

## RISE - Raman SEM Imaging of Single Layer and Twisted Bilayer Graphene

Ute Schmidt<sup>1</sup>, Hans Zimmermann<sup>2</sup>, Stefanie Freitag<sup>2</sup>, and Thomas Dieing<sup>1</sup>

<sup>1</sup> WITec GmbH, Ulm, Germany (www.witec.de)

<sup>2</sup> Carl Zeiss Microscopy GmbH, Munich, Germany (www.zeiss.com)

Due to its unique properties as one dimensional material, graphene gained a lot of attention not only within fundamental research [1,2] but more and more from applied research for its integration in electronic circuits and sensors. For these applications the quality of the graphene is of major interest in terms of defects such as wrinkles, folding or lattice mismatches. Furthermore the phononic and electronic properties of double layer graphene are strongly influenced by the stacking disorder. A key requirement for graphene research is the ability to identify and characterize all present carbon allotropes, both on lab scale as well as on mass production scale. The characterization tool of choice should for this purpose be non-destructive, fast, with high resolution and provide the maximum structural and electronic information.

Scanning Electron Microscopy (SEM) is known for its ability to characterize surfaces from the millimeter range down to highest resolution, allowing overview images of the sample and zoom in to the nanometer scales on the regions of interest. This technique enables highest resolution imaging of the graphene samples, revealing their defects on the nanometer scale. Raman spectroscopy and more precise confocal Raman imaging of graphene reveals structural and electronic information with diffraction limited resolution. A combination of these two techniques in a single instrument fulfills all the above mentioned requirements for graphene characterization.

Fig. 1a shows the SEM image of a twisted bilayer graphene flake obtained by chemical vapor deposition on a 300 nm thick Si/SiO<sub>2</sub> substrate. The gray shades in the SEM image correspond mainly to an increase of the number of graphene layers. The RISE image (Fig. 1b) shows that the entire surface is covered by graphene, as well as that there are zones of different layers as revealed by the measured G and 2D Raman bands for graphene. By analyzing the intensity and full width at half maximum (FWHM) of these two characteristic Raman bands, it is possible to determine the stacking order and twist angle of the analyzed graphene grain [3-5]. Fig. 2 shows the variations in peak intensities of I<sub>G</sub>/I<sub>2D</sub> and changes of the FWHM of the 2D Raman band together with the associated variation of the twist angle and stacking order.

The aim of the contribution is to present the complementary information that graphene researcher can achieve by combining Raman and SEM imaging in one integrated system

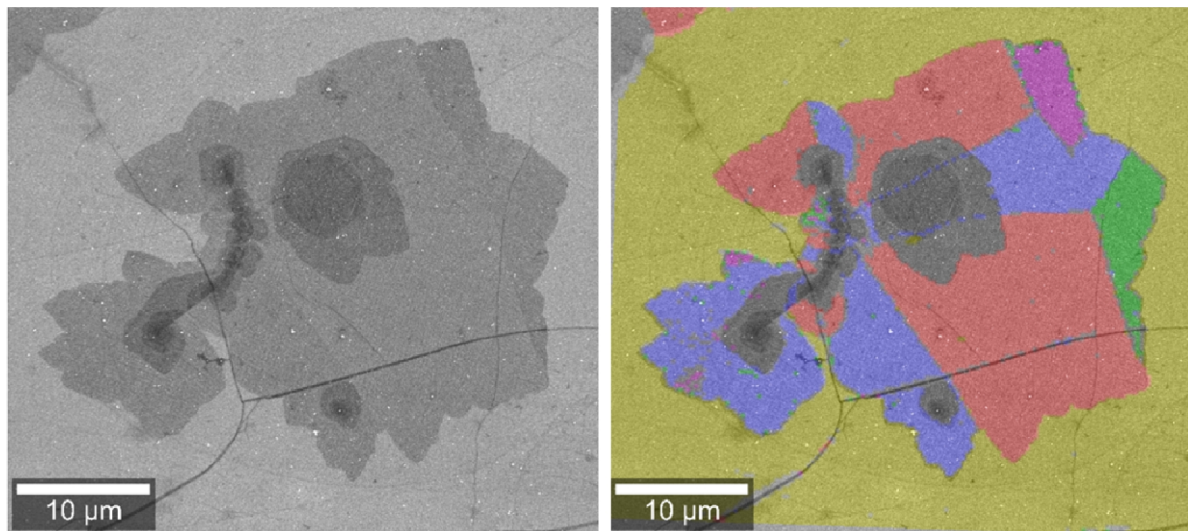
[1] L.M. Malard, M.A. Pimenta, G. Dresselhaus and M.S. Dresselhaus, *Physics Reports* **473** (2009), p. 51.

