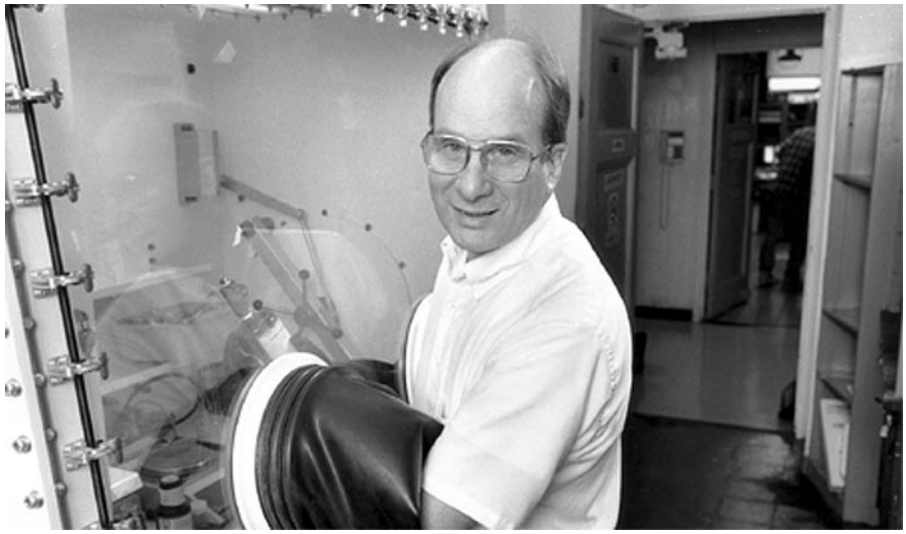
Biography of Louis E. Brus

Just as the shiny luster of silver and gold has attracted people for ages, these metals strongly attract their own kind at the nanoscale. Louis Brus, elected to the National Academy of Sciences in 2004, shows that strong attractive forces are generated between noble metal particles when irradiated by wavelengths from the visible spectrum. His Inaugural Article in this issue of PNAS discusses the basic science behind this and the ways this optical property could be harnessed to fuel the growing need for nanoscale research tools (1). A physical chemist, Brus has carried out research at the atomic level for more than 20 years and is best known for his work on semiconductor nanocrystals. With the rise of nanomaterials and nanotechnology has come the need for tools to manipulate and organize single particles, especially remote methods using lasers. Brus' article is an outgrowth of a systematic effort to understand single-molecule Raman spectroscopy (2).



Louis E. Brus

A Career in Disguise

Brus was born in 1943 in Cleveland, OH. The son of an insurance executive, he moved several times within the Midwest. In high school in Roeland Park, KS, a suburb of Kansas City, Brus developed an intense interest in chemistry and physics. Brus entered Rice University (Houston, TX) in 1961 with a Naval Reserves Officer Training Corps (NROTC) college scholarship. His freshman year was the first year that a major in chemical physics was offered. "Whether or not I would be good at it, I didn't know," he says, but he declared this major with enthusiasm. During each semester of college, Brus says he took one chemistry, one physics, and one mathematics course, in addition to the liberal arts courses the school required. "Rice was difficult, but an excellent place for science," Brus says. During his summers in college, Brus was required to participate in NROTC activities at sea as a midshipman, which precluded opportunities for other jobs or research. It was not until after his senior year that Brus found the room for a research project. He worked with John L. Margrave, a physical inorganic chemist, "learning how to do optical spectroscopy of high-temperature vapor species."

Brus became hooked on physical chemistry during this time, saying, "I wanted to study it as long as I could." He decided to go to graduate school but saw doctoral research as a way to indulge his interests more than develop a career. "I still did not know I could practice this," he says. "I didn't think of it as a profession." Upon graduating from Rice in 1965, Brus was officially commissioned as an ensign in the Navy but received special dispensation to take a leave of absence and enter graduate school instead of naval service. In the fall of 1965, he began his doctoral studies at Columbia University (New York, NY). Brus explains that he chose Columbia because "it had an explicit program in chemical physics," and, having spent most of his life in the Midwest, Brus confesses, "I had an interest in New York City." For his thesis, Brus worked with mentor Richard Bersohn on the photodissociation of sodium iodide vapor. Brus tried "to understand how excited sodium atoms are produced in the photodissociation process and the rates of their subsequent reactions" (3). The work was slow going, says Brus. "I worked for 14 months before I got any publishable data." And time was a factor in Brus' work. The Navy had granted him only four years to complete his Ph.D. Upon receiving a doctorate in chemical physics in 1969, Brus returned to the Navy as a lieutenant and served as a scientific staff officer at the United States Naval Research Laboratory (Washington, DC). Scientifically, "I managed to choose what I would work on," he says, and he likened the experience to doing postdoctoral research.

