MAY 27, 2020
CONFERENCE ONLINE

Graphene Industrial Forum &2DM 2020

Confocal laser scanning microscopy as a real-time quality-assessment tool for industrial graphene synthesis

Dong Jin Kim[1,2,3,8], Chang-Won Lee[4,8], Yeonjoon Suh[2], Heejeong Jeong[5], Insu Jo[2], Joonhee Moon[6], Mina Park[2], Yun Sung Woo[3,7*], and Byung Hee Hong[2*]

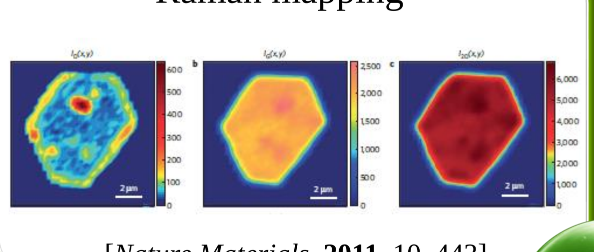
[1]Program in Nano Science and Technology, Graduate School of Convergence Science and Technology, Seoul National University, Seoul 08826, Korea
[2]Graphene Research Center, Advanced Institute of Convergence Technology, Suwon 16229, Korea & Department of Chemistry, Seoul National University, Seoul 08826, Korea.
[3]Graphene Square Inc., Suwon 16229, Korea. [4]School of Basic Sciences, Hanbat National University, Daejeon 34158, Korea [5]Department of Physics, Faculty of Science, University of Malaya, Kuala Lumpur 50603, Malaysia
[6]Division of Analytical Science Research, Korea Basic Science Institute (KBSI), Daejeon 34133, Korea [7]Department of Advanced Materials Application, Korea Polytechnics, Gyeonggi-do 13122, Korea
[8] These authors contributed equally: Dong Jin Kim, Chang-Won Lee

❖ Introduction

For the industrial quality control (QC) of the chemical vapor deposition (CVD) graphene, it is essential to develop a method to screen out unsatisfactory graphene films as efficiently as possible. However, previously proposed methods based on Raman spectroscopy or optical imaging after chemical etching are unable to provide non-invasive and fast analysis of large-area graphene films as grown on Cu foil substrates. Here we report that the reflection mode of confocal laser scanning microscopy (CLSM) provides a high-contrast image of graphene on Cu, enabling the real-time evaluation on the coverage and quality of graphene. The reflectance contrast, R_C, was found to be dependent on the incident laser wavelength, of which the maximum was obtained at 405 nm. In addition, R_C decreases with increasing defect density of graphene. The dependence of R_C on the graphene's quality and laser wavelengths were explained by the tight-binding model calculation based on the Fresnel's interference formula. Thus, we believe that the reflection mode CLSM would be a very powerful quality-assessment tool for the mass production of CVD graphene films grown on Cu.

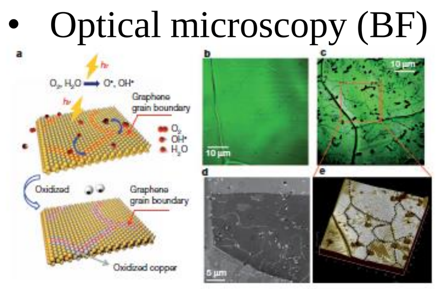
❖ Motivation

- Raman mapping



[*Nature Materials*, **2011**, 10, 443]

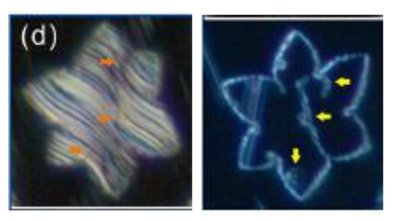
- Optical microscopy (BF)



[*Nature*, **2012**, 490, 235]

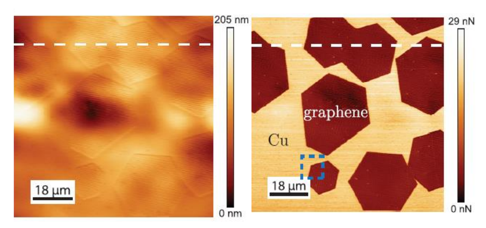
Graphene Imaging

- Optical microscopy (DF)



[*Appl. Phys. Lett.*, **2013**, 103, 043119]

- AFM (Friction)



