Facile deterministic cutting of 2D materials for twistronics using a tapered fibre scalpel

L D Varma Sangani, R S Surya Kanthi, Pratap Chandra Adak, Subhajit Sinha, Alisha H Marchawala, Takashi Taniguchi, Kenji Watanabe and Mandar M Deshmukh

1 Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400005, India
2 Department of Applied Physics, Faculty of Technology & Engineering, The Maharaja Sayajirao University of Baroda, Vadodara 390001, India
3 National Institute for Materials Science (NIMS), Tsukuba, Ibaraki 305-0044, Japan

E-mail: deshmukh@tifr.res.in and mandar.m.deshmukh@gmail.com

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Abstract

We present a quick and reliable method to cut 2D materials for creating 2D twisted heterostructures and devices. We demonstrate the effectiveness of using a tapered fibre scalpel for cutting graphene. Electrical transport measurements show evidence of the desired twist between the graphene layers fabricated using our technique. Statistics of the number of successfully twisted stacks made using our method is compared with h-BN assisted tear-and-stack method. Also, our method can be used for twisted stack fabrication of materials that are few nanometers thick. Finally, we demonstrate the versatility of the tapered fibre scalpel for other shaping related applications for sensitive 2D materials.

Supplementary material for this article is available online

Keywords: 2D materials, twistronics, tear-and-stack
Figure 1. Cutting flakes with TFS and integration with stacking microscope: (a) Schematic of the cutting process using a Tapered Fibre Scalpel (TFS). (b) Photograph of the TFS mounted like a cantilever on a glass slide. The TFS is mounted on a glass slide and bent using scotch tape. The glass slide is then mounted on the micrometer stage to cut flakes. (inset): Optical image of the TFS. (c) and (d) Image of a graphene flake before cutting and after cutting with the TFS respectively.

2D heterostructures are typically made using h-BN assisted tear-and-stack method [15], which is a challenging and probabilistic process. An alternate way of controllably making twisted structures is to slice graphene before making the twisted heterostructure. Most exfoliated flakes are small, of the order of 10 µm, so precise control is required to cut flakes. Oxidation based AFM cutting [16] and electron beam lithography (EBL) patterning [15] followed by etching of the flake are some methods to cut a flake. However, using oxidation based AFM cutting or EBL for sectioning may leave residue and is time-consuming. There is a need for a reliable and facile technique for cutting 2D flakes.

In this paper, we present a simple method to section 2D flakes using tapered fibre scalpel (TFS), which can be integrated with the microscope used for stacking. We demonstrate the sectioning of graphene flakes (dimensions of about 30 µm) and make twisted double bilayer graphene devices. The TFS is very versatile, and we also use it to shape other sensitive 2D materials which might get contaminated by fabrication using lithographic techniques. Further, the TFS can also be used to cut materials a few nanometers thick with ease.

Optical fibres can be tapered using a fibre pulling process that is used extensively for many applications [17] or using HF solution [18]. There are also other examples in literature where AFM tips mounted on a micrometer stage are used in an analogous fashion for cutting 2D materials using the ‘ploughing’ technique [19]. We use the fibre pulling process to make tapered optical fibres (details of optical fibre preparation are provided in S1 of supplementary information (stacks.iop.org/Nano/31/32LT02/mmedia)) and use them as scalpsels for slicing 2D materials. The schematic of the cutting process is depicted in figure 1(a).

We exfoliate h-BN and graphene flakes on 280 nm SiO$_2$/Si substrate. The TFS is placed on a glass slide like a cantilever, as shown in figure 1(b), and the glass slide is fixed on the motorized micrometer precision stage, also used for stacking. We then bring the TFS near the flake and slowly raise the bottom stage containing the chip. Using the focus of the optical microscope, we determine when the chip on the bottom stage comes in contact with the TFS; after this, the flake is cut. We move the TFS in the direction we want to cut at a low speed of 1 µm s$^{-1}$. Higher speed may lead to folding near the edge of the flake. Figure 1(c) and (d) are the optical images of the bilayer graphene flake before and after cutting using TFS.

