

## From the bottom up: Manipulating nanoribbons at the molecular level

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Bottom-up synthesis of graphene nanoribbons from molecular building blocks (left). The resulting ribbon, or heterojunction, has varied widths as a result of different width molecules; and a scanning transmission microscope image of graphene nanoribbon heterojunction (right), with larger-scale inset of multiple ribbons. Credit: Lawrence Berkeley National Laboratory

Narrow strips of graphene called nanoribbons exhibit extraordinary properties that make them important candidates for future nanoelectronic technologies. A barrier to exploiting them, however, is the difficulty of controlling their shape at the atomic scale, a prerequisite for many possible applications.

Now, researchers at the US Department of Energy's (DOE) Lawrence



Berkeley National Laboratory (Berkeley Lab) and the University of California, Berkeley, have developed a new precision approach for synthesizing graphene nanoribbons from pre-designed molecular building blocks. Using this process the researchers have built nanoribbons that have enhanced properties—such as position-dependent, tunable bandgaps—that are potentially very useful for next-generation electronic circuitry.

The results appear in a paper titled "Molecular bandgap engineering of bottom-up synthesized graphene nanoribbon heterojunctions," published in *Nature Nanotechnology*.

"This work represents progress towards the goal of controllably assembling molecules into whatever shapes we want," says Mike Crommie, senior scientist at Berkeley Lab, professor at UC Berkeley, and a leader of the study. "For the first time we have created a molecular nanoribbon where the width changes exactly how we designed it to."

## Nanoribbons past and present

Previously, scientists made nanoribbons that have a constant width throughout. "That makes for a nice wire or a simple switching element," says Crommie, "but it does not provide a lot of functionality. We wanted to see if we could change the width within a single nanoribbon, controlling the structure inside the nanoribbon at the atomic scale to give it new behavior that is potentially useful."

Felix Fischer, Professor of Chemistry at UC Berkeley who jointly led the study, designed the molecular components to find out whether this would be possible. Together, Fischer and Crommie discovered that molecules of different widths can indeed be made to chemically bond such that width is modulated along the length of a single resulting nanoribbon.



"Think of the molecules as different sized Lego blocks," explains Fischer. Each block has a certain defined structure and when pieced together they result in a particular shape for the whole nanoribbon. "We want to see if we can understand the exotic properties that emerge when we assemble these <u>molecular structures</u>, and to see if we can exploit them to build new functional devices."

Until now, nanoribbon synthesis has mostly involved etching ribbons out of larger 2D sheets of graphene. The problem, according to Fischer, is that this lacks precision and each resulting nanoribbon has a unique, slightly random structure. Another method has been to unzip nanotubes to yield nanoribbons. This produces smoother edges than the "top-down" etching technique, but it is difficult to control because nanotubes have different widths and chiralities.

A third route, discovered by Roman Fasel of Swiss Federal Laboratories for Materials Science & Technology along with his co-workers, involves placing molecules on a metal surface and chemically fusing them together to form perfectly uniform nanoribbons. Crommie and Fischer modified this last approach and have shown that if the shapes of the constituent molecules are varied then so is the shape of the resulting nanoribbon.

"What we've done that is new is to show that it is possible to create atomically-precise nanoribbons with non-uniform shape by changing the shapes of the molecular building blocks," says Crommie.

## **Controlling quantum properties**

Electrons within the nanoribbons set up quantum mechanical standingwave patterns that determine the nanoribbon's electronic properties, such as its "bandgap". This determines the energetics of how electrons move through a nanoribbon, including which regions they accumulate in and



which regions they avoid.

In the past, scientists spatially engineered the bandgap of micron-scale devices through doping, the addition of impurities to a material. For the smaller nanoribbons, however, it is possible to change the bandgap by modifying their width in sub-nanometer increments, a process that Crommie and Fischer have dubbed "molecular bandgap engineering." This kind of engineering allows the researchers to tailor the quantum mechanical properties of nanoribbons so they might be flexibly used for future nanoelectronic devices.

To test their molecular bandgap engineering, Crommie's group used scanning tunneling microscopy (STM), a technique that can spatially map the behavior of electrons inside a single nanoribbon. "We needed to know the <u>atomic-scale</u> shape of the nanoribbons, and we also needed to know how the electrons inside adapt to that shape," says Crommie. UC Berkeley professor of physics Steven Louie and his student Ting Cao calculated the electronic structure of the nanribbons in order to correctly interpret the STM images. This "closed the loop" between nanoribbon design, fabrication, and characterization.

## New directions toward new devices

A major question in this work is how best to build useful devices from these tiny molecular structures. While the team has shown how to fabricate width-varying nanoribbons, it has not yet incorporated them into actual electronic circuits. Crommie and Fischer hope to use this new type of nanoribbon to eventually create new device elements - such as diodes, transistors, and LEDs - that are smaller and more powerful than those in current use. Ultimately they hope to incorporate nanoribbons into complex circuits that yield better performance than today's computer chips. To this end they are collaborating with UC Berkeley electrical engineers such as Jeffrey Bokor and Sayeef Salahuddin.



The required spatial precision already exists: the team can modulate nanoribbon width from 0.7 nm to 1.4nm, creating junctions where narrow nanoribbons fuse seamlessly into wider ones. "Varying the width by a factor of two allows us to modulate the bandgap by more than 1eV," says Fischer. For many applications this is sufficient for building useful devices.

While the potential applications are exciting, Crommie points out that a central motivation for the research is the desire to answer basic scientific questions like how <u>nanoribbons</u> with non-uniform width actually behave. "We set out to answer an interesting question, and we answered it," he concludes.

**More information:** Molecular bandgap engineering of bottom-up synthesized graphene nanoribbon heterojunctions, *Nature Nanotechnology*, DOI: 10.1038/nnano.2014.307

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