While in Washington, DC, Brus collaborated with Jim Comas, Ming Chang Lin, and Jim McDonald on surface chemistry and infrared chemical lasers. In the early 1970s, lasers were a relatively new phenomenon, according to Brus. To create a laser beam, "you need a population inversion," explains Brus. "You need more molecules in the excited state than in the ground state. As [George] Pimentel originally discovered, chemical reactions can create this population inversion," negating the need for electricity to generate a laser beam. Brus and his colleagues studied which gases and reactions produced the best population inversions. Whereas the military had an interest in using lasers as weaponry, Brus' work was directed at the fundamental science, "measuring all types of reactions" to find those that might be useful (4). In 1973, Brus left the Navy permanently and was hired by AT&T Bell Laboratories (Murray Hill, NJ), based in part on a strong recommendation letter written by Bersohn.

Phase Change

The research at Bell Laboratories differed from what Brus had worked on thus far. "They focused on the science relevant to electronics and telecommunications," says Brus. "My gas phase work was not relevant, and there were great opportunities in condensed phases. It was a great place for a young scientist to come to work." He first worked on matrix isolation spectroscopy with Vladimir Bondybev. Together, they sought a "fundamental understanding of how energy flows in molecules in solids." Brus and Bondybev studied how excited electronic energy becomes vibrational

This is a Biography of a recently elected member of the National Academy of Sciences to accompany the member's Inaugural Article on page 1280.

energy and then heat energy over time. Based on the two types of motions inside a solid—phonons (running waves) and local vibrations around an impurity molecule—they bombarded solids, typically solid argon, at a low temperature (4 K) with laser beams to drive impurity molecules into an excited state and then observed the subsequent dynamics. Light was not only the instigator of the movement but also the readout of the experiment. "Through changing of the colors of emitted light, you are following the internal dynamics," Brus explains. The work resulted in the discovery of how local modes function in relaxation of the molecule from its excited state (5, 6).

In the early 1980s, Brus "became more interested in complex, mainstream chemistry." He switched from studying solids to work with liquids at room temperature. This shift required new techniques. "The experiments go hand in hand with the improvement in the apparatus," he says. "I had to go deeper into laser technology." To study time-resolved Raman spectra of intermediate species in reactions, Brus used a laser that could produce two or more pulses of different colors with a time delay (7). One laser beam excited the molecules, and the next probed the resulting structures.

Some of Brus' most significant work began as "an accidental observation in 1983." He began to look at organic oxidation and reduction (redox) reactions taking place on the surfaces of photoexcited semiconductors. For a semiconductor to conduct, the electrons must leap from their normal orbital up to another. The band gap, or the energy divide that an electron must surmount between orbitals, is crucial in a semiconductor's functioning. Brus used aqueous colloidal semiconductors to obtain higher surface area for the reactions. He noticed that the band gap decreased over several days and traced this to an increase in particle size with time. "The band gap of the semiconductor was experimentally a function of particle size," observed Brus (8), explaining that small particles are less stable because of their higher surface area and thus spontaneously grow. He discussed this band gap evolution with physicist and colleague Michael Sturge and performed "theoretical modeling of quantum confinement and electrostatics in three dimensions" (9). To make smaller and smaller particles for study, he and postdoctoral fellows Paul Alivisatos and Moungi Bawendi collaborated with organometallic synthetic chemist Michael Steigerwald (10, 11). According to Brus, "this collaboration was absolutely essential and very productive." Collectively, they began to understand the evolution from molecules to bulk semiconductors. During this time of the mid-1980s, Brus realized that these nanocrystals were highly important for microelectronics. The team could see that the future of computing depended on smaller and smaller semiconductors.

