lattice with a three-dimensional hyperplane.

The main symmetry axes of the icosahedral phase show twofold, threefold, and fivefold rotational symmetry, and these are all retained in the cut through the six-dimensional cube, as evidenced by the agreement between theoretical and experimental diffraction patterns.

While the six-dimensional cube is translationally periodic, the projection of it in three dimensions exhibits a three-dimensional version of the quasi-periodicity known as Penrose tiling, after the British physicist Roger Penrose, who several years ago devised a way to cover a flat surface with a pattern having fivefold symmetry. Recent high-resolution electron microscope images of the icosahedral phase, such as the one shown, which was made by Les Bursill and Peng Ju Lin at Arizona State University, dramatically confirm the earlier diffraction evidence for this kind of quasi-periodicity.

There are models for the icosahedral phase other than this one. But the prospect that other problems in crystallography may be solvable by recourse to higher dimensional spaces makes it interesting, indeed.

Spatial Quantization in Three Dimensions

The energy of an electron roaming freely through an infinite, empty space is not quantized but is continuously variable. However, confining the electron causes the energy to become quantized in discrete levels, whose separation increases as the size of the region to which the electron is confined decreases.

A crystalline solid, while not an exact analog of the elementary "particle in a box" problem because of the presence of the positively charged atomic nuclei, exhibits a similar behavior. In semiconductors, for example, the valence or bonding electrons and the conduction electrons that carry electrical current are free to roam through the crystal. But the energy levels of these electrons are so closely spaced that they form nearly continuous valence and conduction bands because the walls of the box (the faces of the crystal) are so far apart.

With state-of-the-art microfabrication techniques, however, it has become possible to make solid structures with exceptionally small dimensions. At the APS meeting, Mark Reed of the Texas Instruments (TI) Central Research Laboratories in Dallas reported that he and several TI coworkers have made what they call quantum dots, which are the first man-made objects to show spatial quantization in three dimensions.

The microfabrication techniques in question are molecular beam epitaxy, electron beam lithography, and reactive ion etching. With molecular beam epitaxy, researchers can grow artificial crystals called "superlattices" atomic layer by atomic layer with each layer having a different composition. In the present case, the materials are gallium arsenide and aluminum gallium arsenide and the layers are 20 angstroms thick. Because the energy of the gallium arsenide conduction band lies at a lower energy than that of aluminum gallium arsenide, free electrons initially in either material end up in the gallium arsenide, leading to a confinement in one direction.

Electron beam lithography and reactive ion etching combine to provide the other two dimensions of confinement. Neither is any longer an exotic technique for microelectronics researchers, but they are not trivial to apply either. In brief, a computer controlled scanning electron beam draws the desired pattern, and reactive ion etching carves it into the semiconductor with a high fidelity.

In the end, Reed and his colleagues produced dots 0.25 micrometer on a side comprising a gallium arsenide–aluminum gallium arsenide superlattice on an aluminum gallium arsenide substrate layer. To monitor the properties of the dots, the investigators examined the luminescence due to the recombination of excitons. An exciton is the entity formed when an electron that is photoexcited from the valence to the conduction band of a semiconductor remains bound to the vacancy or hole left behind in the valence bands. The properties of the luminescence emitted when the electron returns to the valence band are sensitive to such details as changes in the energy levels within the bands.

Differences between the luminescence spectra from excitons confined in one dimension and from those confined in quantum dots provides what Reed calls the first preliminary evidence for three-dimensional spatial quantization. With smaller dots, spatial quantization effects would be much stronger, so making such structures is on the agenda.

As for applications, as the size of the transistors in microcircuits shrink in the upcoming years, it may become quite important to understand the behavior of quantum dots.

—Arthur L. Robinson