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### Full length article

## Rapid fabrication of transparent conductive films with controllable sheet resistance on glass substrates by laser annealing of diamond-like carbon films

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#### ABSTRACT

We report a laser-based method for directly fabricating large-area, transparent conductive films with customizable electrical resistance on glass. In this method, a diamond-like carbon (DLC) film is deposited first on a glass substrate by pulsed laser deposition, which is then annealed in a helium shielding environment by a 2 kW continuous-wave fiber laser with a wavelength of 1070 nm, which is transparent to glass but is absorbed by DLC to transform the amorphous carbons to graphene. When a 510 nm thick film was annealed at a scanning speed of 1 m/s by a 200  $\mu$ m top-hat laser beam, the sp<sup>3</sup> fraction was decreased from 43.1% to 8.1% after the annealing process, and the transformed film showed a transparency of ~80% (at 550 nm) and a sheet resistance of ~2050  $\Omega$ /sq. We also showed that sheet resistance and transparency can be controlled by changing processing parameters. To show the scalability of the method, a 15 mm wide line beam was used to produce a 15 mm × 15 mm film. This method is simple, fully scalable, transfer-free and catalyst-free, and we believe that the fabricated films can have many applications with further research, such as transparent heating films, electromagnetic shielding films, and transparent electrodes.

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#### 1. Introduction

Graphene is arguably one of the most promising materials for many challenging applications due to its unique and exceptional properties [1–3]. With a rare combination of transparency in the visible range [4] and high electrical conductivity, for example, graphene is considered a next generation material for transparent conductive films and many researchers around the world are working to develop an efficient and cost effective method for producing large-area transparent conductive films based on graphene [5-7]. As of today, the most popular method of fabricating transparent conductive graphene films seems to be chemical vapor deposition. In this method, a graphene film is generally grown on a catalytic metal surface, which needs to be transferred to an actual substrate in a separate procedure [8–10]. For example, Bae et al. fabricated a large-scale, 30 inch graphene film by transferring a CVD grown film on a polyethylene terephthalate (PET) substrate by using a roll-to-roll method [5].

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Recently, laser-based methods have been studied for fabricating graphene films, such as laser exfoliation of highly ordered pyrolytic graphite (HOPG) [11], laser-based epitaxial growth of graphene on silicon carbide [12], laser-induced chemical vapor deposition (LCVD) [13–15], and laser direct writing of graphene [16–19]. In each of these methods, a high-density, high-precision optical energy at a selected wavelength is used as an energy source to thermally transform or evaporate the given carbon source. For instance, Xiong et al. developed a femtosecond laser based nanofabrication process for making graphene patterns on insulating substrates using a co-sputtered Ni/C thin film under ambient conditions [16].

Here, we report a simple and fully-scalable laser-based method for fabricating large-scale, graphene-based transparent conductive films with customizable sheet resistance on glass substrates using amorphous carbon as the carbon source. In this study, a DLC film was first deposited on a glass substrate, which was then annealed by a 2 kW continuous-wave laser in a shielding gas environment to prevent oxidation. The laser power and beam scanning speed was calculated based on the beam size to obtain the optimal process condition. By the annealing process, the sp<sup>3</sup>





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bonds in the film are transformed to sp<sup>2</sup> bonds, and graphene is formed from amorphous carbon. This method is fast and transfer free, and also is fully scalable with an adoption of a higher power laser and appropriate beam shaping optics. Due to its simplicity, high throughput, and cost effectiveness, this is a type of method that is preferred by industries. We believe that this study for the first time demonstrated the possibility of the laser-based method for the fabrication of large-area transparent film heaters and transparent electrodes and it has a potential to become a new class of method in this field.

# 2. High speed fabrication (1 m/s) of line-shaped transparent conductive films using a circular laser beam

Fig. 1a schematically shows the process of fabricating a lineshaped transparent conductive film. In this study, two lasers were used for the whole process: a 6 W picosecond laser for depositing DLC on a glass substrate and a 2 kW fiber laser for the annealing of DLC. First, a DLC film was deposited on a borosilicate glass substrate using pulsed laser deposition (PLD), where a 355 nm wavelength picosecond laser (Coherent Talisker 355-4) was used as an energy



**Fig. 1.** Fabrication of line-shaped transparent conductive films by laser annealing of DLC films with a circular top-hat beam (beam diameter 200 µm, laser power 110 W, scanning speed 1 m/s). (a) Schematic drawing of the overall experimental procedure for fabricating line-shaped transparent conductive films (b) Deposited line-shaped DLC patterns on a borosilicate glass substrate. Film thickness and width are 510 nm and 140 µm, respectively. (c) Optical microscope image of a deposited line-shaped DLC film on glass. (d) Optical microscope image of the top surface of a laser-annealed film (e) Optical microscope image of the time by setting the image focal plane at the bottom of the annealed specimen shown in Fig. 1d. The horizontal blue line located below the specimen shows that the DLC film became transparent (f.g) XPS results of the DLC film (shown in Fig. 1c) for C 1s and O 1s. (h) Raman spectrum of the transformed film measured at the location shown in Fig. 1d. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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