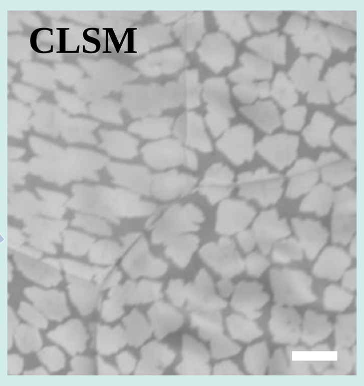
[*ACS nano*, **2014**, 8, 5010]

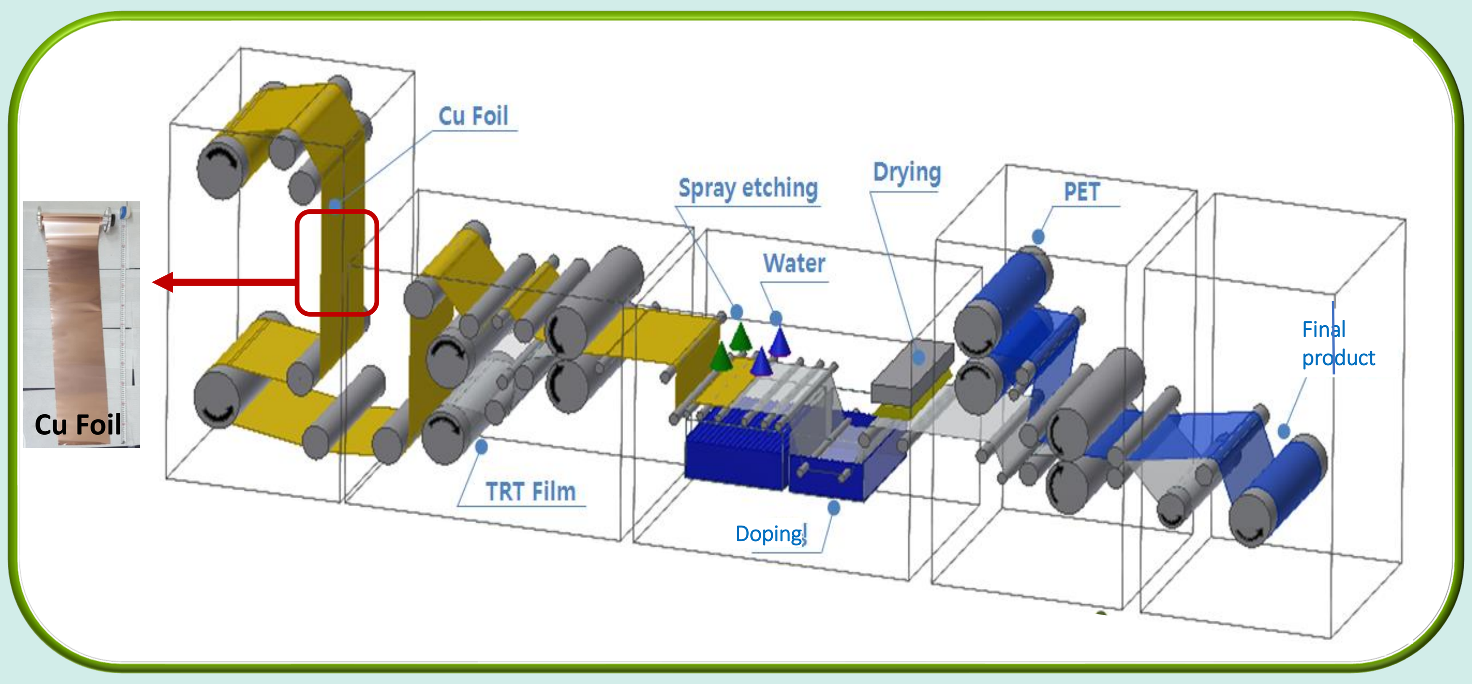
Cons
- ✓ Tim-consuming
- ✓ Destructive
- ✓ Non-exclusive
- ✓ Non-applicable in large area

Pros
- ✓ Non-destructive
- ✓ Exclusive
- ✓ Rapid & large area scanning

- R2R system & in-situ monitoring for mass production of CVD graphene (Graphene square Inc.)

