Microelectronic Engineering 158 (2016) 1–5

Contents lists available at ScienceDirect

Microelectronic Engineering

journal homepage: www.elsevier.com/locate/mee

Patterning of multilayer graphene on glass substrate by using ultraviolet picosecond laser pulses

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ARTICLE INFO

Article history:
Received 21 October 2015
Received in revised form 6 January 2016
Accepted 13 January 2016
Available online 16 January 2016

Keywords:
Ultrafast laser
Picosecond laser
Laser ablation
Multilayer graphene
Thin films
Patterning graphene

ABSTRACT

This paper presents an approach that involves directly patterning multilayer graphene on a glass substrate by using ultraviolet picosecond (PS) laser irradiation. The PS laser is ultrafast, with a pulse duration of 15 ps, and can be operated at a wavelength of 355 nm. In this study, the multiple pulse ablation threshold fluence for patterning multilayer graphene was 5.2 J/cm^2, with a pulse repetition rate of 200 kHz and at a fixed scanning speed of 250 mm/s. The effect of laser parameters on the width, depth, and quality of patterning was explored. To investigate laser-nonablated and laser-ablated multilayer graphene, the characteristics of graphene thin film were measured using Raman, transmittance, and electrical analyses. The experimental results revealed that the PS laser is a promising and competitive tool for ablating multiple layers to several layers of graphene thin films and even for completely removing graphene thin-film layers. The PS laser technique can be useful in developing graphene-based devices. Moreover, this approach has the potential for industrial applications.

1. Introduction

Graphene has attracted considerable attention as a promising material for the highly anticipated all-carbon-based electronic applications because of its favorable electrical, mechanical, and chemical properties [1,2]. From the viewpoint of the small-scale practical production of electronics, considerable efforts have been devoted to developing various graphene-based devices with high carrier mobility (20000 cm^2 V^{-1} s^{-1}) and high carrier density (10^{13} cm^{-2}) for medical sensing applications [3,4] such as the nonenzymatic detection of hydrogen peroxide [5,6]. The surfaces of graphene-based devices are fabricated using multilayer or monolayer graphene with a nonuniform thin film to form the desired thickness and area [7,8]. However, applications of graphene-based materials in certain electronic devices require patterned structures [9,10]. In general, the precise patterned structures for scaling down detection devices depend on the fabrication process.

To obtain the desired graphene structures, laser machining, which is a direct-write, noncontact patterning technique, was used for fabricating micrometer- and nanometer-sized patterns on devices [11]. Compared with the removal of materials by using a conventional long-pulsed laser (>1 ns), using an advanced ultrafast pulsed laser with a nonequilibrium energy transport induces minimal diffusion into any solid material and creates a small heat-affected zone [12,13]. The pulse width of an ultrafast laser ranges from 10^{-12} to 10^{-15} s. The laser ablation threshold can be precisely controlled to only a small area (focal region) of laser intensity. Because of the ultrashort pulse width, an extremely high laser ablation peak intensity may occur within the focal region of material structures with low pulse energy [14,15]. Consequently, ultrafast laser patterning techniques for patterning conductive thin film surfaces can be beneficial in developing integrated electronic applications.

To date, several studies have investigated laser-based approaches for patterning graphene or graphene-related materials that can be considered conductive materials in the development of electrode structure-based devices. Wakaya et al. [16] presented a nanosecond KrF excimer laser that operates at an ultraviolet (UV) wavelength to ablate graphene from a SiO_2/Si substrate, in which the electrical resistance increased at a laser power density of 1.4 MW/cm^2. Kiisk et al. [17] demonstrated intense single pulses of a nanosecond laser at a visible wavelength for ablating monolayer graphene above the threshold energy density of 200 mJ/cm^2. Chen et al. [18] conducted a mask-free programmable patterning of graphene by using femtosecond laser pulses at a wavelength of 800 nm. However, long- and short-pulsed laser beams represent cases in terms of the involved ablated-matter interaction, which reveals the process condition of thermal diffusion at the laser energy into a material. Although several studies have investigated the laser patterning of graphene, studies demonstrating the direct UV ultrafast laser irradiation on multilayer or monolayer graphene are scant. In addition, because

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http://dx.doi.org/10.1016/j.mee.2016.01.012
0167-9317/© 2016 Published by Elsevier B.V.

lasers operating at UV wavelength are small, the process resolution may be affected. Ultrafast laser pulses can be used to fabricate accurate microscale structures.

This study investigated the feasibility of precisely patterning multilayer graphene on a glass substrate by using an ultrashort pulsed picosecond (PS) laser, direct-write technique that has considerable potential for application in industrial processes [19,20]. The ablation threshold for patterning of multilayer graphene through roller imprinting can be demonstrated by using an ultrafast laser operating at a wavelength of 355 nm with a pulse duration of 15 ps. This study provides useful information regarding the applicability of the PS laser technique for ablating multiple layers to several layers of graphene thin films and even for completely removing graphene thin-film layers in developing graphene-based devices for future products.

