Magnetism and Superconductivity Fight for Control in High-T_c Superconductors

The high-temperature copper-oxide superconductors, which offer resistance-free current flow at temperatures extending well above 100 K, are formed by doping certain copper-oxide compounds or by adding excess oxygen to them. The parent compounds—all antiferromagnetic insulators—couldn't be more different from their superconducting offspring: Magnetism and superconductivity are generally antithetical. Yet it's hard to deny one's heritage. Many theoretical and experimental studies of high-temperature superconducting materials have turned up hints of coexisting magnetism, especially in weakly doped materials, where stronger parental influence is to be expected. But left unanswered are such questions as whether the magnetic phase competes or cooperates with the superconductivity, and whether the two phases coexist microscopically or form spatially separate phases.

Recent experiments[1–5] have given particularly dramatic evidence that the ordered arrangement of spins on the copper atoms seen in the parent compounds is always lurking in the shadows, quick to pop up whenever superconductivity is weakened, even in samples that have been "optimally doped" to give the highest critical temperature, T_c. The new studies bolster theories of competing magnetic and superconducting phases.

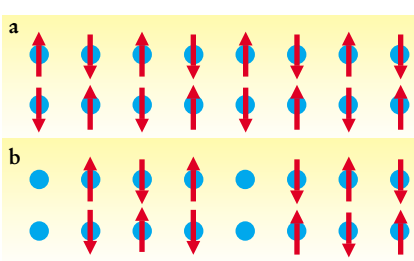

FIGURE 1. ARRANGEMENTS OF SPINS (arrows) on copper atoms (circles) in high-temperature superconductors. **(a)** Antiferromagnetic order seen in the insulating phase. Every spin is antiparallel to its neighbors. **(b)** A spin density wave that might occur at moderate doping levels. The pattern repeats every eight lattice spacings. There is charge on the sites that have no spin.

Researchers interested in exploring the competition between forces that pair electrons and those that align the atomic spins have found it useful to look at the area around magnetic flux lines threading through the material.

Perhaps one of the biggest contributions of the recent experiments has been the demonstration that an applied magnetic field can serve as a tuning knob for exploring the tradeoff between superconductivity and magnetism. Up to now, it's been more typical to study the presence of such phases in cuprate materials by varying the doping levels to take them from an antiferromagnetic insulating regime through the underdoped to the optimally doped region. That approach, however, requires a new sample for each experiment. Now, it seems, one can alter the relative strengths of the superconducting and magnetic phases simply by tweaking the applied magnetic field.

The vortex core

The recent experiments focused on the vortices that are formed when a high-T_c material sits in a magnetic field. The cuprates are type-II superconductors: Although weak magnetic fields are excluded by the Meissner effect and strong fields totally destroy superconductivity, fields of intermediate strength can penetrate in discrete flux lines, surrounded by vortices of swirling supercurrents. The superconducting pairs are broken within the vortex cores, but survive beyond some characteristic distance.

The state that forms in such vortex cores is expected to hint at the origin of the superconductivity. "It's the state you perturb to get superconductivity," says Gabriel Aeppli, who led two of the recent experiments.[1,3] (Aeppli is affiliated with NEC Research in Princeton, New Jersey, and Risø National Laboratory in Denmark.) In the case of conventional, low-temperature superconductors, the vortex core state is a normal metal. By analogy, some researchers had expected that the vortex core in high-temperature materials would be the "normal" resistive state—that is, the strange metal seen above T_c. The recent experiments now show, says Aeppli, that "the state from which superconductivity emerges is a spin-ordered state rather than a conventional metal."

Piers Coleman of Rutgers University comments, "It's amazing to see the magnetic order appear in the vortex state, as if it's a feature of the normal state that's in the background, behind the superconductivity." Magnetic or spin order refers here to a particular pattern of the spins on the copper atoms known as a spin density wave. In the antiferromagnetic arrangement seen in the parent insulator, each spin is antiparallel to its nearest neighbor, as illustrated in figure 1a. The experiments on cuprate superconductors suggest that the spins may have a more complex up-and-down pattern, such as that shown in figure 1b. Spin order is expected to be accompanied by an ordered arrangement of charges, whose repeat distance, or wavelength, is half that of the spin wave.