In situ monitoring for QC (Quality control)





❖ Results & Discussion

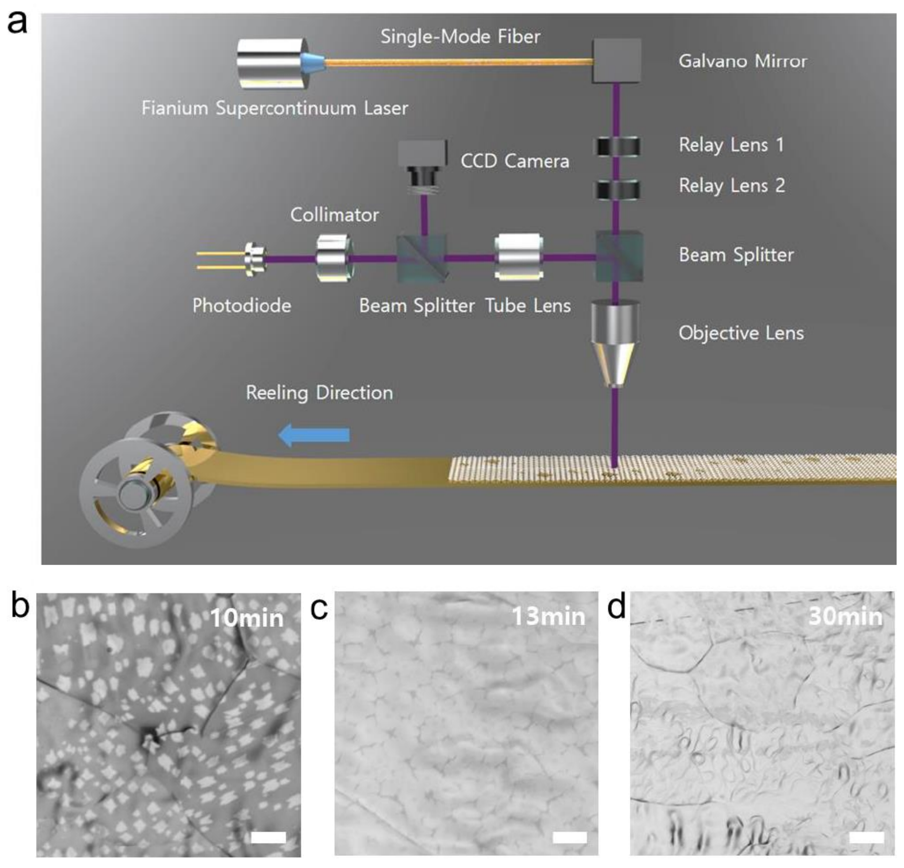


Fig. 1 | Concept of *in-situ* monitoring of the as-grown CVD graphene on Cu using CLSM. (a) The concept of monitoring the as-grown CVD graphene on Cu foil during the continuous roll-to-roll synthetic process using the reflective mode CLSM. (b)-(d) The CLSM images of the CVD graphene with various coverage from sub-monolayer to fully covered on Cu foil controlled by growth time of 10, 13, and 30 min, respectively. Scale bar: 10μm

