Toward a One-Angstrom Electron Microscope

By Stephen J. Pennycook

"A new technique developed at ORNL is revolutionizing electron microscopy."

Steve Pennycook obtains high-resolution images of a high-temperature superconducting material using a scanning transmission electron microscope modified to take advantage of his Z-contrast technique.

A new technique developed at ORNL is revolutionizing electron microscopy. It has demonstrated a resolution improvement of 35% over conventional transmission electron microscopy (TEM), and, equally important, it provides an image that is simple to interpret and reveals a material's atomic composition directly. Based on a scanning transmission electron microscope (STEM), this technique represents a fundamentally new approach to electron microscopy because, for the first time, it provides a direct view of materials on the atomic scale.

The goal of state-of-the-art electron microscopy is to "see," or resolve, features as small as one angstrom (Å), which is one ten-billionth of a meter (10^{-10} m) and about one millionth of the diameter of a human hair. The distances between neighboring atoms are typically 1 to 2 Å in all materials, including semiconductors, superconductors, metals, and ceramics. An instrument that resolves features this small would enable scientists to view directly the structure of these materials on the atomic scale and to locate and identify atoms at defects and interfaces in these materials. This capability would improve the understanding of the origins of bulk materials properties, such as the strength of structural materials, the ability of superconductors to carry electrical current, and the speed of electron transport through candidate materials to be used in compact electronic devices. From such knowledge, we could tailor new materials for improved properties, leading the way to stronger ceramics, faster computers and communications, and other technologies of the future.

ORNL Concept Economical

The principles governing conventional TEM imaging were developed 40 years ago. Since then, significant improvements have been made, particularly in resolution at the atomic level. The 1-million-volt (MV) Atomic Resolution Microscope at DOE's Lawrence Berkeley Laboratory, which can resolve individual atoms that are only 1.6 Å apart, currently has the world's highest resolution. Plans are being made by a Japanese company to achieve 1-Å resolution through technological development. If successful, such a TEM machine would still require a team of trained operators to use and maintain it, and a building in which to house it. The estimated cost for such a microscope, which would probably require 5 years to develop, is $12 million.

ORNL is proposing an alternative method for attaining 1-Å resolution—one that would require only one operator, a normal-size room, and only $1.5 million, practically one-tenth the cost of the conventional approach. The new instrument would be shared with members of the scientific community through the standard policy of collaborative research. By modifying a commercially available 300-kilovolt (kV) STEM, manufactured by VG Microscopes in England, ORNL could achieve a resolution of 1.3 Å—the highest ever obtained. This modification could be accomplished as soon as next year. This is possible because the ORNL approach essentially uses only existing technology; stable 300-kV power supplies already exist for conventional TEMs, obviating the need for extensive technological development.

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ORNL Concept Simpler

Besides being more economical than the conventional TEM, the ORNL approach is inherently superior because it produces only one image of a material's structure, whereas a conventional TEM image, which is an interference pattern, can take many forms. Like the ORNL technique, a conventional TEM forms a picture of the atomic arrangements inside materials—actually thin sections of bulk materials, penetrated by electrons from the microscope. Scanning tunneling microscopes and scanning electron microscopes provide views only of surfaces.

"In effect, we can use the electron microscope to simply photograph the atomic arrangements inside materials."

How a conventional TEM forms an image is shown in the figure on p. 57. A broad beam of electrons from the TEM illuminates the material. As the electrons pass through the material and bombard atoms, they are scattered in many directions. The images of atoms are obtained by "tracking" the scattered electrons: an electron lens gathers and focuses the scattered electrons onto a screen, giving a magnified interference pattern or image. Although this seems similar to the operation of a camera or optical microscope, there is one crucial difference! The electron beam is different from ordinary light because, like laser light, it is highly coherent. In coherent imaging, two objects can combine either "in phase" or "out of phase," giving a signal that is either brighter or darker than it was before; thus, atoms can look bright or dark, depending on the setting of the lens and the thickness of the sample. An analogy would be making a photograph by changing the focus of a camera so that the sky turns dark and the trees turn bright. Because TEM images of atoms cannot be interpreted simply by eye, a computer-simulated image must be generated and compared with the experimental image.

