

Bell Labs researchers' now-clouded string of papers set other scientists agog over results that promised to revolutionize several fields. Then the storm broke

# Winning Streak Brought Awe, And Then Doubt

Even in the best of times, 95% of Jan Hendrik Schön's experiments fizzled out. It's the other 5% that made him one of nanotechnology's brightest stars and the envy of physicists worldwide. In paper after paper, he and his collaborators reported that a simple turn of a dial could transform normally poor electrical conductors into semiconductors, metals, or even superconductors, a malleability never seen before. That opened up entirely new vistas for exploring the physics of materials—the stuff of Nobel Prizes.

Today, though, few researchers would trade places with Schön, a physicist at Bell Laboratories, the research arm of Lucent Technologies in Murray Hill, New Jersey. On 10 May, Bell Labs officials launched an investigation of Schön's work, after outside researchers revealed what appears to be duplication of data in multiple papers (*Science*, 24 May, p. 1376). Schön is the lead author on all the papers under scrutiny and the only author whose name appears on all. The investigation is the first of its kind in the 77-year history of Bell Labs, the world's most famous corporate research outfit.

Schön says he stands behind his measurements and is doing everything he can to cooperate with the inquiry, which is being conducted by an outside committee and is expected to be completed by the end of the summer. But, until then, a cloud hangs over a spectacular body of work.

Some researchers say that the suspect data have cast doubt on all of Schön's results. "I can't trust any of the work," says Harvard University chemist and nanotechnology expert Charles Lieber. Others, however, point out that much of the disputed data seems to be supporting material, not the primary results in each paper, which detailed the observations of everything from high-temperature superconductivity to quantum-mechanical signatures never before seen in organic materials.

Unfortunately, Schön's most provocative results have not been independently verified, despite years of effort by other labs and tens of millions of dollars spent on research in the area.

Even before the storm of controversy broke, other scientists were starting to raise questions about how Schön and his colleagues achieved their stunning results and why no one else has been able to repeat them.

In interviews conducted over the past 6 months—most of them before the investigation began—the Bell Labs team and others in the field retraced the whirlwind trajectory of the work and weighed its enormous promise against those simmering questions. The answers, when they come, will have enormous significance not just for the fate of one bright young researcher but also for scientists around the world trying to follow his lead, and for the future of one of the hottest ventures in condensed-matter physics.

## A question of speed

That venture got its start in Bell Labs' room 1E318, a somewhat dingy, crowded lab located one floor below the birthplace of the transistor. The room was the longtime lab of superconductivity physicist Bertram Batlogg.

One day in the mid-1990s, Batlogg and his colleagues were brainstorming ideas about work on plastic electronics when he hit on one that he just had to try.

A Bell Labs team led by physicist Ananth Dodabalapur, now at the University of Texas, Austin, had succeeded in making field effect transistors (FETs) using a variety of organic materials laid down in thin films. FETs are the bedrock electrical switches of computer circuitry. In a typical version, a pulse of electrons sent to one electrode, called the "gate," creates an electric field that repels electrons sitting in the semiconductor lying directly below, effectively spiking it with positive charges. These charges boost the conductivity of this semiconductor "channel," making it easier for electrons to flow through this channel between two other electrodes. And, presto, the device switches from off to on (see diagram on p. 35).

But electrons don't move at the same rate through all semiconductors. Dodabalapur's organic transistors weren't about to give Intel's inorganic ones a run for their money. They were painfully slow. Electrical currents crept through the organic channels at a pace orders of magnitude below their speed through even the worst silicon-based devices. The team didn't know whether organics were inherently slow conductors or whether the problem lay in the way the devices were constructed.

Batlogg suggested a way to find out. In the thin organic films that Dodabalapur's team was using, the organics invariably organized spontaneously into tiny crystallites, like gravel on a path. It was possible that charges were zipping through the perfectly ordered organics within each crystallite but were getting hung up at the ragged borders as they hopped from one crystallite to the next. Batlogg suggested growing larger single crystals and using them to measure the speed of electrons. Because single crystals don't have grain boundaries, the researchers would see what the materials could really do.