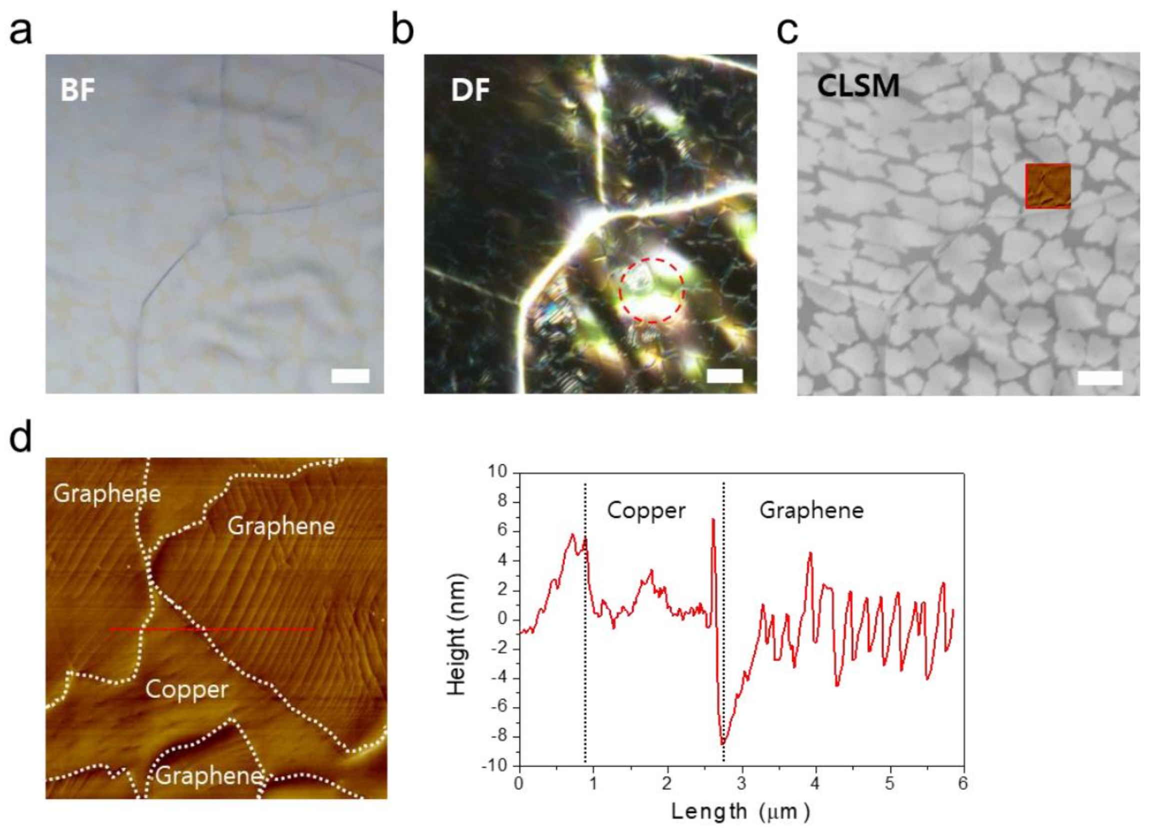


Fig. 2 | Comparison of BF, DF and CLSM images of CVD graphene on Cu. The images of CVD-grown graphene on Cu foil with coverage of sub-monolayer in the same region obtained by (a) bright field microscope, (b) dark filed microscope, and (c) CLSM. The morphology of graphene domains on Cu foil was measured by AFM, as shown in (d). The height profile in the right, which is taken at a red dotted line in (d). Scale bar: 10μm.