2. Experimental

Fig. 1a and b shows the experimental laser setup and process, respectively. The surface of multilayer graphene was patterned using an ultrafast laser fabrication system comprising a PS laser (Coherent Inc., model: Ultra 355-04C, USA), galvano-mirror scanner, and high-precision XYZ sample stages. Generally, the graphene-based samples were prepared by high-temperature chemical vapor deposition (CVD) and acid etching treatments [23–25]. In this study, multilayer graphene with graphene ink coated on soda lime glass substrates (5-mm thickness) was formed using roller imprinting, and the homogeneous multilayer thickness was approximately 2.60 ± 0.05 μm. Here, the diameter of stainless–steel roller was 6 mm, and the graphene ink was prepared through cleaned graphite powder (<20 μm) in an aqueous surfactant solution. Note that the roller imprinting can promote adhesion of graphene coating onto the glass substrate by controlling temperature and humidity. On the other hand, scanner control application software was used in this study to control all laser patterning routines. The laser delivers a beam with a 355-nm center wavelength at a maximum power of 4 W and can produce pulses of 15-ps duration at a pulse repetition rate of 200 kHz. The transverse mode of the laser was TEM_{00}, and the beam quality factor (M^2) was less than 1.3. An F-theta lens with a focal length of 150 mm was used in this study. The diameter of the laser beam was 1.3 mm at a $1/e^2$-level of intensity. The diameter of the focal spot was approximately 7 μm on the sample surface. The galvano-mirror scanner was used to steer the laser beam to scan across the substrate surface with a predetermined pattern structure. The scanning speed was adjustable from 5 to 2000 mm/s. The optimal dimensions of the ablated region on the surface of the device with a programmed path were 8 mm × 8 mm, and the pitch of control machining was 20 μm.

3. Results and discussion

3.1. Patterning of multilayer graphene by using laser pulses

In this study, patterned structures of multilayer graphene were formed using continuous Gaussian PS laser irradiation at a UV wavelength. The ablated structures were analyzed using an optical microscope (OM, Olympus Inc., model: BX51) and a three-dimensional laser confocal microscope (LCM, Olympus Inc., model: LEXT OLS4000). The feature size of an area ablated with a Gaussian beam according to energy fluence (laser pulse energy) can be expressed as [21]

$$D = \omega_0 \sqrt{2 \ln \left(\frac{F_0}{F_{th}} \right)} \quad (1)$$

where D is the diameter of the ablated area, ω_0 is the focused beam radius (intensity: $1/e^2$) on the sample, F_0 is the laser peak fluence, and F_{th} is the process threshold fluence. In this study, the PS laser ablation threshold fluence (single-pulse ablation threshold fluence) of multilayer graphene derived according to Eq. (1) was 0.30 J/cm^2. To pattern line structures on multilayer graphene, the accumulated fluence with the multiple pulse effect can be derived as follows [22]

$$F_{th}(N) = F_{th}(1) N^{\zeta - 1} \quad (2)$$

where N is the number of laser pulses and ζ is the accumulation coefficient. The ablation threshold (multiple pulse ablation threshold fluence) for patterning multilayer graphene was 5.2 J/cm^2 at a pulse frequency of 200 kHz and fixed scanning speed of 250 mm/s. Effectively patterning

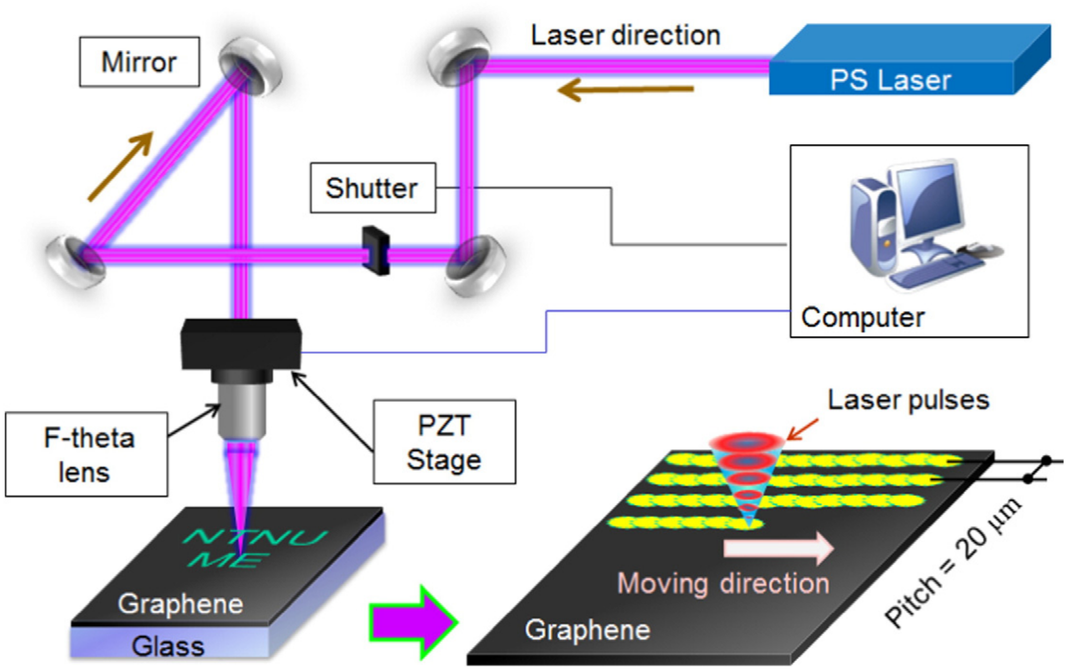

Fig. 1. Experimental setup of a UV PS laser system for mask-free and programmable patterning of multilayer graphene structures, with each patterned structure being scanned using overlapping multiple pulses.