The catch was that making high-quality single crystals out of organics is much more easily said than done. "Organics are synonymous with crappy stuff," Batlogg said in February. Even the best organic



**High flyer.** Jan Hendrik Schön dazzled physicists with results many have tried to emulate.

CREDIT: BELL LABS



crystals typically harbor 1% to 2% impurities, often solvent molecules left over from their original synthesis. Their presence can disrupt the regular crystalline order of the material enough to make it impossible to grow a single crystal.

Dodabalapur's team members were too busy with their thin-film transistors to steer their research in a new direction. So, Batlogg offered to help. In 1997, he and Bell Labs chemist Bob Laudise recruited Christian Kloc, a chemist then based in Konstanz, Germany, who was an expert at growing crystals. Kloc quickly hit upon a new strategy for both purifying organics and growing crystals at the same time. Kloc's progress meant Batlogg needed another set of hands to put the crystals through their electrical paces. His longtime friend (and Kloc's former boss) Ernst Bucher in Konstanz recommended Schön, who jumped at the opportunity and left for New Jersey even before finishing his Ph.D.

Schön set up shop in Batlogg's lab, and the results came quickly. When Schön slapped electrodes on a variety of different organic crystals, none came even close to matching the speed of the standard crystalline silicon semiconductor. The best organic, called pentacene, just kept pace with low-grade amorphous silicon, a semiconductor commonly used in solar cells. Dodabalapur's team had already achieved similar speeds with their thin-film FETs—devices that were chock-full of grain boundaries.

This was bad news for organic-crystal research. If grain boundaries didn't hinder the flow of current, then there wasn't much anyone could do to improve the crystals' plodding speed. Plastic electronic devices, it seemed, were destined to be slow.

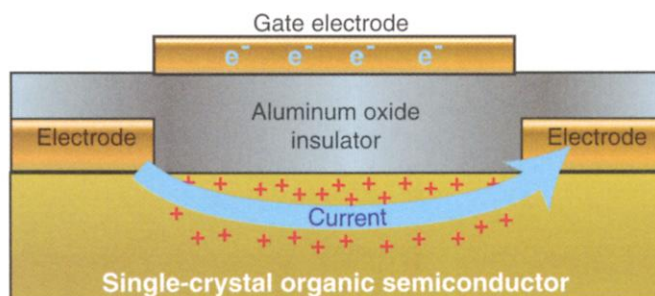
### Up for a challenge

The science of working with single crystals of organics, however, was just picking up speed. Horst Stormer—a Nobel Prize-winning physicist who was then at Bell Labs but has since moved to Columbia University in New York City—spurred the team on by issuing a challenge. "He said [that] if any semiconductor is decent, you can make it into a transistor," Batlogg recalled. Transistors had already been made with thin films of organics. But those are relatively simple devices to make. Researchers place metal electrodes on a wafer of inorganic crystalline silicon, then add a layer of the soft organic material atop the wafer's tough ceramiclike surface.

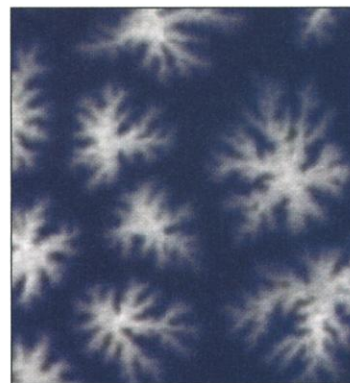
What Stormer was proposing was much harder: starting with one of Kloc's fragile,

millimeter-sized single crystals of organics as the substrate and planting metal electrodes on top. The upside-down approach was necessary because organics grown on wafers spontaneously form tiny grains, or crystallites. As a result, single-crystal FETs must be built from the crystal on up. "The difficulty was the prospect of putting down hard materials on soft materials, held together only with weak van der Waals bonds," said Art Ramirez, a physicist at Los Alamos National Laboratory in New Mexico, during an interview in March. "You have to do the deposition very, very carefully."

