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The History and Development of the Helium Ion Microscope

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Summary: The helium ion microscope has recently emerged as a commercially available instrument. However, its roots go back more than 60 years to the development of the field ion microscope in Berlin, first reported in 1951. Over the intervening years, numerous researchers have pursued the development of a gas field ionization source with the goal of producing a suitable source for an ion microscope. This proved to be an elusive goal until early in this century when a number of discoveries led to a successful source, and shortly thereafter, an instrument fully able to exploit its advantages. Many individuals and many technical advances have come together to make this new class of microscope. The long history of this quest is reviewed along with the recent advances that led to the achievement of this milestone. A brief summary of the current status of the technology and its applications are given. SCANNING 33: 1–7, 2011. © 2011 Wiley Periodicals, Inc.

Key words: helium ion microscope, gas field ionization, field ion microscope, ion source, ion microscope, focused ion beam, field emission, history

Introduction

Most new technologies develop at varying rates. Progress lies dormant for long periods of time that are punctuated with periods of frenzied activity. The development of the helium ion microscope (HIM) exhibits these same patterns. The fluctuating rates of progress are sometimes the consequence of technical hurdles that cannot be overcome at a given point in time, but are more readily surmounted when some other supporting technology matures and is brought into play. Market forces too can foster development—or discourage it, as competing technologies flourish or encounter performance limitations. In the past 60 years, many individuals and many technical advances have come together to make this new class of microscope. For this reason, Newton's metaphor, "Shoulders of Giants," applies very well to our advancement of the science and technology of the HIM.

Although the underlying technology of the HIM was conceived 60 years ago, it has become a commercial success in just the past 5 years. This technology relies upon an ion beam that originates in a nanometer scale volume that is adjacent to a single atomic site. To master this technology and control an individual atom for days and even weeks at a time is no simple matter. As if directed by Feynman's prophetic lecture in 1959, we have come to "the bottom" of the atomic length scale, and here we can exploit the properties of individual atoms.

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The Field Ion Microscope

At the Zeiss laboratories in Peabody, Massachusetts, October 11 is a day for celebrating the field ion microscope (FIM). Building on earlier work in Berlin on the FIM (Müller, '51), it was on this day at Penn State University in 1955 that Erwin Müller, and his graduate student Kanwar Bahadur, used their FIM to resolve clearly the individual atoms in a piece of crystalline wire (Müller, '55). For mankind this was a landmark achievement, the first convincing direct visual evidence that the atomic notion of Democritus was indeed true. And most amazingly, this was achieved not with a room-size apparatus and racks of electronics, but a diminutive instrument blown from glass that could readily fit in a backpack. The circumstances of this occurrence have been excellently chronicled by another of Müller's graduate students (Melmed, '96). None of Müller's original FIMs, nor any photographs of such are known to exist (Cutler, 2011), but Figure 1

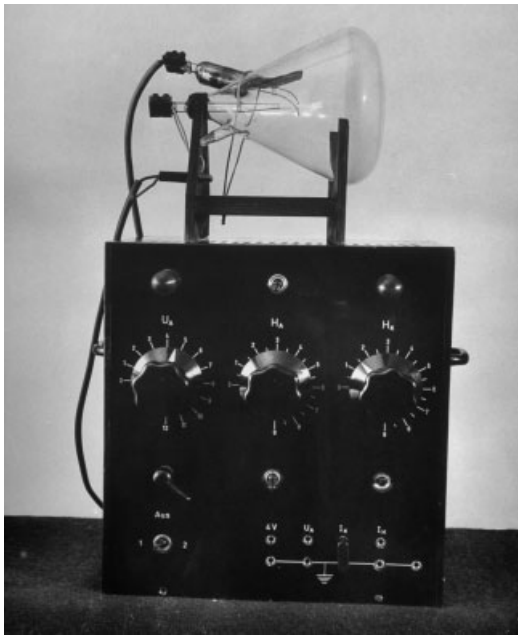

Fig 1. An early example of Erwin Müller's apparatus. Field ion microscopes (and field emission microscopes) were originally blown from glass with the pointed emitter at one end, and a phosphor coating on the opposite end for viewing the emission pattern.

shows a similar device used by Müller during this same time period.