[2] A.C. Ferrari, *Solid State Communication* **143** (2009), p. 47.

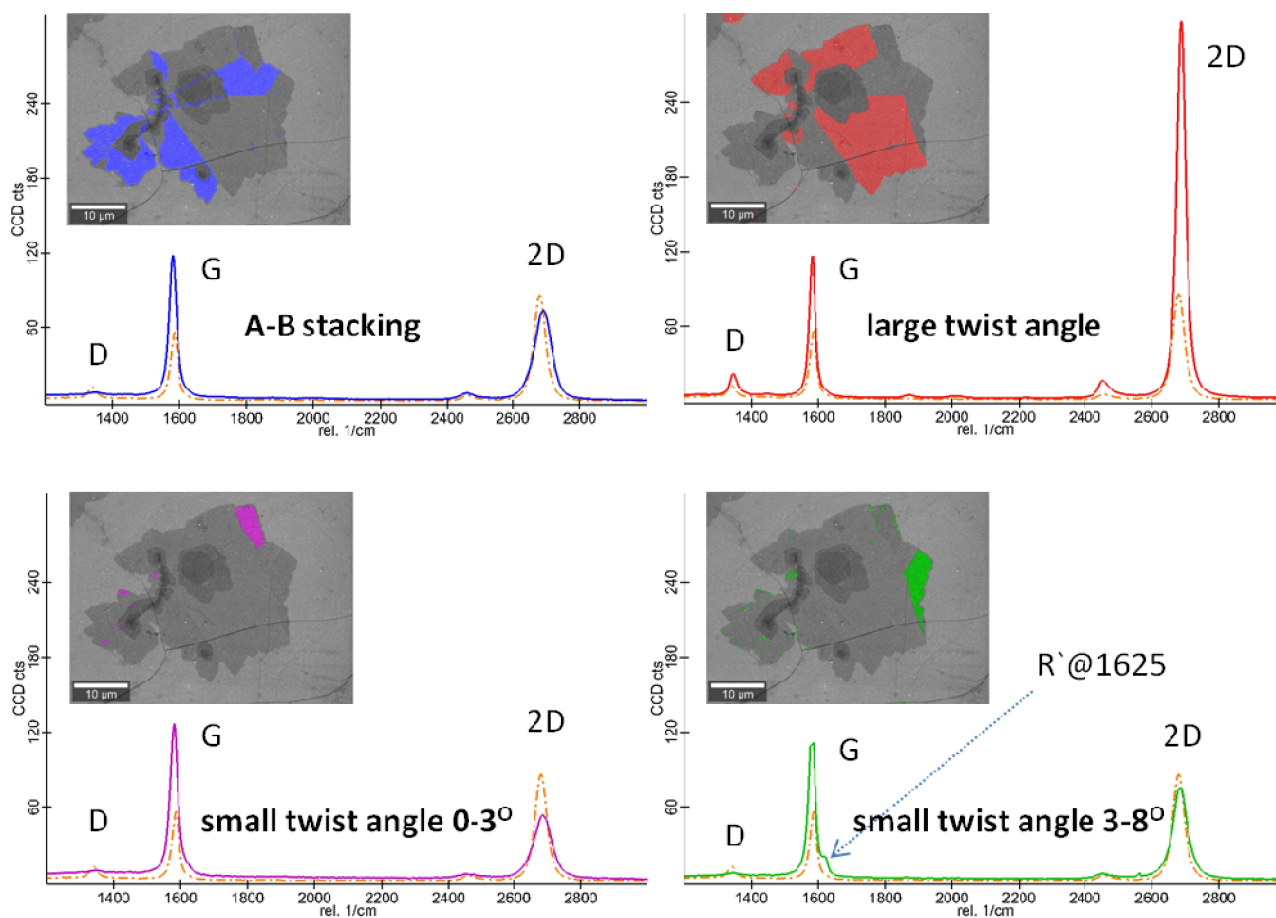
[3] Y. Chen, L. Meng, W. Zhao, Z. Liang, X. Wu, H. Nan, Z. Wu, S. Huang, L. Sun, J. Wang, and Z. Ni, *Phys. Chem. Chem. Phys.* **16** (214), 21682.

[4] T. Ohta, T. Becheem, J.T. Robinson, G.L. Kellogg, *Phys. Rev B* **85** (2012) 75415.

[5] S. D. Costa, J. E. Weis, O. Frank, M. Kalbac, *Carbon* **98** (2016), 592.



**Figure 1.** SEM and RISE image of a grain of twisted bilayer of graphene.



**Figure 2.** RISE images and Raman spectra highlighting areas with different stacking order and twist angle. As reference the Raman spectrum of the monolayer of graphene is shown in dashed line in each spectrum.