

## Graphene ablation by an optical fiber delivered laser

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Nano and micropatterning graphene is of ultimate importance for graphene-based electronic, photonic and plasmonic devices. In the majority of the reported work, patterning is achieved via optical or electron-beam lithography, with the latter offering nanometer-scale spatial resolution. However, lithography is a multi-step and high cost process. Importantly, it requires a resist to be deposited onto graphene, which can be a source of contamination. Laser ablation, on the other hand, is an attractive, single-step, alternative patterning method, which requires significantly less instrumentation and involves no direct contact with the sample. Laser ablation has been successfully used to generate holes and patterns in graphene [1-6], with feature sizes as small as  $\sim 0.5 \mu\text{m}$  [3,6]. Most reports to date describe graphene ablation using femtosecond lasers [1-4,6], with some mentioning the use of a controlled, inert, atmosphere [4] and the appearance of an intense Raman D band around the hole [1,2,5]. Outside the graphene area, fiber delivered-laser beams have been responsible for a technological revolution in micropatterning and microcutting, allowing for simpler and more flexible optical setups [7]. However, the fiber delivery of femtosecond laser pulses, such as those used so far for graphene ablation, is challenging because fiber chromatic dispersion and optical nonlinearity tend to degrade the pulses. Fiber beam delivery became even more attractive with the development of photonic crystal fibers (PCFs) [8], which allow for, e.g., delivery via an air-core [9] or a large modal area fiber, thus increasing the waveguide damage threshold. In addition, PCFs with exotic cross sectional profiles can present modal distributions that are not possible with conventional fibers.

Here, we present a graphene laser ablation setup that uses fiber delivery of a nanosecond laser beam. The fabricated hole's edges do not present a significant D band in the Raman spectrum, meaning a predominance of zig-zag edges. The setup was designed to produce on graphene an optical image of the fiber's modal profile, with the ablated region closely reproducing this profile. As a proof of principle experiment, 2 cores of a 3-core PCF were simultaneously excited and imaged onto a graphene sample, resulting in the immediate creation of a  $2.4\text{-}\mu\text{m}$ -wide microribbon between the two generated holes.

The ablated graphene samples used in this work were synthesized by the chemical vapor deposition (CVD) (and more specifically high-vacuum CVD) method on copper foil substrate using methane [10]. The samples were then transferred to silica substrates using a wet transfer method with Poly-(methyl methacrylate) (PMMA) as a support film [11]. To ensure the quality of the growth and transfer processes, the silica substrate was optically inspected before and after PMMA removal, and Raman spectroscopy measurements were performed on the transferred graphene. For ablation, a Q-switched Nd:YAG laser was employed which delivered 0.7 ns pulses at 1064 nm with a repetition rate of 1 kHz. The pulses were launched into an optical fiber using an objective lens. At the fiber output a pair of  $10\times$  objective lenses collimated and subsequently imaged the fiber output modal distribution onto the graphene samples. Two types of optical fibers were employed: a standard telecommunications fiber (STF), in which case the fundamental mode (Gaussian-like intensity profile with a  $\sim 15\text{-}\mu\text{m}$  full width  $1/e^2$  modal diameter) was excited; and a 3-core PCF, in which case two cores were simultaneously excited.

Figure 1 shows the results obtained with the STF and an average power of  $\sim 1$  mW. Figure 1(a) shows an optical microscopy image of the sample after ablation. A round superficial hole is clearly observed. Ablation can be further confirmed and better observed via comparison of the confocal mapping of the 2D Raman band before (Fig. 1(b)) and after ablation (Fig. 1(c)). No graphene residue could be detected in the hole's region. The ablated region shape, as well as its dimensions ( $\sim 14$   $\mu\text{m}$  diameter), closely matches that of the fiber mode. The Raman spectra at the ablation edge (see insets) exhibit minimal D-band intensity. To demonstrate ablation with more complex modal structures, a 3-core PCF (Fig. 2(a)) was used. Modes were simultaneously excited in two cores (see inset). The average power into the graphene sample was  $\sim 0.5$  mW. Figure 2(b) shows the 2D Raman band mapping of the ablated region, confirming the presence of two holes. The distance between hole centers is  $\sim 7.4$   $\mu\text{m}$  (c.f. 5- $\mu\text{m}$  hole distance), indicating a small level of defocusing. Nevertheless, a 2.4- $\mu\text{m}$ -wide, 7.5- $\mu\text{m}$ -long microribbon is carved between holes, which can be lengthened by beam scanning and can be attractive for microdevice fabrication. Other hole's shapes can also be obtained with other specialty PCFs.

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## Figures

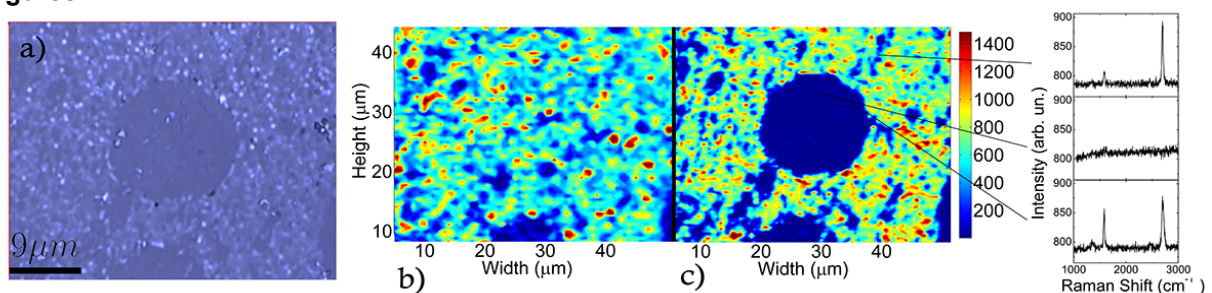


Fig. 1. (a) Optical image of the ablated region. 2D Raman band mapping before (b) and after (c) ablation. Insets show individual Raman spectra taken at the indicated positions.

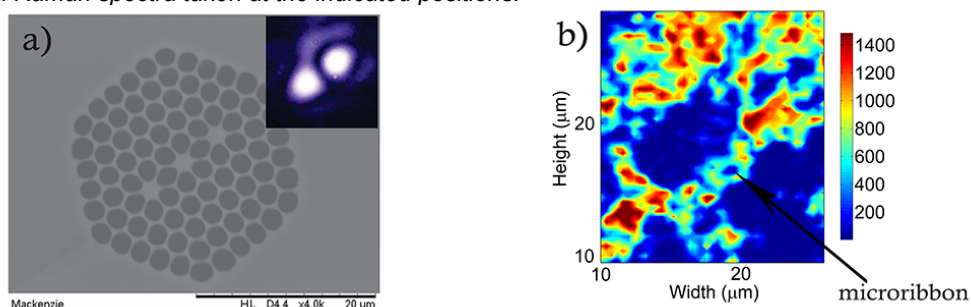


Fig. 2. (a) Scanning electron microscopy image of the PCF cross section (inset: excited modal profile). (b) 2D Raman band mapping showing the two ablated holes and a graphene microribbon in between.