A detailed description of the FIM and its principles of operation can be found in the comprehensive book of this period (Müller and Tsong, '69). The modern FIM uses a stainless steel vacuum vessel instead of glass, but fundamentally it operates much like the blown-glass variety. A sharpened tungsten needle is biased positively relative to an opposing counter electrode that is grounded. Opposite the needle is a grounded fluorescent screen, which provides a view of the FIM emission pattern. In the modern instrument, a microchannel plate helps to intensify the image so it can be more easily seen, but the original instruments required either a dark-adapted eye or photographic plates. The FIM also requires some form of cryogenic cooling such as liquid nitrogen to cool the emitter, the helium gas, and to improve the base vacuum levels. Finally, a small amount of helium is provided in this otherwise high-vacuum vessel. With this relatively simple set of requirements, the FIM operates according to equally simple principles: helium atoms are ionized where the electric field is sufficiently large—adjacent to the most protruding atoms at the apex of the needle. For each of the most protruding atoms, a continuous stream of helium ions is produced, each accelerating along the same trajectory, and contributing to a single bright spot on the fluorescent

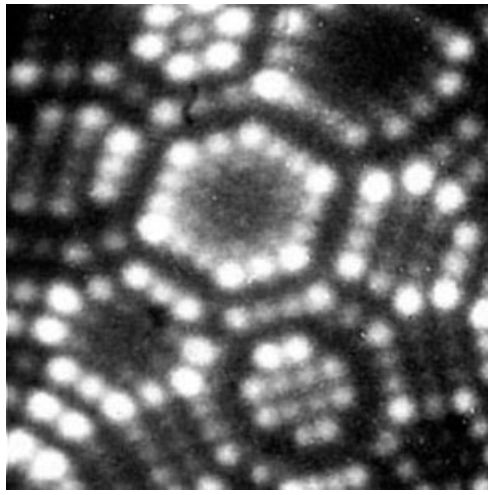

Fig 2. One of Müller's original FIM images as seen on the phosphor. The brightest dots correspond to the most protruding atoms at the end of the needle-shaped emitter. The geometrical patterns arise from the crystalline arrangement of atoms being truncated by a roughly spherical end form. (Image courtesy of the Penn State University Department of Physics). FIM, field ion microscope.

screen. The resulting FIM image (Fig. 2) is crisp and clear, providing valuable insight into the atomic arrangement of the chosen emitter needle. Anyone who has had the pleasure to operate a FIM can attest to the satisfaction of seeing the emitter atoms in real time, and even seeing them re-arrange or be evaporated in response to simple controls that are available to the operator.

Müller's work was soon reproduced at other research facilities throughout the world and Müller was recognized for this achievement with many accolades and awards. Over the next two decades, the blown-glass FIM was replaced with an all-metal apparatus (Ryan and Suiter, '65), and researchers tried new gases such as neon (Nishikawa and Müller, '64), and new emitter materials such as gold (Averback and Seidman, '73). To this day, the basic FIM remains a valuable instrument to scientists for observing or controlling the end shape of a needle with atomic precision (Rezeq *et al*., 2006; Rahman *et al*., 2008) and for studying atomic processes that occur at surfaces (Kellogg, '94).

In the 1960s, FIM technology inspired a series of extensions that led to the 3D Atom Probe (Müller *et al*., '68; Panitz, '73). These instruments (Fig. 3) coupled the high-resolution properties of the FIM with a means of actually identifying the individual atoms on the emitter. The latter is achieved by using high-voltage pulses to field evaporate the atoms one by one and performing time of flight measurements on their subsequent trajectories. For many researchers, the modern 3D Atom Probe (see www.Cameca.com) offers the ultimate in atomic level information. In many regards, this technology

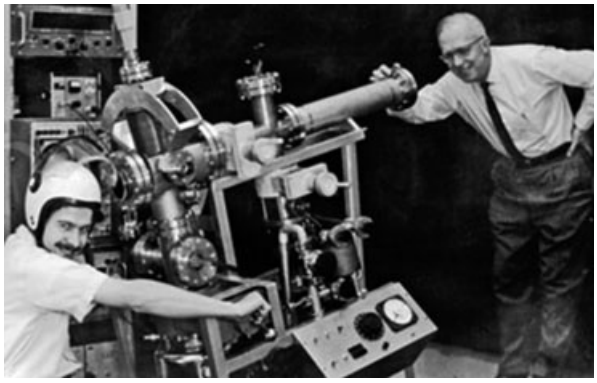

Fig 3. John Panitz (left), Erwin Müller (right), and the 3D atom probe (center). Photo courtesy of J.A. Panitz.

is a "cousin" to the HIM since they are both descendents of Müller's original FIM.