To succeed in making a single-crystal or-



**Electrifying.** In an organic FET, electrons sent to a gate electrode induce positive charge in a semiconductor such as pentacene (right), causing current to flow.



ganic transistor, Schön needed another key ingredient: a thin insulating barrier to prevent charges from shuttling back and forth between the electrodes when they're not supposed to do so. All transistors rely on such insulating barriers, which come in a wide variety of chemical flavors.

Schön decided to make his insulator out of aluminum oxide, a decision that became the key to the group's biggest successes and its greatest mystery. Not long after joining Bell Labs, Schön flew from New Jersey to Konstanz to finish up work on his Ph.D. "I got back to Konstanz and was sputtering aluminum oxide for solar-cell coatings," he recalled in February. "No one else was using the machine, so I decided to try it out" for the organic transistors. The machine coated the organic crystals with a neat insulating layer.

Later, Schön used a low-temperature scheme to deposit the gate electrode on top of the aluminum oxide while protecting the fragile organics. When he hooked up the electrodes to a power supply and flipped the switch, he recalled later, the results jumped off the screen. Not only had the fragile organic crystals not cracked, broken, or turned to ashes, but they had changed from insulators to semiconductors, conducting current when prompted by the gate voltage.

The result was a paper published in the 11 February 2000 issue of *Science* (p. 1022). "That was the first time the community took notice of the crystals," Batlogg said.

The paper caused a sensation in the condensed-matter physics community, because it held out the prospect that researchers could make electronic devices out of an enormous variety of organic materials. Those wouldn't necessarily be any better than silicon FETs for computer circuitry. But they would give researchers a new way to track how electrical charges move through a wide variety of materials. According to the Institute for Scientific Information, the paper has since been cited

130 times, making it not only Schön's most highly cited paper but also one of the top 0.01% of all physics papers published in 2000.

Schön, Batlogg, and Kloc were just getting warmed up. And Stormer was ready with a new challenge. "Horst said, 'Any real semiconductor has a quantum Hall effect,'" Batlogg noted. The effect, a stepwise change in voltage that

occurs when a semiconductor studded with electrodes is placed in a magnetic field, is a hallmark of the quantum-mechanical behavior of electrons. Most physicists thought it could be observed only in materials so pure that electrons move through them without scattering off obstacles. Because organic crystals

normally harbor so many impurities, "I never thought we'd see it," Schön said.

But, on 23 December 1999, just before taking off to Germany for the holidays, Schön ran the experiment and reported seeing the telltale voltage steps. "We showed the result to our [Bell Labs] colleagues, and everyone thought we were joking," Batlogg said. Added Schön: "It was a nice Christmas present." Later, Schön also noted that he had witnessed a related effect called the fractional quantum Hall effect, the effect for which Stormer had shared his Nobel Prize in 1998. "Wow. I thought this was fantastic," Stormer recalled in an interview a week before news of the disputed figures broke.

### Superconductors and beyond

The Bell Labs trio didn't revel in its success for long. By now, Kloc was churning out high-quality crystals of a variety of organics, including ones made of C<sub>60</sub>, the soccerball-shaped carbon molecule also known as a



buckyball or buckminsterfullerene. Back in 1991, a Bell Labs team that included Ramirez had turned the normally insulating  $C_{60}$  into a superconductor by spiking it with potassium atoms. The potassiums harbor extra electrons, which could move around through the crystal and pair up as they go, a signature of superconductivity. Theoretical results suggested that if researchers could add three extra electrons for each  $C_{60}$  molecule in the crystal, they could get it to superconduct without potassium atoms.

Where to get those electrons? Schön, Batlogg, and Kloc wondered whether they could use the electric fields produced by their FETs to yank them from the FET electrodes and shunt them into a channel made from  $C_{60}$ . If the density of charges got high enough, perhaps the material would resemble a metal like copper, or, if they got really lucky, perhaps even a superconductor.