From Industry to Academia

Brus continued his work on semiconductors, especially silicon, through 1996 at Bell Laboratories. "The management of the laboratory supported me very strongly," he says. In the mid-1990s, the company became Lucent and began moving in new directions, and Brus realized he needed a change. "My research would be invigorated if I went and did something else with broader horizons," he recalls. "That something else was becoming an academic. I was psychologically ready to teach." Brus returned to Columbia University as a professor in 1996 and is currently S. L. Mitchell Professor of Chemistry. However, his transition back to academia was not an easy one. Having spent his entire career in private industry, Brus "had never done many of the things necessary for an academic," such as teach a course or write a grant. He remembers George Flynn, then chairman of Columbia's chemistry department, and George McLendon, now at Duke University (Durham, NC), as helpful mentors in his transition. Brus now enjoys teaching and sees teaching large undergraduate courses, where most students will not necessarily become scientists, just as important as instructing graduate students how to do research.

"It is truly amazing how much nanocrystals have grown. I'm going sideways as fast as I can to find novel problems."

In the academic environment, Brus has continued to work in the area of nanoscience in several ways. One of Brus' last projects at Bell Laboratories, with Jay Trautman and Tim Harris, focused on developing confocal single molecule spectroscopy for characterizing single nanoparticles (12, 13). In a particle that is only 50 Å wide, there are still approximately 3,000 atoms. Even the best synthetic methods cannot make atomically identical nanocrystals of this size. The properties of each atom count, making it essential to characterize individual nanocrystals. At Columbia, Brus has trained graduate students in microscopy and explains his realization that "these optical methods would be good for characterizing single carbon nanotubes" (14, 15). In addition to optical microscopy, Brus has also used electric force microscopy (16, 17), which measures the presence of single electrons on individual nanocrystals. He is currently pursuing theoretical questions of nanocrystal organization with colleague David Reichman (18) and electronic structure with colleague Rich Friesner (19).

The current popularity of work on the nanoscale has Brus reeling. "We had no idea it would be as popular as it is," he says. "It is truly amazing how much nanocrystals have grown. I'm going sideways as fast as I can to find novel problems." Brus explains that his Inaugural Article does not necessarily highlight the main subject of his research, which is understanding the nature of semiconductor nanocrystals. "The Inaugural Article is an outgrowth of our effort to understand how it is possible to see the Raman spectra of individual molecules." Raman spectra are commonly used to identify structure in molecules and materials. Upon excitation with a light beam, most of the light is scattered elastically, but some light interacts with the molecules inside the material, producing a characteristic scatter pattern unique to that material. Brus is curious how noble metal particles can act as nanoscale antennas to concentrate electromagnetic fields.

The Inaugural Article explores why there is an enhancement of the local electromagnetic field when two particles are close together. The calculations show that the same effect should also create a large optical force between the two particles (1). The phenomenon had been recognized, "but no one had explored it systematically," say Brus. He hopes to discern whether it can be used in a practical sense. "We're always looking at ways to manipulate particles or organize them." Brus is also trying to explore the effect experimentally, saying, "theory is only theory; what actually happens may be different."

On a Larger Scale

Outside of research, Brus is "a voracious reader of history and biography." He and his wife of 34 years, Marilyn, have three grown children, and he maintains a garden in the shady yard of his home. Gardening provides Brus with "some occupational therapy and relief from science." Tongue in cheek, he

expounds, "I like to dig holes in the ground because it tires you out, and you've got something to show for your work, unlike what sometimes happens in months of toiling away at research."

However, the toiling away has paid off. Brus was awarded the Irving Langmuir Prize in Chemical Physics from the American Physical Society in 2001 and is the 2005 recipient of the Chemistry of Materials Prize from the American Chemical Society for his work on nanocrystals. Brus is quick to point out all those who have worked with him along the way. "Work like this is collaborative," he says. "What I do is truly interdisciplinary. It is great fun to work with people who can teach you new things."

Tinsley Davis, *Freelance Science Writer*

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