Evidence for spin and charge order was found in the recent studies by neutron-scattering[1–4] and scanning tunneling microscope[5] (STM) experiments, respectively. These experiments found not only that order arose when a magnetic field was applied to the cuprate superconductors but—more surprisingly—that the order was not confined to the core of the vortices. Instead, the ordered spin or charge arrangement extended a considerable distance into the superconducting region.

What had been known

Earlier neutron-scattering experiments, done in zero magnetic field, had caught spin order cohabiting with high-temperature superconductivity. In most cases, the spin order was fluctuating in space and in time. Fluctuating spin density waves were especially obvious in the weakly superconducting, underdoped members of the lanthanum strontium copper oxide (LSCO) family, but they have also been seen in underdoped yttrium barium

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copper oxide (YBCO) superconductors. In optimally doped samples, fluctuating spin density waves were seen only near or above T_c.

The role of fluctuating spin order in superconductivity has been debated (see PHYSICS TODAY, June 1998, page 19). Until the recent experiments, however, magnetic fields were known only to suppress rather than enhance the visibility of spin fluctuations in the cuprates.

About three years ago, a few members of the LSCO family, having the particular doping of one hole to every eight copper atoms, were found, surprisingly, to have static spin order. As Marc Kastner of MIT puts it, "Most people would not expect that the same electrons could participate in static spin density waves and superconductivity." Some of these experiments had suggested that the static spin density wave might cooperate with the superconductivity, but the recent studies show that the two phases compete instead.

Moreover, the recent STM experiments give the first direct evidence for either spin or charge order in a third family of cuprates: the bismuth strontium calcium copper oxides (BSCCO).

Neutron scattering

Thanks to their magnetic moment, neutrons scatter off atomic spins, and hence can detect periodic arrangements of spin—although they can't tell where that periodic structure is centered. Last spring, a collaboration led by Aeppli reported results from neutron-scattering measurements on optimally doped LSCO.[1] Low-energy spin fluctuations had been seen in this material above T_c, but they died out as the sample was cooled below T_c. Aeppli and his colleagues found that the low-energy spin fluctuations reappeared when a magnetic field was applied, and got stronger as the magnetic field was increased. The wavenumber of the spin fluctuations was too well defined to come from spin fluctuations confined to the core regions. Moreover, the spin fluctuations appeared coherent over areas with diameters at least three times those of single vortices.

These neutron-scattering experiments were done at the Risø Laboratory by researchers from there, Oak Ridge, NEC Research, and the Universities of Tokyo, Loughborough in the UK, and Karlsruhe in Germany.

Neutron-scattering studies of static spin density waves have provided additional insight. More than a year ago, Susumu Katano and coworkers from the Japan Atomic Energy Research Institute in Tokai, together with collaborators from Kyoto and Tohoku Universities, found that the static spin order for an underdoped LSCO sample was enhanced in a magnetic field.[2]

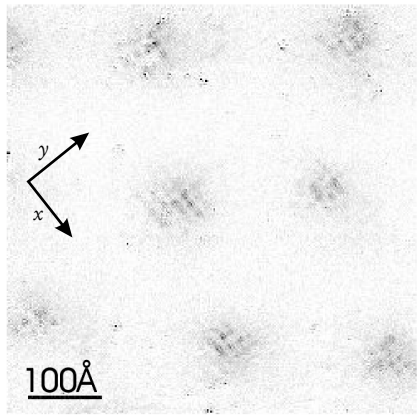

FIGURE 2. CHECKERBOARD PATTERNS in this scanning tunneling microscope image of a copper-oxide plane suggest static charge order extending out from seven vortices. The magnetic field is perpendicular to the plane. Copper–oxygen bonds make a 45° angle with the horizontal. Intensities indicate the autocorrelation of the local density of electronic states. Darker areas are regions of higher densities. The zero-field signal has been subtracted to enhance the signal seen in an applied field. (Adapted from ref. 5.)