The ORNL technique uses a fundamentally different approach, shown in the upper right schematic on p. 57. A finely focused electron beam, or "probe," is scanned across the sample. A ring-shaped, or annular, detector picks up those electrons scattered through large angles (like foul balls in a baseball game). These electrons are not coherently scattered, however; the coherently scattered electrons, which are used to form a conventional image, pass through the central hole in the detector (like baseballs "hit up the middle" simultaneously during batting practice). The images produced by the ORNL technique have characteristics typical of an incoherent image such as a photograph—much as the image you see through your camera lens is the same as that you see without it. This new technique makes it possible to view atoms in the same direct way, and we always see atoms as white, never as black. In effect, we can use the electron microscope to simply photograph the atomic arrangements inside materials.

Our technique works even for relatively thick samples in which the scanning probe itself undergoes complex coherent diffraction effects as it passes through the crystal. At first sight, we might expect this interaction to introduce some anomalous characteristics, but detailed theoretical analysis has shown that the image is practically unchanged by these effects. Because calculations are not needed to correct for these effects, computer simulation of images is simpler and images can even be roughly interpreted by eye.

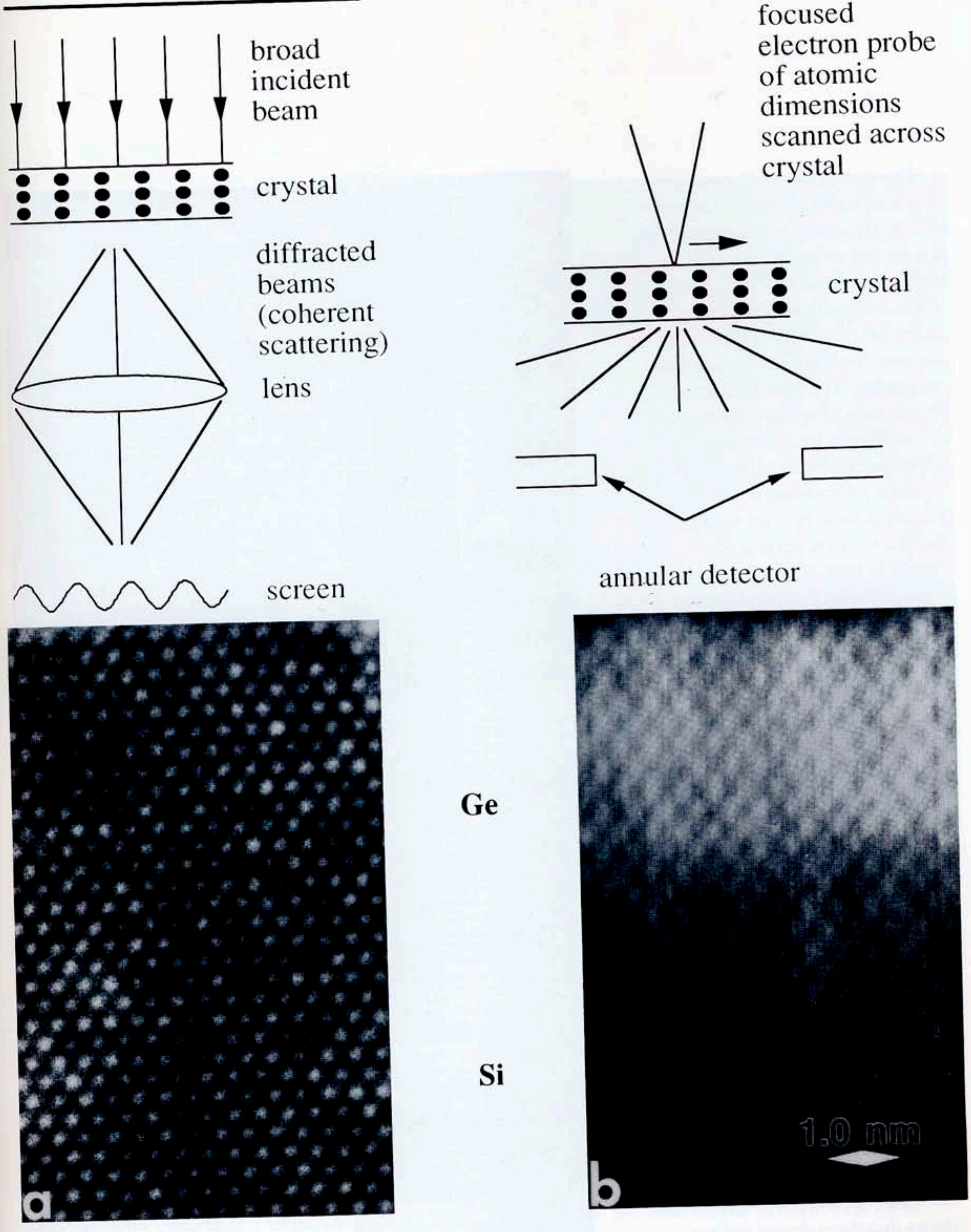
In all optical systems, the ultimate resolution depends on the wavelength of the light or electrons used. Two objects separated by less than a wavelength cannot be resolved as separate objects but will be blurred into one. Because electrons have a much shorter wavelength than light, electron microscopes have a much better resolution than light microscopes. Shorter-wavelength electrons are produced by using a higher accelerating voltage, usually measured in kV. Only in recent years has the resolution of conventional TEM machines exceeded that needed to resolve the atomic separations in most practical materials. However, an incoherent image will always show higher resolution than a coherent image taken under comparable conditions. For this reason, our existing STEM, operating at only 100 kV, provides images having resolution comparable to those taken by conventional microscopes operating at 200 or 300 kV, and a STEM operating at 300 kV would give the world's highest resolution.

