Laser induced defects on CVD graphene upon femtosecond pulsed illumination

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Nowadays, there is a frantic activity worldwide to exploit the unique optical properties of graphene and other 2D crystal materials in photonic applications ranging from solar cells and light-emitting devices to touch screens, photodetectors and ultrafast lasers¹. In this line, it has been recently shown experimentally that two-photon absorption (TPA) in graphene is an extremely intense phenomenon². On the other hand, graphene is an ideal memory material because of its transparency, conduction properties and solution processability³. According to this, a novel technique for 3D optical data storage is proposed. Storage capacity as high as 10 TB in a 120-mm-diameter optical disk, namely three orders of magnitude greater than capacities achieved in conventional DVD disks, can be obtained.

The goal of this work is to investigate the creation of defects onto graphene lattice, after illumination by a focused femtosecond laser beam and then to optimize the procedure in order to develop a novel three dimensional optical data storage memory, with high spatial resolution, based on graphene. The memory material will consist of a stacking sequence of a building block, a PMMA host layer with thickness of ~1 μ m in which stacks of mono or multi-layer graphene are attached on opposite sides. The TPA derived luminescence intensity is proportional to the square of the laser intensity while its nonlinear nature provides 3D imaging resolution smaller than 400 nm. Moreover, graphene removal is achieved upon femtosecond laser illumination and it can be used to pattern graphene with very high spatial resolution⁴. The defects induced in the graphene lattice will be used as the recording mechanism in the proposed 3D optical memory. Defected spots correspond to logical '0' whereas non-defected areas correspond to logical '1'.

In a first step, we examine the generation of defects on a single building block consisting of chemically deposited graphene on the top of Si/SiO_2 substrate. The graphene lattice is illuminated using femtosecond pulses with different experimental parameters such as laser power and exposure time in order to create defects which will be both as large as needed for being clearly detected and as small as needed for giving high storage density. Our first target is to investigate whether controlled defects can be created on the aforementioned graphene system. The fabricated samples are characterized by SEM and AFM imaging as well as Raman spectroscopy.

Raman spectroscopy is an integral part of graphene research⁵. Detailed Raman mapping of graphene took place before and after the laser illumination. The Raman spectrum of pristine graphene consists of two distinctive features, known as G and 2D peaks which are located at around 1580 and 2680 cm⁻¹, respectively. The presence of defects gives rise to another two features at around 1350 cm⁻¹ (D peak) and 1615 cm⁻¹ (D peak), which initially are forbidden in non-defected graphene as a result of Raman selection rule (**Figure 1**). The I(D)/I(G) ratio has been widely used to evaluate the amount of disorder in graphene samples. The defected areas exhibit much higher I(D)/I(G) ratio compared to the non-defected ones indicating successful generation of defects^{6,7}. Microscopy and spectroscopy give the same results concerning the size and shape of the defected areas.



Figure 1. Raman spectra of CVD graphene on Si/SiO_2 substrate without defects (top) and with defects (middle & bottom) in the lattice induced after femtosecond laser illumination

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