By changing the relative orientation of the chip before cutting, and moving with low speed while cutting, we can slice graphene flakes along any direction. Further, this method of slicing of flakes makes use of a stacking stage, which is readily available for making van der Waals heterostructures. The TFS is easy to fabricate, and also ensures that the flake is minimally contaminated.

More details of the cutting procedure can be found in S1 of supplementary information. Videos of the cutting process are also provided as supplementary materials.

After slicing the graphene, the twisted devices are made using conventional van der Waals pick up technique. Figure 2(a) shows a schematic of the pick-up process of the two portions of the cut graphene flake. To prepare the twisted
graphene stack, we first align the h-BN on a PC-PDMS stamp with the graphene such that it covers only one section of the sliced graphene flake [15] as shown in figure 2(b). We thus pick up one cut portion of the graphene. We then rotate the chip with the second portion of graphene by a specified angle $\theta$ and pick-up the second half of the graphene using van der Waals forces to complete the twisted stack as shown in figure 2(c). The color enhanced optical micrographs from figure 2(b) and (c) can be found in S4 of the supplementary information. We shape the stack into a Hall bar geometry via reactive ion etching [20]. The resulting Hall bar devices of the twisted double bilayer graphene device that proves the efficacy of our technique. Figure 3(a) shows image of a device fabricated using the TFS assisted slice-and-stack method. Figure 3(b) shows the 4-probe resistance as a function of gate induced charge density and perpendicular electric displacement field (D) of a twisted double bilayer graphene device fabricated using the TFS assisted slice-and-stack. The data from the twisted double bilayer graphene device can be used to extract the bandgaps and bandwidths of the system [22].

Figure 3(c) shows the device resistance as a function of gate voltage for a device made using this method. The moiré peaks are distinctly visible. Moiré patterns were consistently obtained in devices twisted with this method. More resistance as a function of gate voltage curves showing moiré peaks for other devices are provided in S3 of supplementary information.

Thus far, we have used the TFS for slicing graphene quite frequently. However, the TFS can also be used for shaping other 2D flakes in a regular desired geometry. For example, 2D materials like the high T$_c$ superconductor Bi$_2$Sr$_2$Ca$_2$Cu$_2$O$_{10+\delta}$ (BSCCO) in a few unit-cell limit are of great interest [23–25]. However, the time-dependent degradation of BSCCO in ambient conditions [23] makes it difficult to work with. The minimal invasiveness and rapidity of cutting with the TFS, along with its simple integration with the micrometer precision stage used for stacking, make the TFS ideal for shaping purposes. We regularly use the TFS to rapidly shape BSCCO flakes without using any chemicals. This shaping step is crucial to our devices, as it helps us control the device geometry. Figure 4(a) and (b) show an optical micrograph of a BSCCO device before and after shaping respectively.

## Table 1. Comparison table of success of first half flake pickup using tear-and-stack and using slice-and-stack method.

| Method                | Twisted stack number | 1st section picked up at |
|-----------------------|----------------------|--------------------------|
| Tear-and-stack        | Stack 1              | 6th attempt              |
| Stack 2               |                      | 3rd attempt              |
| Stack 3               |                      | 7th attempt              |
| Stack 4–10            | Not successful       |                          |
| TFS assisted          | Stack 1              | Not successful           |
| slice-and-stack       | Stack 2–10           | 1st attempt              |

As is clear from the data, using the slice-and-stack method, we are able to make twisted stacks with ease in the first attempt reliably. The yield of the devices ‘twisted’ successfully is better than the tear-and-stack method. Additionally, unintentional straining in the stack [21] can result from the tear-and-stack method, which can be avoided using the slice-and-stack method.

Commonly used methods for cutting graphene before stacking includes oxidation based AFM cutting [16] and EBL [15]. Cutting graphene using oxidation with the help of AFM tips takes a large amount of time (around an hour). EBL patterning is another reliable method to slice flakes, but is time-consuming and can cause contamination of flakes. Using the TFS, we can achieve rapid cutting of graphene flakes (at a speed of $1 \mu m s^{-1}$) with minimal contamination.