Z-Contrast Technique

Besides producing superior resolution and an easily interpretable image, the ORNL technique also gives information on the species of atom imaged. For example, it enables scientists to distinguish between silicon (Si) and germanium (Ge) atoms in a material (see figure on p. 57) because the number of electrons scattered by the sample is directly related to the composition. Because atoms of higher atomic number (Z) scatter more electrons, they produce a brighter image. We call this technique Z-contrast imaging because, although atoms are always white in the image, the heavier a species is, the whiter it appears.

Conventional high-resolution imaging

Z-contrast imaging



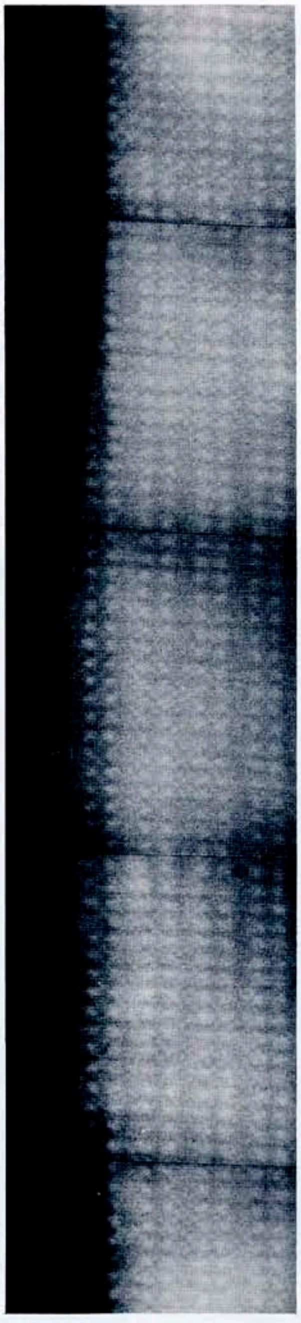
"The images produced by the ORNL technique have characteristics typical of an incoherent image such as a photograph."

In a conventional transmission electron microscope, a broad beam of electrons illuminates the sample and the diffracted beams are focused back to form an interference pattern on a screen. This pattern can take many different forms depending on lens focus and sample thickness, making interpretation extremely complex. With a powerful scanning transmission electron microscope using ORNL's Z-contrast imaging, a fine beam of electrons scans the sample and an annular detector collects electrons scattered to high angles, producing a direct, easy-to-interpret, high-resolution image that is simply a map of the scattering power of the sample. Atoms are always seen white, but the heavier the atom, the brighter the image. The image is a direct view of a material's structure and composition.

