MBE deserves a place in the history books

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Molecular beam epitaxy is widely used in research and industry to make semiconductor devices and structures. However, despite its ability to control matter with near-atomic precision, the technique is overlooked in most histories of nanoscience and nanotechnology.

It is common to read that nanotechnology began in December 1959 when Richard Feynman delivered a talk with the title "There's plenty of room at the bottom"[1]. According to this version of events, Feynman's foresight was confirmed when Gerd Binnig and Heinrich Rohrer invented the scanning tunnelling microscope (STM) in 1981, with the atomic force microscope following five years later[2]. Finally, nanotechnology really took off in the late 1990s when governments around the world started investing hundreds of millions of dollars in the field.

However, as a number of historians have pointed out, the history of nanotechnology is more complicated than this: Feynman's lecture, for example, was initially much less influential than is widely believed[3,4]. Simple histories of nanotechnology also tend to overlook some experimental techniques: molecular beam epitaxy (MBE), for instance, played a central role in the growth and development of nanoscience and nanotechnology, yet it is rarely mentioned in popular or historical accounts.

Put most simply, MBE allows researchers to make new materials and nanostructures by, in the words of the *The New York Times*, "spray painting… with atoms"[5]. Pure sources of material are vaporized in separate ovens, and the atoms or molecules released by the sources are transported as a 'beam' to a substrate, where they are deposited. By varying the source materials and controlling the release of the atoms and molecules, scientists can build — one atomic layer at a time — nanostructures with precisely controlled compositions.

BELL LABS

Figure 1 Al Cho (right) and Charles Radice working on an early MBE machine at Bell Labs in 1970. Although MBE was invented before the scanning tunnelling microscope and has had a major impact on both research and industry, it features much less prominently in accounts of the history of nanotechnology.

Compared with other deposition techniques, MBE is relatively slow, depositing as little as a nanometre or so of material per minute and requiring a much higher vacuum and more stringent control of impurities. However, its leisurely rate is also an advantage, allowing for the ordered growth of crystalline films. In fact, the word 'epitaxy' comes from the Greek for 'above' (*epi*) and 'in an ordered manner' (*taxis*).

The origins of MBE lie in the convergence of a number of research areas including solid-state physics, surface physics and materials science in the early 1950s. In 1954, for example, two years after finishing his physics doctorate at the University of Göttingen, Herbert Kroemer had a post at the Central Telecommunications Laboratory run by the German Postal Service in Darmstadt. As the theorist in a small semiconductor research group, he proposed "a non-stoichiometric mixed crystal of different semiconductors with different energy gaps"[6]. Several years later, working for American firms like RCA and Varian, Kroemer suggested several ways to exploit heterostructures made of two different semiconducting materials.

Almost four decades later, after sharing the 2000 Nobel Prize in Physics for his work on semiconductor heterostructures, Kroemer remarked that his lab managers at the time had found it difficult to envisage short-term applications for his ideas, partly because researchers were unable to fabricate the materials needed to translate the ideas into real devices. "I promised myself that if a new technology for building heterostructures arose," he recalled, "I'd get back into it"[7]. Kroemer's patience would be rewarded, but he had to wait for more than a decade.

