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## Patterning of graphene from ps to fs pulses

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### Abstract

Single-layer graphene is a one-atom thick carbon crystal with unique material properties: extremely high mechanical strength and elasticity, very high electrical and thermal conductivity, uniform absorption over all of the optical and infrared spectrum.

These properties make it highly attractive for many applications, among which flexible electronics. The properties of graphene depend on the number of layers so that patterning with a controlled number of layers affords flexibility in the fabrication of graphene devices; selective ablation leading to the control of the number of graphene layers down to a monolayer has recently been demonstrated using picosecond laser pulses.

In our work we study selective patterning of graphene as a function of pulse length from 340 fs to 14 ps both on glass and on PET polymer substrates. We characterize the patterned graphene using optical transmission and Raman measurements.

Keywords: ultrashort pulses; graphene; ablation; laser

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### 1. Introduction

Graphene is one of the so-called two-dimensional materials (As thin as it gets, 2017). A single layer of graphene is one of the layers of graphite (Geim and Novoselov, 2007); its mechanical, electrical and optical

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properties differ markedly from the properties of bulk graphite and make it a candidate for many applications, among which flexible and transparent electronics (Jang et al., 2016).

The patterning of graphene with ultrashort laser pulses has been demonstrated in several works (for example Sahin et al., 2014, Bobrinetskiy et al., 2015, Van Erps et al., 2015). Selective ablation of multilayer graphene (Lin et al., 2015), in which a determined number of graphene layers can be ablated, is of particular interest for the flexibility that it affords in fabricating graphene-based devices. In the work of Lin et al, 2015, pulses of ps duration were used for ablation, with an intensity, according to the mechanism proposed in the article, at which multiphoton absorption does not play a significant role.

This leaves open the issue of selective ablation with shorter pulses in regimes where multiphoton absorption is one of the mechanisms responsible for ablation.

In our study, we compare selective ablation of multilayer graphene with laser pulses of different duration, starting from 15 ps down to 340 fs.

## 2. Experiments

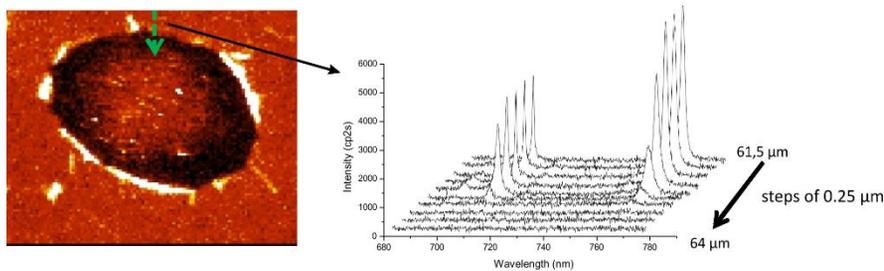


Fig. 1. (Right panel) 2D Raman map of a single-laser shot ablation spot on single layer graphene; the bright areas along the ablation edge may correspond to rolled-up graphene. The relatively bright areas inside the ablation spot do not correspond to the presence of graphene, as is demonstrated by a Raman spectrum (not shown), and are probably due to filter leakage in outside the spectral region of interest. (Left panel) Raman spectra measured along the line marked in green in the right panel. The D line of graphene appears only in the immediate vicinity of the ablation edge.

The graphene samples were purchased from Graphene Supermarket (Calverton, NY, USA) - monolayer graphene on glass and PET – and from Graphenea (San Sebastián, Spain) – monolayer on glass and trilayer on quartz and PET.

We determined the single-shot ablation for successive layers on multiple-layer graphene for three different pulse durations: 340 fs, 3 ps and 14 ps. For the ablation threshold determination, we used the second harmonic output (520 nm wavelength) of a Spectra-Physics Spirit-HE, delivering up to 30  $\mu\text{J}$  of energy at the focus of a 100 mm lens; the pulse duration is tunable via software at the laser output. We placed the graphene sample at a slight defocus (corresponding to a beam radius of about 20  $\mu\text{m}$ ) for a better resolution of the steps corresponding to ablation of different graphene layers.

According to our preliminary results, both the threshold for the ablation of a single layer and the ablation of multiple layers are approximately equal for 340 fs and 3 ps, while they increase by a factor of two for the longer 14 ps pulses.

Additional experiments were performed with a different Spectra-Physics Spirit, under similar focusing condition, with a fixed pulse duration of 380 fs and energy in its second-harmonic output limited to 14  $\mu\text{J}$ .

A Raman analysis of single-shot ablated areas of monolayer graphene on glass (Fig. 1; Graphenea) in this second set of experiments shows that (similar to previous studies, for example Sahin et al., 2014) graphene is

cleanly ablated by an ultrashort pulses, with minimal damage next to the edge. Areas next to the edge where the 2D Raman line is stronger may correspond to rolled-up graphene.

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