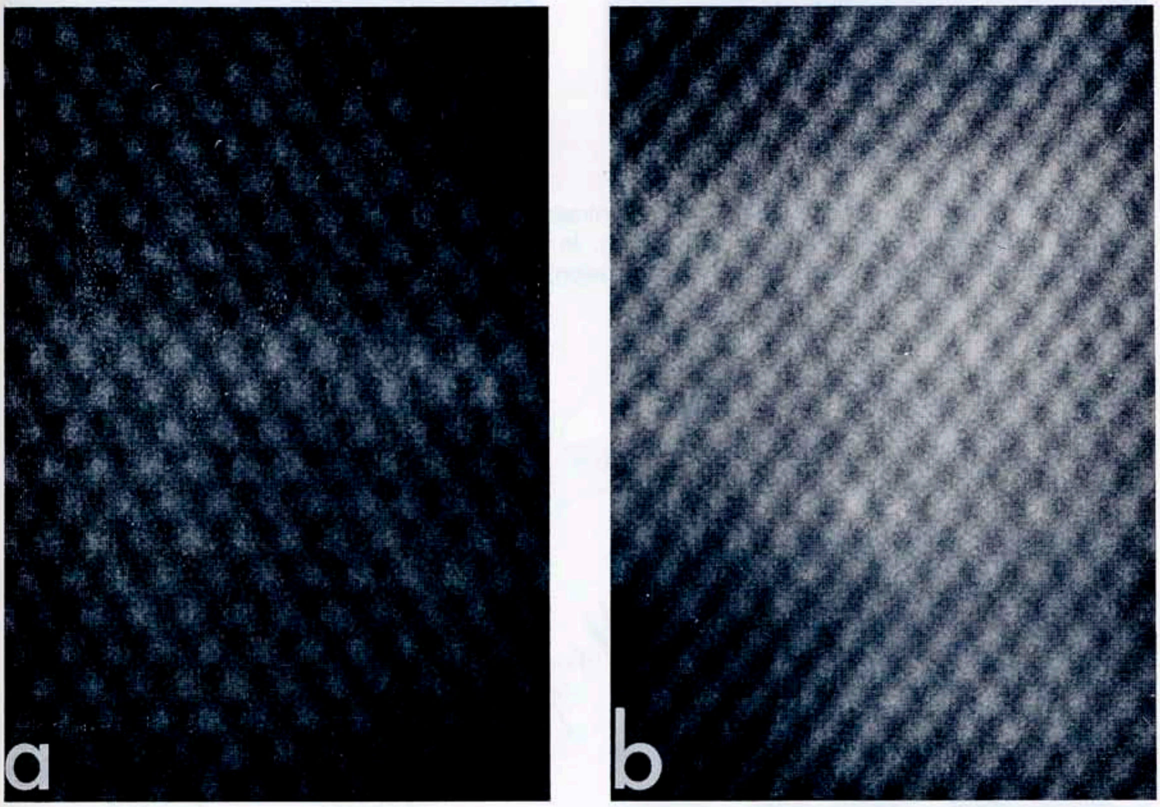
This compositional sensitivity, together with the simple form of the image, enables scientists to locate and study in detail the defects and interfaces of technologically important materials, the regions that ultimately control bulk materials properties. The new Z-contrast image now provides a direct view of the atomic structure and chemistry of these regions. This detailed information could stimulate new theories to explain the microscopic origins of desirable bulk properties, which should lead to the development of new and improved materials.

The photograph on this page shows a cross section of a superconducting thin film of yttrium-barium-copper oxide ($YBa_2Cu_3O_{7-x}$) deposited onto a magnesium oxide (MgO) substrate. Such films are being made and studied because of their potential use in high-speed electronic devices and superconducting motors. The Z-contrast image of the superconducting film is dominated by the orderly rows of bright Ba atoms, with the fainter (lighter)Y atoms barely showing between adjacent Ba rows. The atoms of the substrate are so light in mass that it looks completely dark in the left part of the figure. Undulations, or steps, are clearly present on the surface of the substrate, but the Z-contrast image shows that these do not induce defects in the superconducting film; one or two layers away from the substrate, the current-carrying planes are perfectly continuous. Such information is critical in interpreting the properties of these films. It implies that the current-carrying capacity of these films (1) is not limited in a major way by surface roughness of the substrate and (2) is determined by the boundaries between different grains in the film.