The Gas Field Ionization Source

The prospect of using this same FIM concept to serve as a practical ion source was probably considered by Müller from the very start, but his efforts were instead applied toward the 3D Atom Probe. It was up to others with different interests to adapt the FIM to serve as an ion source. Such an ion source has the virtue of high "reduced brightness," the current per area, per solid angle, per extraction voltage. In the world of electron and ion optics, a high reduced brightness is essential for any beam to be subsequently focused to a small probe size. An excellent review article (Tondare, 2005) explains the exciting potential of the gas field ionization source (GFIS) and the various strategies pursued by different research groups. A small energy spread is another prerequisite to achieving a small focused probe size. And experiments confirmed that the energy spread from one of the individual beamlets could be smaller than 0.5 eV (Ernst *et al*., '93). The only remaining challenge was to produce enough current from a single ionization site. The FIM emission pattern often has hundreds and even thousands of emission sites, all taking a roughly equal share of the available helium gas. Therefore, a single beamlet might have just a picoampere of current or less—not suitable for many GFIS applications. Some major improvements in the GFIS beam current were realized by researchers at the Oregon Graduate Center (Orloff and Swanson, '75) and at Cornell (Hanson and Siegel, '79). Further improvements in emission current relied upon the atomic scale sharpening techniques which had been developed for making good scanning tunneling microscope tips (Fink, '86; Binh, '88). These advances offered the potential of significant improvements to the GFIS performance.

However at this time, the steady GFIS progress had already diminished. The 1980s saw the advent of the gallium liquid metal ion source (LMIS). This new technology was relatively easy to operate (no cryogenics required) and offered high total emission currents (Orloff *et al*., 2003). And for the purpose of sputtering at high rates, the LMIS opened new frontiers of microfabrication (Giannuzzi, 2005).

The further pursuit of the GFIS was then limited to researchers that needed an even smaller probe size (<5 nm), or could not tolerate the electrical, optical, and chemical effects arising from the implanted gallium atoms. Applications such as these included high-resolution imaging, semiconductor mask repair, lithography, and nanofabrication. In the 1970–1990 time frame, GFIS-based systems were pursued by the Levi-Seti group at the University of Chicago (Escovitz *et al*., '75), Seliger's group at the Hughes Research in Malibu (Rensch *et al*., '79), Aihara's group at JEOL (Aihara, '87), and Siegel's group at Cornell (Hanson and Siegel, '81). Each of these groups attempted to collect sufficient current by using multiple emission sites on the emitter and consolidating them into a single beam. Subsequently, the beam could be focused and scanned as necessary for imaging or lithography applications. Building such a system turned out to be difficult for several reasons. First, the individual beamlets diverged rapidly, making it difficult to effectively consolidate the beams. Second, the emission sites tended to fluctuate rapidly causing inconsistencies in the patterns or the images. Third, the designs typically used liquid helium at high consumption rates, making GFIS imaging systems expensive to operate. Finally, the boiling cryogen (or any cryomechanical refrigerator) introduced vibrations into the system that caused the Angstrom-sized emission site to appear many times larger. The Levi-Seti group achieved some level of success using a hydrogen beam (Fig. 4) to produce transmission images of a variety of samples (Escovitz *et al*., '75.)

Within the Cornell group, Gary Hanson, Paul Schwoebel, and Joel Cubby began working on tip shaping techniques to concentrate the GFIS emitting sites so that the emission current could be more easily consolidated. Inspired by their work Dr. Siegfried Kalbitzer at the Max Planck Institute (MPI) in Heidelberg, Germany gave several of his graduate students the assignment of making a GFIS "SuperTip" that could one day become the ion source for a GFIS scanning system. After 8 years of work, the MPI group perfected a fabrication recipe that provided high brightness (Wilbertz *et al*., '92). The recipe was reported to be reproducible, and able