Now, we discuss the electrical response of the fabricated twisted double-bilayer graphene device that proves the efficacy of our technique. Figure 2(a) shows an optical micrograph of a BSCCO device fabricated using the TFS assisted slice-and-stack method. Figure 2(b) and (c) can be found in S4 of the supplementary information. We shape the stack into a Hall bar geometry via reactive ion etching as irregular edges can influence device characteristics even in large systems [20]. The resulting Hall bar devices of the twisted double bilayer stacks are electrically characterized here. The twisted devices made show moiré bands, proving that it is indeed twisted. We present evidence of the moiré peaks in one of the devices made using the slice-and-stack method subsequently.

The statistics of 10 consecutive attempts of making twisted devices using the tear-and-stack method and slice-and-stack method is shown in table 1. As is clear from the data, using the slice-and-stack method, we are able to make twisted stacks with ease in the first attempt reliably. The yield of the devices ‘twisted’ successfully is better than the tear-and-stack method. Additionally, unintentional straining in the stack [21] can result from the tear-and-stack method, which can be avoided using the slice-and-stack method.
Figure 3. Data from a twisted graphene device made using slice and stack method: (a) Optical micrograph of a fully fabricated dual gated twisted double bilayer graphene device made using the slice-and-stack technique. Dual gates allow independent control over the perpendicular electric field applied on the device and charge density of carriers in the device. (inset) Colour enhanced optical image of the twisted double bilayer graphene stack made using this method. The two graphene layers, covered by hBN from top and bottom sides, are clearly visible in the tweaked image. (b) A color scale plot of 4-probe resistance of the device having a twist angle $\theta = 1.18^\circ$, as a function of charge density ($n$) and perpendicular electric displacement field ($D$). It demonstrates features which are characteristic to a small-angle twisted double bilayer graphene device. (c) A line slice plot of resistance vs. charge density is shown for $D/\varepsilon_0 = -0.25$ V nm$^{-1}$, showing all the three resistance peaks corresponding to the charge neutrality point gap at $n = 0$ and the two moiré gaps at $n = \pm 3.2 \times 10^{12}$ cm$^{-2}$.

Figure 4. Shaping other materials using TFS: (a) and (b) Optical images of the BSCCO device before and after shaping of BSCCO flake respectively. Inset: higher magnification image. (c) and (d) optical images of ABA and ABC flake before and after separating ABA and ABC portions respectively. Encircled region in (c) is ABC part which was identified using Raman analysis. (e) and (f) optical images of h-BN flake before and after cleaning its surroundings respectively.
The TFS were also used in isolation of an area with ABC stacked trilayer graphene stacking in a larger flake. ABC trilayer is typically a smaller fraction of exfoliated flake and easily converts to ABA graphene on heating [26] if in contact with ABA trilayer graphene. So, it is necessary for it to be isolated from surrounding ABA graphene regions before being fabricated into a device. Traditional lithography techniques involve heating and chemicals, which can result in contamination. With the TFS, we easily isolate the ABC trilayer graphene from the ABA trilayer graphene. Figure 4(c) and (d) show the trilayer graphene flake before and after the ABC trilayer region is separated out.

In general, stacking picks up the flakes in the neighborhood of our flake of interest, which can be an issue while making devices. We remove any unwanted flakes around our flake of interest easily with the help of the TFS and a micrometer precision stage. Figure 4(e) and (f) show images of a h-BN flake before and after its surroundings are cleaned with the TFS respectively. Other examples of slicing 2D flakes are provided in S2 of supplementary information.

We have demonstrated the efficacy of the slice-and-stack technique of making twisted graphene stacks. Additionally, the slice-and-stack technique can be used for graphene slicing with a TFS for rapidity, reliability, and minimal contamination. The tapered fibre scalpel described is easily integrated with existing stacking setups and easy to use. In general, the TFS can be used for slicing any 2D materials rapidly and can also be used to create twisted stacks of other few nanometer-thick materials like transition metal dichalcogenides (TMDCs). Additionally, it can also be used for cleaning areas near a flake of interest while stacking.

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ORCID iDs

Kenji Watanabe https://orcid.org/0000-0003-3701-8119
Mandar M Deshmukh https://orcid.org/0000-0002-1401-1080

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