IBM researchers first demonstrated how drastically a grain boundary can affect the current-carrying capacity of a film. They showed that, if two grains are misaligned by only 10°, the current that can be passed from one grain to another drops tenfold. One obvious question arises from this work: because grain boundaries represent an imperfection in the crystal, they are natural sites at which impurities may collect and form a thin insulating layer of some impurity compound. If such a layer exists, then future research should focus on eliminating the impurities. On the other hand, if the current-carrying capacity is reduced by the structural imperfection itself, then the only way to increase capacity is to accurately align all the grains—a very different research direction and a clear case for microscopic examination. Our Z-contrast imaging has demonstrated clearly that low current-carrying capacity is



This cross-sectional view of a superconducting thin film on a magnesium oxide substrate (seen black) shows that continuity of the current-carrying planes is achieved only a few atomic spacings from the substrate surface. (This is a montage of five separate photographs. The horizontal black lines are the joints between photographs, not defects in the film).

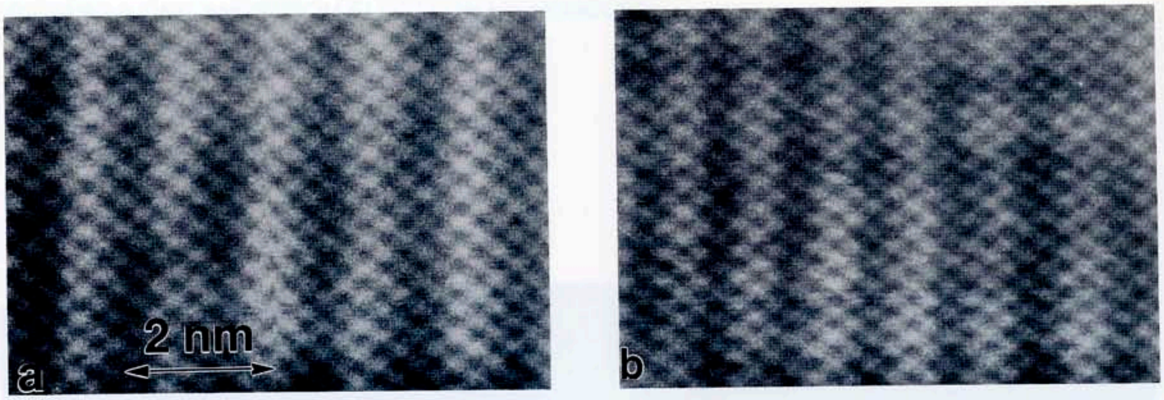


Quantum wells only 2 (left) and 13 (right) atomic layers thick are seen as horizontal bright bands having atomically sharp interfaces. These quantum wells are formed by a gallium arsenide layer sandwiched between layers of indium-gallium-phosphide.

indeed an intrinsic effect of structural imperfection, rather than the presence of impurities. Therefore, we are currently concentrating on methods to accurately align the grains of the film to maximize the current-carrying capacity. This example illustrates how the microscopic view of a material's structure and composition can play a key role in identifying the best approach to improving a material's properties.

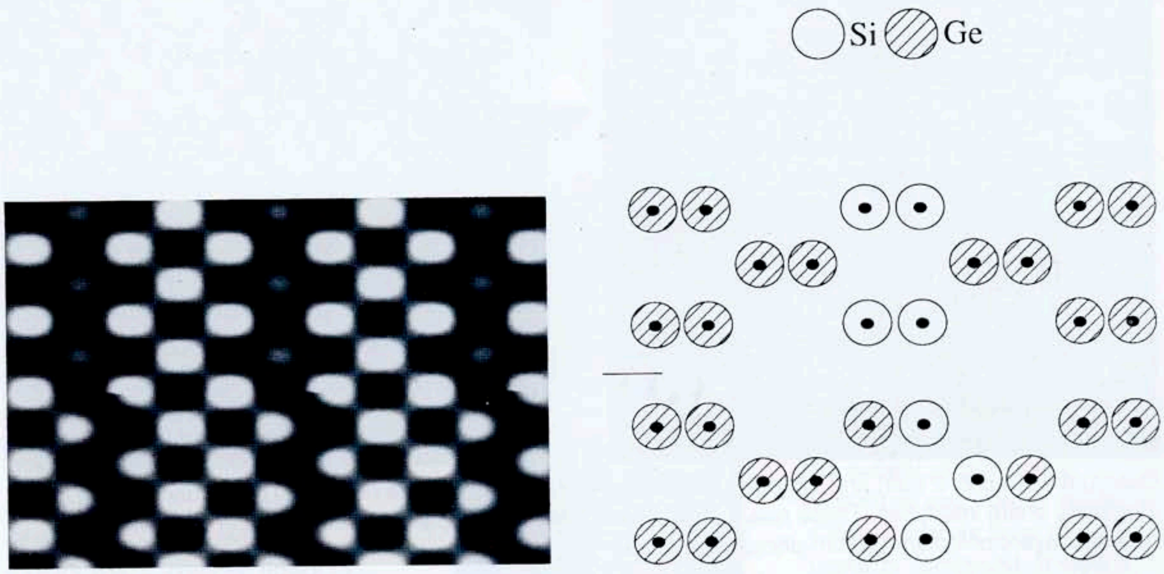
Similar insight can be gained into the behavior of semiconductor materials. A tremendous effort is under way to fabricate artificial structures using techniques such as molecular-beam epitaxy to engineer materials on the atomic scale. For example, a thin layer of one material, perhaps only one or two atomic planes in thickness, can be sandwiched between layers of another to produce a "quantum well" in which electrons behave differently from the way they do in a bulk material. Among their unique properties, quantum wells can be efficient sources of laser light; thus, they are being used in industry to make solid-state lasers for use in fiber-optic communications.