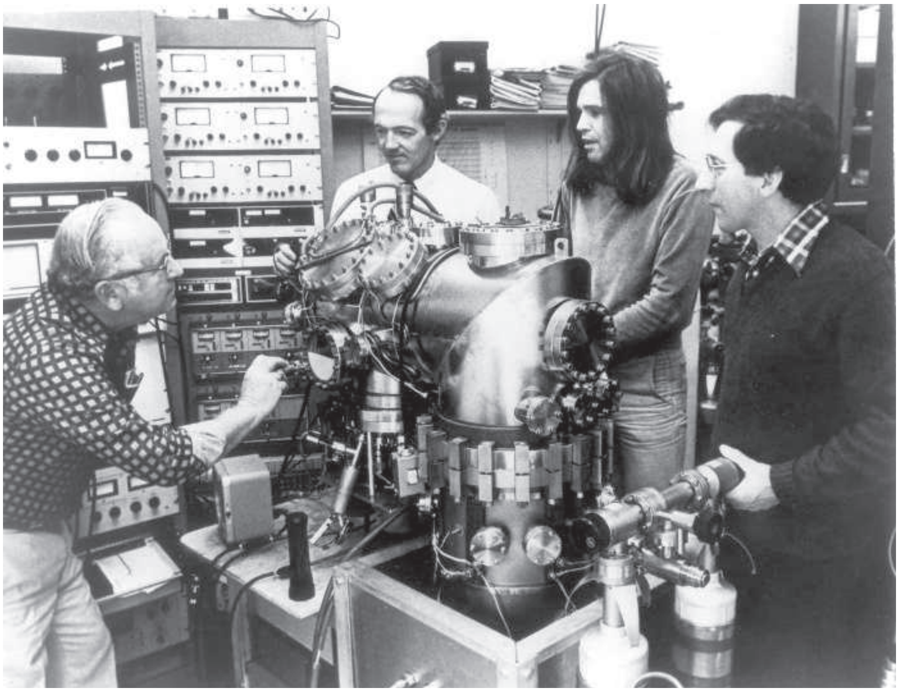

Figure 2 Four pioneers of modulation doping gather around an early MBE machine at Bell Labs in 1978: (left–right) Willy Wiegmann, Art Gossard, Horst Störmer and Ray Dingle. Störmer and his Bell Labs colleague Daniel Tsui shared the Nobel prize for discovering the fractional quantum Hall effect in devices made by Gossard and co-workers with MBE.

BUILDING NEW NANOSTRUCTURES

Throughout the 1960s, researchers grappled with the challenge of making new crystalline materials for semiconductor applications. A good deal of the most significant work was done at the IBM Thomas J. Watson Research Center in Yorktown Heights, New York, and at Bell Laboratories in Murray Hill, New Jersey, between 1968 and 1973. At Bell Labs, for example, John R. Arthur Jr published a paper in July 1968 that described construction of epitaxial gallium arsenide layers using molecular beams of these elements[8], and he filed a patent application for his work in the same year. Managers at Bell and IBM viewed GaAs, with its wide bandgap and high electron mobility, as a good candidate for high-speed electro-optical devices, and researchers liked the fact that the lattice constant of GaAs closely matches that of aluminium arsenide, another III–V semiconductor. Consequently, structures built of alternating layers of AlAs and GaAs would figure prominently in early MBE studies.

Alfred Y. Cho, an ambitious Chinese scientist who is sometimes called the 'father of MBE', started working with John Arthur soon after he joined Bell Labs in 1968 (Fig. 1). Together they blended basic science with instrumental engineering as they improved their fabrication technique and studied the properties of the new materials they made. Cho had previously worked on ion-beam propulsion for space applications, which, he later explained, was the reason why his "MBE systems all look like ion engines!"[9]. At first, however, they could only produce a few hundred angstroms of GaAs per day, grown in layers that were too thin to accurately characterize.

Over at IBM, meanwhile, Leo Esaki and Ray Tsu were grappling with a theoretical concept known as a superlattice. Whereas the heterostructures proposed by Kroemer contained just two layers of different materials, superlattices could be made of many very thin layers of two or more different semiconductors. Moreover, if a superlattice could be built with layers that were shorter than the mean free path of the electrons, a series of allowed and forbidden energy bands would be created in the device. Esaki and Tsu argued that this could prove to be "a valuable area of investigation in the field of semiconductors"[10]. However, when they submitted their first paper on this topic to a journal, it was rejected on the grounds that it was too speculative with "no new physics". (The first paper on the STM was rejected for similar reasons[2].) Luckily for Esaki and Tsu the Army Research Office thought differently and supported their superlattice research for several years.

Around this time Leroy Chang returned to IBM from a sabbatical at the Massachusetts Institute of Technology, and he and Esaki started to build their own MBE equipment, partly as a way to explore superlattices further. Esaki's work, both theoretical and experimental, encouraged both IBM and Bell Labs to explore the use of MBE to make semiconductor structures, and when he shared the Nobel prize in physics in 1973 for earlier work on tunnelling in semiconductors, his influence increased further.

Like Cho and Arthur, the IBM group built their first MBE systems from scratch and had to master various engineering issues, such as developing ways to monitor epitaxial growth *in situ* and control the shutters that allow the atoms into the vacuum chamber. The IBM and Bell groups also had to convince lab managers and outside colleagues that MBE had genuine potential for what we now call nanofabrication. However, the need for pure elemental sources and ultrahigh vacuums led some observers to joke, according to Cho, that the acronym really stood for "mega buck equipment"[11]. Others saw it as a threat to established (but less precise) fabrication techniques rather than an opportunity.