Two experiments have since studied exactly how that enhancement varies as a function of temperature and magnetic field. One of these studies was performed by Aeppli and his collaborators, together with colleagues at the French Atomic Energy Commission in Grenoble and at Berlin's Hahn–Meitner Institute.[3] Using underdoped samples of LSCO, the researchers found that the static spin order sets in at the same temperature, regardless of field strength.

The other recent study of static spin waves, done on oxygen-doped lanthanum copper oxide, was reported by Kastner and coworkers from MIT, the University of Toronto, and NIST in Gaithersburg, Maryland.[4] Both this group and the Aeppli group find that the static spin order increases roughly linearly with the applied magnetic field H, especially for small H (the field dependence is also consistent with $H \ln H$, as predicted by some theories).

The number of vortices also grows linearly with the applied field. "Each time we add a new flux line," says Kastner, "the spins being created must be in phase with those that were there before, or we wouldn't be able to see them with neutron scattering." That suggests some kind of communication among the spins. The two kinds of order—magnetic and superconducting—must permeate everywhere in the sample, concludes Kastner.

Complementary studies

The neutron-scattering experiments imply, but don't directly prove, that the spin density waves are localized at the vortices. The missing piece of the puzzle has now been supplied by a complementary STM experiment done on BSCCO by Séamus Davis of the University of California, Berkeley, and Lawrence Berkeley National Laboratory, together with colleagues from Berkeley and from the University of Tokyo.[5] By measuring the electron density of states, the STM experiment shows that charge order, which is known to be associated with spin order, is not only localized at the vortex cores in a cuprate superconductor but extends well outside the vortices.

Figure 2 shows a measure of the electron density in one of the copper-oxide planes, where most of the supercurrent is expected to flow. One sees there the cores of seven vortices created by a magnetic field perpendicular to the plane. Each vortex is surrounded by a faint checkerboard pattern, oriented parallel to the copper–oxygen bonds. This pattern suggests a two-dimensional (2D), periodic charge structure centered on each core. Figure 3 shows a magnified version of the charge density pattern around one vortex, together with a schematic representation of a charge modulation.

If the charge order seen in STM studies reflects the same underlying phenomenon as the spin order seen in the neutron-scattering data, the spin fluctuations are indeed centered on the vortex core. Furthermore, the period of the charge order found in the STM is half that of the fluctuating spin density wave that shows up in the neutron-scattering experiments, consistent with theoretical expectations.

Interestingly, the charge order seen in the STM pictures of a slightly overdoped sample is static, whereas only fluctuating spin order was found by the neutron-scattering studies of optimally doped materials.

Researchers have sighted evidence for spin order around the vortex cores of YBCO superconductors using nuclear magnetic resonance[6] and muon spin resonance experiments.[7] Both of these techniques, like STM,

can sense the local environment, but they are sensitive to spin rather than charge order.

Theory

Steven Kivelson of UCLA comments that the high-temperature community is "currently interested in the general issue of competing orders. In many proposals about what's going on in high T_c, people say its key feature is a proximity to some ordered state." That state might be antiferromagnetism, ordered stripes of spin and charge regions, stripe orientational order, or a so-called staggered flux phase. (For a discussion of coexisting superconductivity and magnetism in other materials, see PHYSICS TODAY, September 2001, page 16.)