The figure above shows quantum wells of a gallium arsenide layer sandwiched between layers of indium-gallium-phosphide. Such a direct view of the atomic structure and composition is extremely valuable. Suppose that, as is almost always the case in research and development, the prototype device does not perform exactly as predicted. Is this discrepancy because the physics of the device is not sufficiently understood or because the desired structure was not achieved by the complex growth and processing procedures used? The figure at the top of p. 60 shows an example of growth as the problem. In this case, the desired structure was an atomic-scale superlattice—a series of alternating Si and Ge



$(Si_8Ge_2)p$ $(Si_2Ge_6)p$

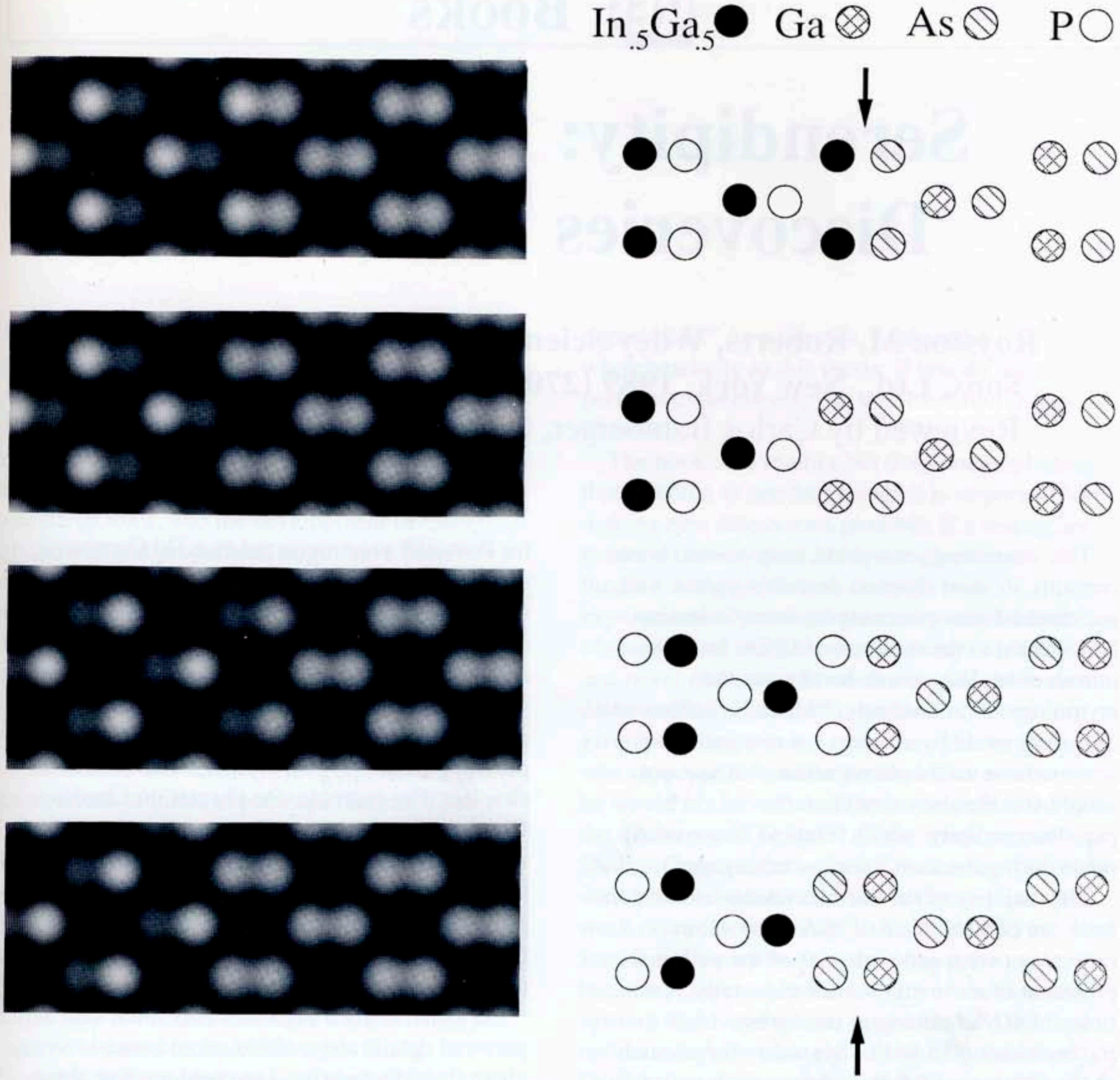
Interdiffusion of atoms is clearly shown in this Z-contrast image of ultrathin silicon-germanium superlattices grown at 400°C using molecular beam epitaxy (left). In the superlattice grown at 350°C (right), the interfaces are more abrupt (indicating less interdiffusion) but irregular because of steps on the growing surface.