AN MBE COMMUNITY EMERGES

By 1975, both IBM and Bell Labs funded several groups that were actively working to improve the MBE technology and study the new materials they could produce. As interest in MBE grew, experienced researchers began to consider how the technique could be used to build real semiconductor nanostructures, including lasers and diodes. Kroemer, for instance, started to experiment with an MBE machine donated by the US Army once he moved to the University of California, Santa Barbara in 1976.

Meanwhile, other researchers in the US, Japan and Europe (where the German physicist Klaus Ploog played a central role) began to experiment with the new degrees of freedom offered by MBE. Often working in competition with each other and colleagues in academic labs, the IBM and Bell groups incrementally improved MBE technology, introducing refinements such as computer-controlled shutter systems and *in situ* monitoring.

One of the young experimentalists attracted to MBE was Arthur C. Gossard, who arrived at Bell Labs in 1960 after getting his doctorate in physics from Berkeley. At first he continued to study ferromagnetic and superconducting materials with nuclear and electron resonance techniques. Around 1973, however, managers at Bell Labs encouraged him to study defects in semiconductors, especially those used for laser applications. It was the need to fabricate samples for this research that led him to MBE. However, managers initially discouraged him, saying that MBE was too complex for a neophyte to take on. Within a few years, however, Gossard's small group was publishing papers describing the nanoscale semiconductor structures they had built with their rudimentary equipment[12].

By the late 1970s, Gossard's team, like researchers at other labs, were experimenting with a technique that became known as modulation doping (Fig. 2). By carefully controlling the introduction of source materials into the MBE chamber, researchers could implant dopants at precise locations in the semiconductor nanostructure they were fabricating. And by cleverly mixing doped with updoped layers, they could tune the structure and composition of the material and vary its electrical properties. The design and fabrication of new semiconductor materials by MBE has been described by Federico Capasso, another former Bell Lab researcher, as "bandgap engineering".

As the research community became big enough to support dedicated MBE conferences, commercial firms began to pay attention. By the early 1980s, firms like Riber offered the first off-the-shelf MBE systems, which helped make the technique even more accessible. After a decade of incremental improvements and refinements, a fully fledged instrumental community had coalesced around MBE and its ability to fabricate nanostructured materials. Today, there are hundreds of MBE machines in use at university and industry labs throughout the world, with a typical machine costing around a million dollars. Away from the research lab, scaled-up MBE machines produce semiconductor structures for lasers that are found in hundreds of millions of compact disk and DVD players.

Like the STM, MBE has also been associated with Nobel glory. In October 1981 two physicists at Bell Labs, Horst Störmer and Daniel Tsui, started to characterize a sample of Al-doped GaAs that Gossard's team had fabricated with its MBE machine. Störmer and Tsui went on to discover the fractional quantum Hall effect, for which they shared the Nobel prize in 1998 (along with Robert Laughlin, who did the theory). In their Nobel lectures, both physicists acknowledged the pivotal role that Gossard's instrumental expertise had played in their discovery, with Störmer calling such "creators of materials…the true heroes of our trade"[13].

THE VALUE OF HIDDEN HISTORIES

MBE's success as a research tool and as a commercial process that provides the basis for billions of dollars of commerce seems to be partly responsible for its relative invisibility in the history of nanotechnology. MBE originated more than three decades ago at prestigious corporate laboratories that explored the basic science behind the microelectronic devices upon which their business rested. Over time, it matured to become a common yet flexible tool that was essential for research in many areas of nanoscience and technology. Whereas the results of this research make headlines, the instrumental techniques that underpin the research receive little publicity: in many ways, techniques such as MBE are like cinematographers in the movies — their job is to make something or someone else look good.

Recognizing that nanotechnology has a 'hidden history' that involves MBE (as well as, for example, Langmuir–Blodgett films and Arthur von Hippel's work on 'molecular engineering') may be of value to policy makers and researchers grappling with concerns about society's reaction to nanotechnology. When government investment in nanotechnology took off in the US and elsewhere around the turn of the century, new materials and devices for electronics featured prominently.

In recent years, however, attention has shifted to health and safety issues, mostly revolving around relatively low-tech nanoscale particles. Given these concerns, and worries about the social and economic impacts of nanotechnology, it is fair to say that efforts to anticipate the concerns of tomorrow would benefit from a better understanding of the past, including all hitherto hidden histories of nanotechnology. And, if we are going to understand the rich and complex history of nanotechnology, then MBE surely deserves a place in the narrative.

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