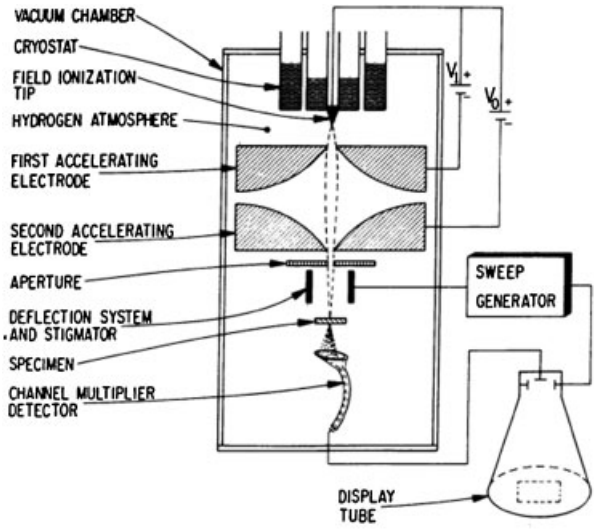

Fig 4. Diagram of the Levi-Seti hydrogen GFIS for a transmission ion microscope. (Reproduced with permission of Wiley-Blackwell).

to produce a GFIS with stable emission current for a variety of gas species.

Early Commercial Efforts at Micrion and FEI

In the mean time, Micrion Corporation (Peabody, MA) was making ion beam instruments that used the gallium LMIS for the semiconductor and data storage markets. The president of Micrion (Nick Economou) had been monitoring the publications of the MPI group and determined that their "Super Tip" GFIS might have some advantages relative to LMIS. Micrion's chief scientist (Bill Thompson) traveled to MPI to qualify their GFIS technology for potential mask repair applications. Also playing a consulting role in this project was Bill Ward, co-founder, and CTO of Micrion, who would later make key discoveries that enabled the ultimate success of the HIM.

In the period from 1992 to 1994, Micrion was awarded two contracts, one from DARPA and one from SEMATECH to build a "Super Tip" based phase shift mask repair system. Micrion licensed the "Super Tip" technology from MPI and the system construction started at Micrion. Bill Thompson gathered his GFIS team together and began his "Kick Off" meeting with these prophetic words: "You are about to build a system that requires one atom to stay in place for days on end. If you think this is too high a risk program feel free to speak up." No one did. The GFIS was cooled by an ingenious cryo-refrigerator that could maintain the source at a temperature of approximately 10 K. The gun housing was designed to permit the source to be tilted or shifted using a "hexapod" source mounting scheme.

Fig 5. The early Micrion GFIS research platform.

The column and remaining system hardware were modified from a standard Micrion 8000 mask repair system (Fig. 5).

In late 1995, the GFIS "SuperTip" based system integration was complete (Thompson *et al.*, '95) and phase shift masks were repaired with helium, neon, and argon beams. Although the currents and sputter yields obtained were not adequate for high speed applications, a probe size of 2–3 nm was obtained with probe currents approaching 20 pA. Although the demonstration was successful, the commercialization was not practical due to issues with stability and lifetime. Also, the vibrations of the source arising from the cryo-refrigerator limited the ultimate performance. The "Super Tip" GFIS was relegated to a skunk works project.

In 1999, FEI Co. (Hillsboro, OR) acquired Micrion Corporation. Bill Ward took over the GFIS R&D activities within FEI and worked to improve the GFIS lifetime and stability. In 2002, FEI chose to spin this project to an external R&D team under the acronym ALIS (Atomic Level Ion Source). The team consisted of Bill Ward, Lou Farkas, and Randy Percival, with Nick Economou, who had earlier left FEI, acting in a consulting role. During the next 3 years, the "Super Tip" concept was abandoned in favor of a build process that would be more consistent. When this process was optimized the ALIS group was reformulated into its own company with venture capital funding in the March of 2005. The new company was also known as ALIS, but now with the acronym standing for Atomic Level Imaging Systems.

ALIS Corporation

The technical challenges to a commercially viable GFIS ion beam system are indeed daunting: The vacuum requirements are extreme when one considers that a single atom landing at or near the

emission site would drastically alter the otherwise stable emission current. Also the emitter must be cryogenically cooled, yet the boiling cryogen or the mechanical cryo-cooler must not produce even nanometer scale vibrations at the emitter. The high voltages involved are near the breakdown strength even under ideal circumstances. In this case the risk is enhanced because the insulators are coated with cryo-pumped gases and the high fields threaten to ignite the helium into a plasma. To realize the full potential of this ion beam, the sample stage must be immune to vibrations, while the electronics must be free of any noise that could deflect the beam by even a fraction of a nanometer. In the April of 2005, the ALIS team was assembled to tackle these challenges. The team of 20 scientists and engineers, led by John Notte, Ray Hill, and Richard Comunale, was given the goal of producing a 1-nm resolution instrument in less than 1 year.