Computer simulation of repeating silicon-germanium units (Si_2Ge_6) across a surface step.

layers. Such structures are expected to form the basis for faster transistors and, therefore, faster computers. The critical factor for speeding up electron transport through the material is the "chemical abruptness" of the interface between the Si and Ge—that is, virtually no intermixing of the Si and Ge atoms at the boundaries between Si and Ge layers is desirable. In the left figure, the view of the superlattice shows considerable intermixing; the bright Ge layer was intended to be only a quarter the width of the Si layer. The view of the superlattice on the right shows greatly reduced intermixing because it was grown at a lower temperature than the other superlattice. This image corresponds very closely to the desired structure. The computer simulation in the bottom figure shows that irregularities in the image can arise from surface steps and that surface steps, although too small to be resolved directly, still affect the image in a simply predictable fashion. Features this small would be directly resolvable with the proposed 300-kV STEM, after modification to make use of the Z-contrast technique, because every atomic column would be imaged separately.

The new ORNL capability will be particularly important for imaging quantum wells, which have four possible interface configurations (see the figure on p. 61) because they are fabricated from compound semiconductors. Each structure would have its particular electronic and optical properties. At our current resolution, we cannot distinguish these structures, but at the higher resolution anticipated at 300 kV, each is clearly distinguishable.



Computer-simulated images of quantum wells for a 300-kV high-resolution STEM. Each atomic column can be clearly imaged, and its composition determined. The four possible interface configurations for the quantum wells shown on p. 59 become distinguishable.

ORNL is currently attempting to procure the first high-resolution 300-kV STEM. If successful, we would then have the highest-resolution microscope in the world, perhaps even the first to reach the goal of 1-Å resolution. From a practical standpoint, it is not only the resolution but also the simple nature and compositional sensitivity of Z-contrast imaging that make it such a powerful technique in electron microscopy. The proposed modified STEM will enable us to look inside any material to determine the atomic structure and chemistry in critical areas. This view will provide the fundamental link between a material's growth or processing history and its properties. This capability will have a major impact on materials science and engineering and will improve our nation's competitiveness in these areas. For only a small price, ORNL could offer the key to the understanding of all materials, both natural and artificial.

Biographical Sketch

Steve Pennycook is leader of the Electron Microscopy Group in ORNL's Solid State Division. A native of England, he earned his doctoral degree in solid-state physics at the Cavendish Laboratory of Cambridge University in 1978 and joined the ORNL research staff in 1982. He is a recipient of three Publication Awards (1986, 1988, and 1989) from Martin Marietta Energy Systems, Inc. He received DOE's 1989 Materials Sciences Award for Outstanding Scientific Accomplishment in Solid-State Physics.