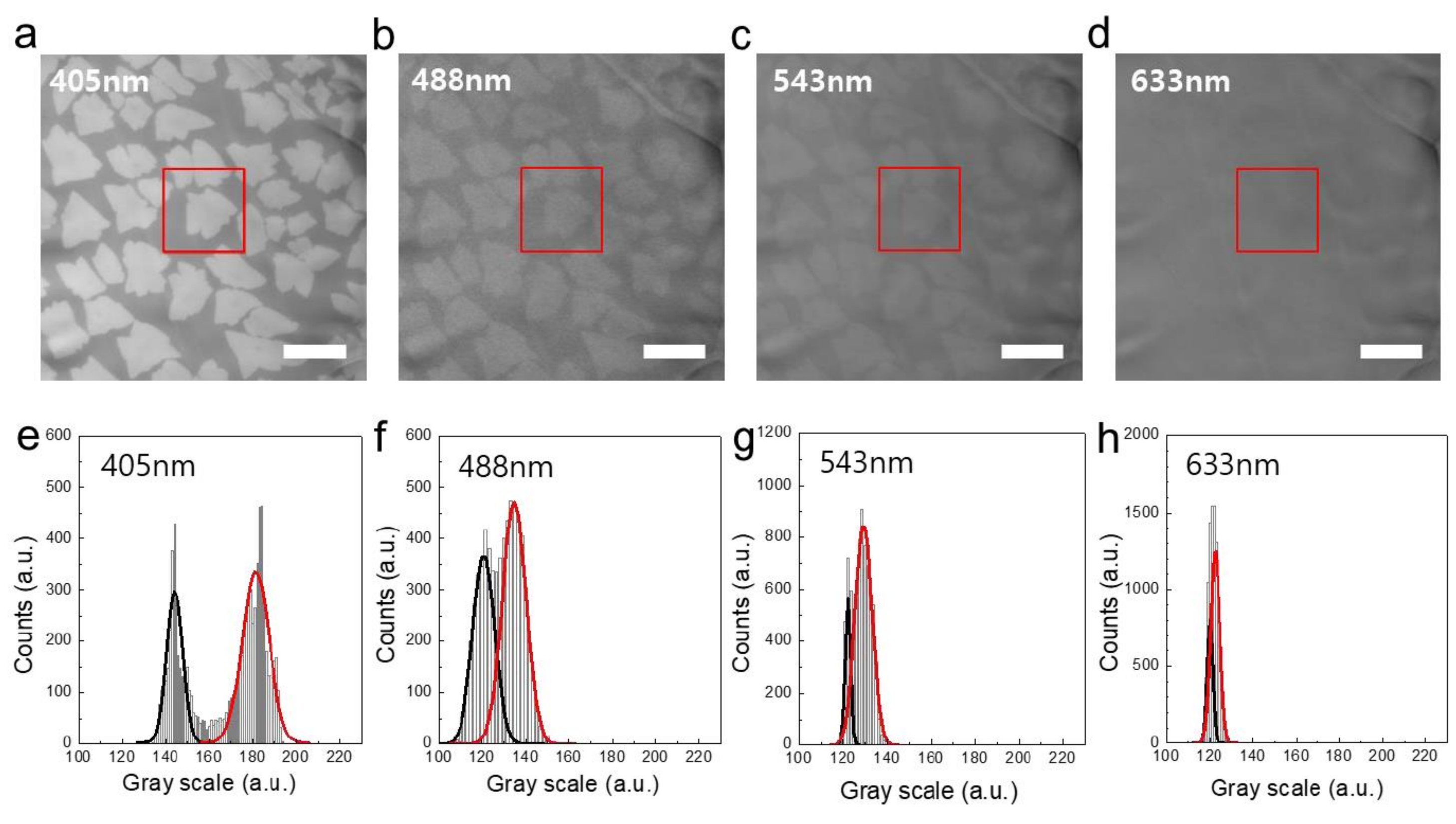


Fig. 3 | Dependence of CLSM images on the incident laser wavelength. The CLSM images of the CVD-grown graphene on Cu foil obtained under four different laser wavelengths of (a) 405, (b) 488, (c) 504, and (d) 633 nm, respectively. The grayscale histograms in (e)-(f) were obtained in the area indicated by the red box in each CLSM image above. The black and red Gaussian curves in the grayscale histogram correspond to dark Cu and bright graphene, respectively.

Fresnel's interference formula

$$Rc(\omega) = \left|\frac{n_{cu}(\omega)+\frac{\sigma_{GP}(\omega)}{c\varepsilon_0}-1}{n_{cu}(\omega)+\frac{\sigma_{GP}(\omega)}{c\varepsilon_0}+1}\right|^2 \Big/ \left|\frac{n_{cu}(\omega)-1}{n_{cu}(\omega)+1}\right|^2$$

$$\sigma = \sigma_{\text{intraband}} + \sigma_{\text{interband}}$$

$$\sigma_{\text{intraband}} = i\frac{e^2\mu}{\pi\hbar^2(\omega+i\gamma)}$$

$$\sigma_{\text{interband}} = \frac{e^2}{4\hbar}\{\theta(\hbar\omega-|2\mu|)\} + i\frac{1}{\pi}\log\left|\frac{\hbar\omega+|2\mu|}{\hbar\omega-|2\mu|}\right|,$$