Both the platform development and the GFIS research continued at a rapid pace (Ward *et al.*, 2006). Although a variety of source build processes had been reported in the literature (Fink, '86; Binh, '88; Kuo *et al.*, 2006; Rezeq *et al.*, 2006), none of these could be made to work consistently on a practical instrument. A breakthrough came later that same year when a reliable in situ process was found that caused the emitter's end shape to facet repeatedly. The faceting process encourages the atoms near the apex to arrange themselves into a stable three sided pyramid. The very apex of the pyramid is likely a single atom, but in the presence of high fields this atom is soon evaporated away, leaving a triple of atoms, the "trimer" (Fig. 6). This faceted end form with the trimer at the apex gives rise to an electric field that peaks at the apex and quickly diminishes in the surrounding region. Only at the vertex—where three facets come together—is the field strong enough to produce helium ionization. Hence, the same limited gas supply is shared by just these three atoms near the vertex, producing about 100 pA of emission current. Under normal operations, one of the three beamlets is aimed down the column, while the other two beamlets are wasted on the beam defining aperture. Once discovered, the in situ process was refined and optimized to the point that it became quite reliable and suitable for automated tip formation in a commercial instrument.

By the early summer of 2006, the system was completed. Dubbed the "Looking Glass" the system was able to produce images with a resolution of 1 nm (Scipioni *et al.*, 2007).

Zeiss

By July 2006, several successful demonstrations were completed, and there was considerable interest in this new technology. Although there were many suitors, the relationship with Zeiss seemed to be a perfect match. Zeiss had accumulated a broad knowledge of SEM and FIB applications. They also knew what application needs were still unaddressed by their present suite of instruments. Under the Zeiss banner, the first HIM (now dubbed ORION) was shipped to NIST in Gaithersburg, MD into the group led by Mike Postek and Andras Vladar. A series of universities and research institutes followed in a steady succession. All the while, there were significant platform improvements that increased the emission current and reduced image vibrations allowing image resolution of 0.35 nm (Fig. 7).

The early adopters of the ORION were intent upon studying the fascinating new properties of the instrument. After all, a focused helium ion beam

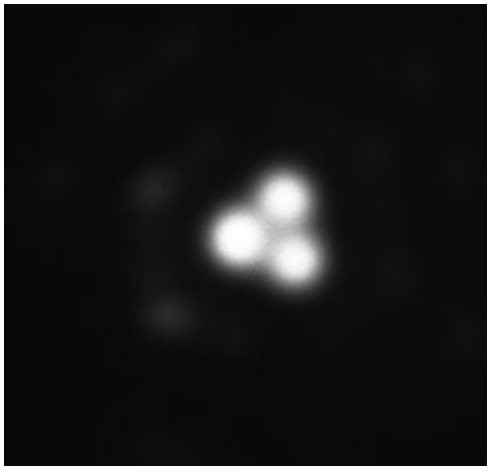

Fig 6. The customary trimer that resides on the apex of the three sided pyramid. The next atomic shelf provides a much reduced electric field, so almost no ionization takes place there.

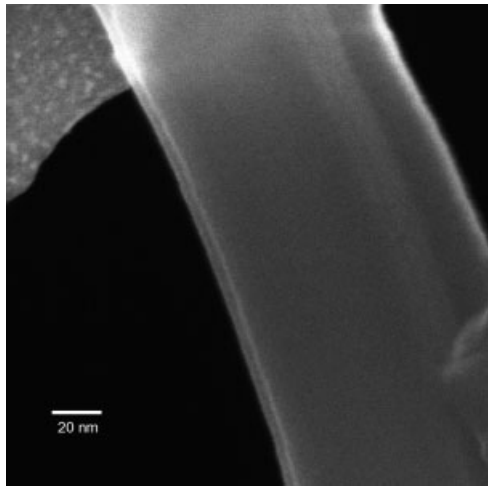

Fig 7. A 200-nm FOV image of an asbestos fiber on a carbon film. An edge analysis indicates that the image resolution is 0.35 nm.