Shou-Cheng Zhang of Stanford University suggested in 1997 that the vortex core state would be the insulating antiferromagnetic state,[8] and in the same year he elaborated on that prediction with three other theorists.[9] The prediction followed from Zhang's SO(5) treatment of competing antiferromagnetic and superconducting orders, which are represented in his theory by 3D and 2D projections, respectively, of a 5D state vector. The prediction of this theory, that the field-induced antiferromagnetic moment is proportional to the applied field, helped pique interest in the vortex core state. Zhang and Jiang-Ping Hu have now relaxed the SO(5) symmetry to agree with experiments that find a fluctuating spin density wave rather than the static antiferromagnetic state he predicted earlier.[10]

Subir Sachdev of Yale University and various collaborators have been thinking about quantum phase transitions and magnetic order in the cuprates for many years, and the recent neutron-scattering experiments spurred them to examine the impact of an applied magnetic field on a magnetic transition. With Eugene Demler (Harvard) and Ying Zhang (Yale), Sachdev has interpreted the observed behavior in terms of the proximity of the superconducting phase to a phase with coexisting superconductivity and static magnetic order.[11] Increasing the magnetic field takes the superconducting phase very close to the phase with coexisting orders and leads to a strong enhancement of the low-energy spin fluctuations. The theorists predict that the static spin density wave signal should go as $H \ln H$. The data deviate from a linear H dependence in a manner that's consistent with this form. Before the STM experiment, Sachdev and a colleague had suggested that one might see a vortex nucleation of static charge order.[12]

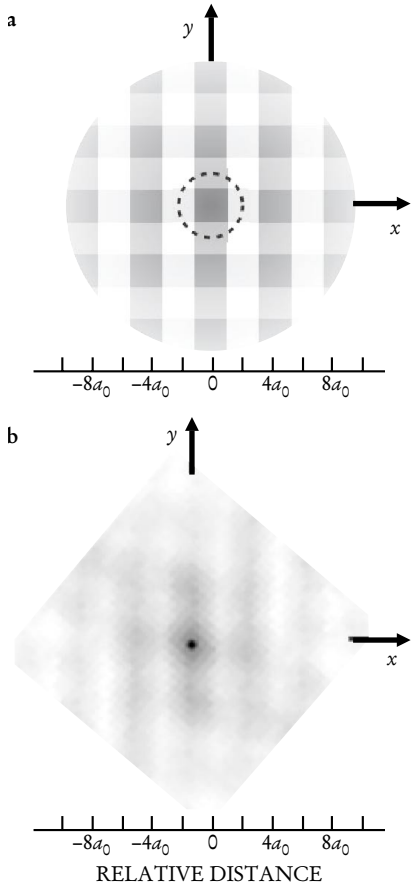

FIGURE 3. CHECKERBOARD IN DETAIL. **(a)** Schematic representation of a two-dimensional modulation of the charge density around a vortex core (dotted circle). The wavelength is four lattice spacings ($4a_0$). **(b)** Closeup of one of the vortices seen in Figure 2, rotated by 45°. Measured spectrum scaled to match the schematic above it. (Adapted from ref. 5.)

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Ultrashort Laser Pulses Can Control X-Ray Switch

The ability to direct pulses of x-ray synchrotron radiation using femtosecond lasers may open a new avenue for time-resolved x-ray studies.

In the molecular world of chemical reactions and conformational changes in biological and other molecules, events often happen on time scales from hundreds to thousands of femtoseconds—the typical period of molecular vibrations. Femtochemistry (described in PHYSICS TODAY, December 1999, page 19) uses ultrashort laser pulses, only a few tens of femtoseconds in duration, to probe electronic changes on such time scales. From these experiments, dynamics information can be extracted. But to obtain direct structural information, one needs a probe whose wavelength matches the angstrom scale of interatomic spacing: x rays are a prime candidate (see the article by Eric Galburt and Barry Stoddard in PHYSICS TODAY, July 2001, page 33).

Obtaining probes of appropriate wavelength, duration, and intensity is no easy matter, however. Laser-produced plasmas can generate sufficiently short bursts of x rays, but the bursts aren't highly collimated and have relatively low flux. Synchrotron sources, in contrast, produce trains of pulses that are tunable and have high brightness but, with durations of 10–100 picoseconds, are orders of magnitude too long.

Recently, a group of researchers from the University of Michigan, led by David Reis, Philip Bucksbaum,