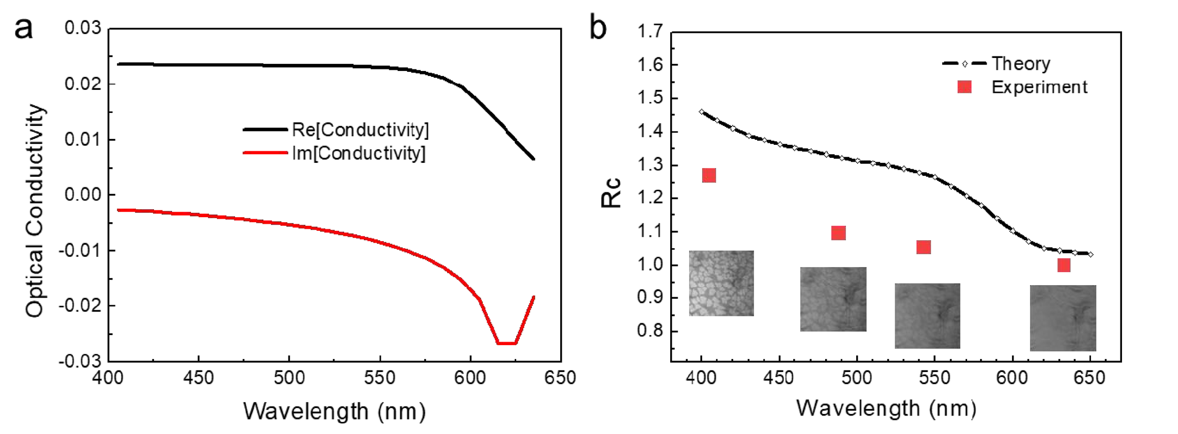


Fig. 4 | Measured and calculated reflectance contrast with optical conductivity. (a) The real and imaginary parts of the optical conductivity of graphene were drawn according to Eqn. (3) and (4) as a function of wavelength. (b) The calculated and measured *Rc* of graphene on Cu were compared at four laser wavelengths of 405, 488, 543, and 633 nm, respectively.

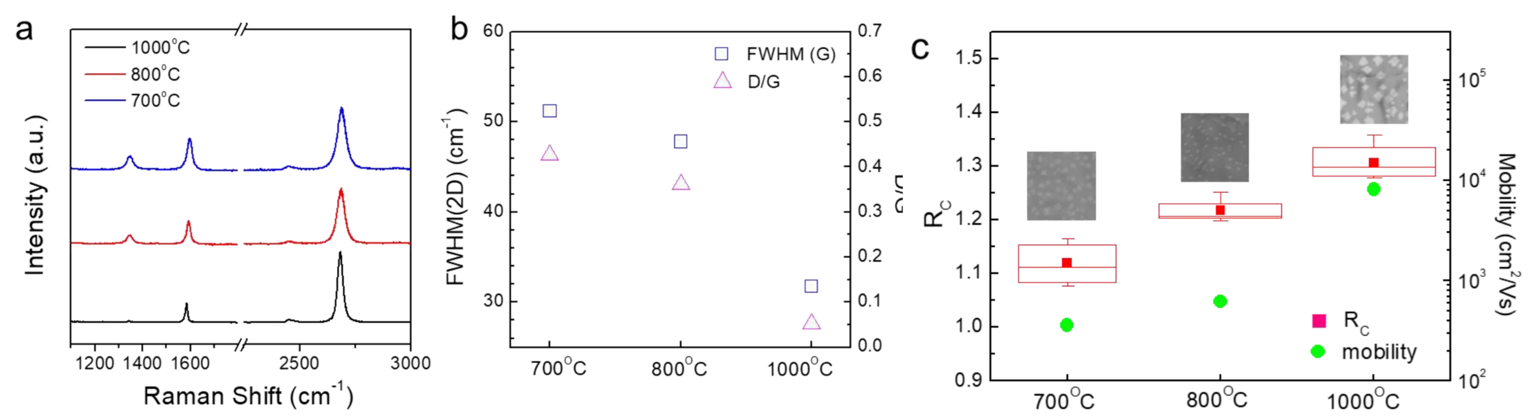


Fig. 5 | Raman analysis and reflectance contrast of CVD graphene grown at different temperature. (a) The Raman spectrum of CVD graphene grown on Cu foil at different temperature of 700, 800, and 1000 °C, respectively. (b) The FWHM of 2D peak and the intensity ratio of D/G peak obtained from the Raman spectrum of (a) are compared with the growth temperature. (c) The carrier mobility and the statistical analysis of *RC* of CVD graphene are compared with the growth temperature.

CONTACT PERSON

Dong Jin Kim
Seoul National University, Korea. dj.kim@snu.ac.kr

Prof. Yun Sung Woo
Dankook University, Korea. yunswoo@dankook.ac.kr

Prof. Byung Hee Hong
Seoul National university, Korea. byunghee@snu.ac.kr

REFERENCES

[1] Qing Yu *et. al.*, *Nature Materials* (**2011**), 10, 443
[2] Dinh Loc Duong *et. al.*, *Nature* **(2012)**, 490, 235
[3] X. H. Kong *et. al.*, *Appl. Phys. Lett.* (**2013**), 103, 043119
[4] Philip Egberts *et. al.*, *ACS nano* **(2014)**, 8, 5010

Graphene Industrial Forum &2DM 2020