But that wasn't a simple proposition. The density of free electrons in a metal is at least 1000 times that of a semiconductor. "If you want to go to very high [electron] concentrations, you have to apply a very high field" to the gate electrode, Schön said. Normally, that turns the organic to ash. So Schön needed not only to build FETs atop  $C_{60}$  but also to have the organics and the aluminum oxide insulator withstand withering electric fields. Nearly all his attempts failed. But in a few cases, Schön's seemingly magic layer of aluminum oxide somehow handled the high currents. In the 28 April 2000 issue of *Science* (p. 656), Schön and his colleagues reported that they had coaxed potassium-free crystals of  $C_{60}$  to superconduct at 11 degrees above absolute zero.

Not bad for starters. But they dreamed of achieving even higher superconducting temperatures. Theorists had suggested that  $C_{60}$  could reach such temperatures if it could be made to conduct positively charged "holes" instead of electrons. Holes are vacant electron sites and can move through a material just as electrons do. But, although chemists could add extra electron-carrying atoms such as potassium into a  $C_{60}$  crystal, there was no chemical method for adding holes.

FETs, however, can manage the task handily. All the researchers had to do was simply reverse the polarity on the electrodes to pull electrons off the  $C_{60}$ 's. In the 30 November issue of *Nature*, Schön, Batlogg, and Kloc reported that the scheme worked

just as theory said it should, allowing a  $C_{60}$  crystal to superconduct at 52 kelvin. Less than a year later, they reported in *Science* that they had tweaked the  $C_{60}$  crystals to push the superconducting temperature up to 117 K (*Science*, 31 August 2001, p. 1570).

The papers not only shattered the record for superconductivity in  $C_{60}$  but also offered researchers the heady prospect of finding high-temperature superconductors without painstakingly doping each material with different impurities. "They defeated chemistry," Princeton University physicist Bob Cava said in February.

More marvels were to come. In 2000 and 2001, papers by the trio were flooding the journals. Schön and collaborators both inside and outside of Bell Labs reported turning organics into everything from light-emitting lasers to light-absorbing photovoltaic devices. They made plastic, a notoriously messy organic compound, superconduct. And they showed that their high-field FETs could work just as well with inorganic superconductors, a development that promised to revolutionize the field of high-temperature superconductivity.

As if that weren't enough, while Schön was rewriting the textbooks on condensed-matter physics, he was also busy pioneering a separate field: molecular-scale transistors. In the 18 October 2001 issue of *Nature*, he and Bell Labs colleagues Zhenan Bao and Hong Meng reported making a novel type of transistor in which the key charge-conducting layer was composed of a single layer of an organic conductor. They followed that with a report in the 7 December issue of *Science* (p. 2138) describing how they

diluted the charge-conducting layer with nonconducting insulating molecules, allowing them to track the conductivity in a transistor through a single molecule. Together, the results were hailed as a triumph of molecular-scale electronics.

Through the beginning of this year, Schön had racked up 15 papers in *Science* and *Nature*, as well as dozens in other journals. Between 1998 and May 2002, he published more than 90 papers and was lead author on 74 of them, a staggering level of productivity. Of his top 20 papers, all are in the top

10% of physics papers for their number of citations, with eight in the top 0.1% (see table). Many of those citations are no doubt Schön and colleagues citing their own previous work. Nevertheless, the work clearly captured the imagination of others in the community. "Hendrik has magic hands," Ramirez said in the March interview. "Everything he does seems to work."

But, even before the revelations in May, questions began to swirl around Schön's work. His magic, other researchers noted nervously, didn't seem to work for anyone else. Over the past 2 years, efforts to slap high-field FETs on different materials have become one of the hottest endeavors in condensed-matter physics. The U.S. Department of Energy, for example, has helped fund a new group specifically to try to reproduce and extend the Bell Labs results.