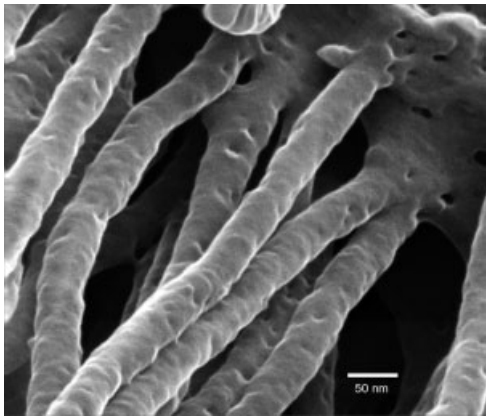

Fig 8. Kidney cells imaged with a 500-nm field of view. Note that the HIM does not require a conductive coating, which would otherwise obscure fine details. Image courtesy of Dennis Brown of Massachusetts General Hospital.

interacts with the sample in a fundamentally different way than an electron beam or a gallium beam. The most significant differences in image quality compared with the SEM are ultimately attributed to the HIM's surface sensitivity, minimal charging, and high resolution. In the most recent year, we are seeing an increase in the ORION users that are as intent on studying their own samples as on studying the instrument. Figure 8 shows a high magnification image of the renal epithelial cells from a mouse kidney. This image was acquired with the use of a low-energy electron flood gun that effectively neutralizes the positive surface charge. Because of this, the sample was imaged without the need for a conductive coating, which is commonly required in the SEM to prevent charging artifacts.

Aside from its strength as an imaging instrument, the ORION has also been used to explore nanofabrication. Some of the earliest results used the helium beam to precisely deliver stress in a membrane (Arora, 2007). Recently, the beam has been used for lithography and beam-assisted chemistry (Veldhoven, 2010). A team at the Harvard University (Bell *et al*., 2009) has demonstrated patterning of graphene films via sputtering using the ORION system. Another team at the National University of Singapore (Pickard and Scipioni, 2009) has used the helium ion beam to sputter nanometer scale patterns in graphene films (Fig. 9). In the recent scientific literature, the graphene milling challenge is "most convincingly demonstrated by helium ion microscopy" (Zhou and Loh, 2010).

Conclusion

After a long and arduous evolution, the HIM has been successfully commercialized. Over the last 60 years, many researchers have contributed to the incremental advancements that have brought the instrument to its current level of performance. We observe now that the instrument is increasingly being used to make significant advances in the imaging of difficult samples—going beyond the capabilities available through a traditional SEM. In addition to imaging, nanofabrication has arisen as one of the major applications of the instrument. For perspective, consider that the SEM was developed and studied in the 1930s and 1940s before an industrial application was found in 1958 (Oatley, '72). In the future, we anticipate that the HIM will make significant contributions to some of the most challenging imaging and nanotechnology applications.

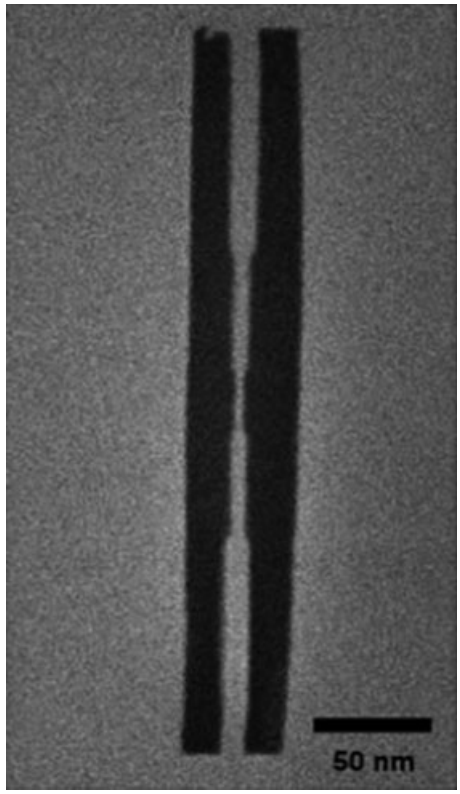

Fig 9. Milling of Graphene with a focused helium ion beam. Image courtesy of Daniel Pickard of National University of Singapore.

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