So far, however, the well is dry. "It's very

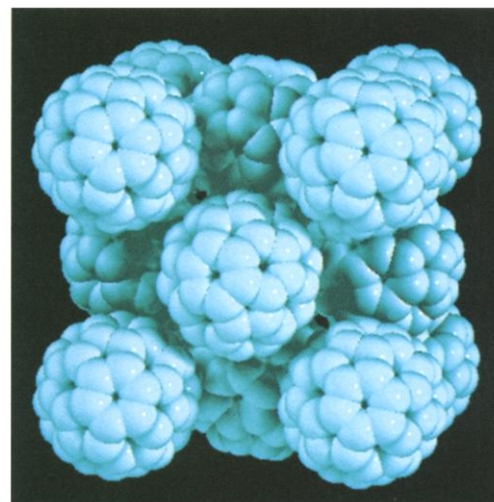
## SCHÖN'S TOP TEN

Topic	Journal	Year	Citations*
Pentacene FETs	<i>Science</i>	2000	0.01%
$C_{60}$ Superconducts at 52 K	<i>Nature</i>	2000	0.01%
Organic FET Laser	<i>Science</i>	2000	0.01%
Organic FET Superconductors	<i>Nature</i>	2000	0.1%
Superconducting Switch	<i>Science</i>	2000	0.1%
Fractional Quantum Hall Effect	<i>Science</i>	2000	0.1%
Plastic Superconductor	<i>Nature</i>	2001	0.1%
Pentacene Photovoltaics	<i>Nature</i>	2000	1%
Elec. Prop. of Single Crystals	<i>Phys. Rev. B</i>	1998	10%
Elec. Cond. of Oligothiophene Single Crystals	<i>APL</i>	1998	10%

\*Ranking among the top percentage of all physics papers in the same year by number of citations, as determined by the Institute for Scientific Information.

Red: Implicated in possible data duplication.

Black: Not suspected of duplication.



**Crystal balls.** Bell Labs team reported new records for superconductivity in  $C_{60}$ .

CREDIT: (BOTTOM) GERARD MEIJER AND MARCO TOMASELLI

unusual to have a result that is 2 years old that hasn't been reproduced," Richard Green, a physicist at the University of Maryland, College Park, said during an interview in February. Added Robert Dynes, a physicist and chancellor of the University of California, San Diego: "Some people are frustrated and discouraged."

Some researchers also complain that Schön's early papers left out key details needed to reproduce the work. "There is an uneasy feeling around the community," said Teun Klapwijk, a physicist at the Delft University of Technology in the Netherlands on 2 May, just before the discovery of the apparent duplication of data. "Why do the papers have so little detail? It's such a unique case of a whole string of papers where each paper shows you the beautiful result you want to see: the Hall effect, lasing, the quantum Hall effect, superconductivity. People kept feeling, 'Is that possible? How do you produce so many results? Is that physically possible?'"

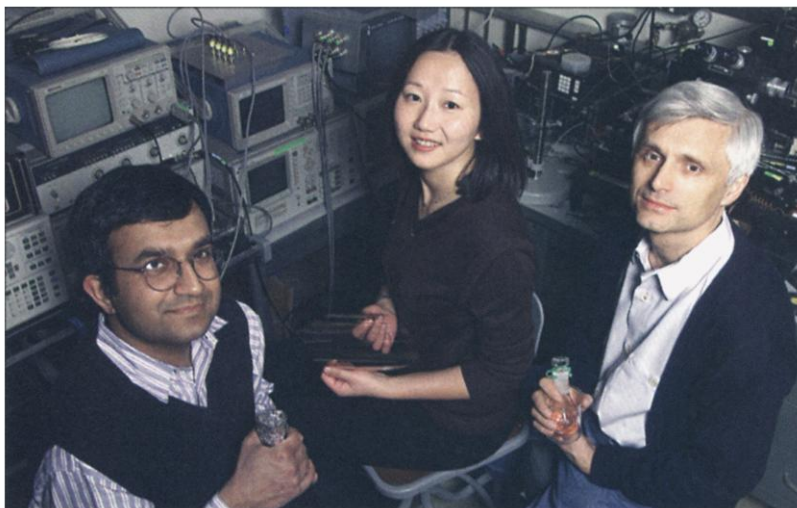
Much of the concern boiled down to Schön's aluminum oxide. Making the electron barrier is a piece of cake. You just vaporize the materials in an apparatus called a sputtering machine and let the vapor rain down on your sample. Add the electrodes, and you're in business. "You really need very little to get into the game," said Ramirez. But when most researchers start turning up the electric fields to drive electrical charges into their organic materials, they can create fields of only about 10 million volts per centimeter before the aluminum oxide starts to bubble, turn black, and vaporize, taking the fragile organics along with it. Somehow, Schön's aluminum oxide gets up to 45 million volts/cm—nearly five times higher than anyone else. "What is the trick?" asked physicist Arthur Hebard of the University of Florida, Gainesville. IBM physicist John Kirtley agreed: "That's the \$64 million question."

#### Magic box

In an interview last February, Schön, Batlogg, and Kloc said they were as eager to find out the answer as everyone else. "I wish we knew," Batlogg said. Added Schön: "If we knew, then we wouldn't have to waste 18 samples out of 20." Schön said he had looked at the aluminum oxide layers under ultrahigh magnification but found nothing remark-

able—just a noncrystalline amorphous layer of aluminum oxide. Whatever the secret, it seems unique to Bucher's sputtering machine in Konstanz, the only place where Schön has managed to grow aluminum oxide layers that withstand the high fields.

Sputtering machines are commonplace in the world of semiconductor electronics, and Bucher's is a run-of-the-mill one at best. The machines vaporize their targets in a vacuum to ensure that outside compounds don't find their way onto a sample. But the vacuum in Bucher's machine is "lousy," said one researcher, capable of reaching a pressure of  $10^{-6}$  torr. By contrast, state-of-the-art molecular-beam epitaxy machines—devices used to lay down



**Team effort.** Bell Labs colleagues such as Ananth Dodabalapur, Zhenan Bao, and Christian Kloc were among Schön's many collaborators.

vaporized materials one atomic layer at a time—can reach  $10^{-12}$  torr.

The upshot, says one researcher, is that Bucher's machine isn't just laying down aluminum oxide: "It has everything in it—your breath, water, other gases." That rain of mystery compounds, the Bell trio speculates, might somehow toughen the material against meltdowns, perhaps by plugging defects that would otherwise snag electrical charges.

Even if other groups manage to make aluminum oxide that's stable in high electric fields, high-field FETs will face further hurdles. For the devices to work, both the underlying crystals and the interfaces between the different layers of semiconductors, insulators, and metals need to be nearly perfect to prevent charges from getting hung up as they travel between layers and burning out the device. As a result, Texas's Dodabalapur said at a meeting of the American Chemical Society (ACS) in April, high-field FETs are so fragile that getting them to work "requires the skill of a jeweler, the persistence of a saint, and the background of a physicist."

Even so, some teams believe they're making progress. Ramirez's group at Los Alamos has made the most headway. At the March meeting of the American Physical Society in Indianapolis, Indiana, Ramirez reported that when he and colleagues ran currents through FETs they had created using  $C_{60}$  crystals made by Kloc, they saw signs of the organic's behaving like a metal—although not a superconductor.

#### Clouded prospects

That was where Schön's saga stood in early May. Since then, the latest chapter—revelations of possible duplication of data—has cast doubt on all that went before. It would make matters far simpler if Schön could

submit his best FETs for independent testing or invite other researchers to make measurements on his equipment. But that's not possible. The small fraction of Schön's FETs that did work in the past were either fried in the process or have degraded, he says. Worse, Schön's magic in making his high-strength aluminum oxide seems to have evaporated, as even he has been unable to reach the high electric fields for about the last 6 months. "We have the same problems now as everyone else," Schön said at the April ACS meeting. "It has been

frustrating. We can empathize with what others have been going through."

Now, one of the most exciting strings of results in modern physics is under a cloud. It's impossible to say which work will stand the tests of time and intense scrutiny. "Maybe some of the most dramatic stuff is right," says James Heath, who heads the California NanoSystems Institute at the University of California, Los Angeles. "What's harder to believe: that everything is wrong, and they made up all of the data, or that some of it is real? It's easier to believe that there are some legitimate results."

If Schön's results hold up, they would point the way to exciting physics and novel devices. If not, the loss could be devastating—not just for the careers of those directly involved but for the credibility of Bell Labs, condensed-matter physics, and science as a whole. Research on organic electronics would of course press on. But, it would march less resplendently than it did before Hendrik Schön set foot in Bell Labs 4 years ago.

—ROBERT F. SERVICE