## **Supporting information**

# Noninvasive Scanning Raman Spectroscopy and Tomography for Graphene

### Membrane Characterization

Stefan Wagner<sup>1</sup>, Thomas Dieing<sup>2</sup>, Alba Centeno<sup>3</sup>, Amaia Zurutuza<sup>3</sup>, Anderson D. Smith<sup>4</sup>, Mikael Östling<sup>4</sup>, Satender Kataria<sup>1</sup>, Max C. Lemme<sup>1</sup>

 <sup>1</sup> Department of Electrical Engineering and Computer Science, University of Siegen, Hölderlinstrasse 3, 57076 Siegen, Germany
<sup>2</sup>WITec Wissenschaftliche Instrumente und Technologie GmbH, Lise-Meitner-Str. 6, 89081 Ulm, Germany

<sup>3</sup>Graphenea S.A., Avenida de Tolosa, 76, 20018 - San Sebastián, Spain

<sup>4</sup> School of Information and Communication Technology, KTH Royal Institute of Technology, E229, 16440 Kista, Sweden

### S1: Application example for Raman tomography

A second chip with cavities was fabricated using a polytetrafluoroethylene (PTFE) layer to cover the cavities side wall after the etching process. A Raman tomography image was recorded of these cavities which can be seen in Fig. S1 a, b. In the image stack (Fig. S1a) as well as in the 3Dimage (Fig. S1b) the PTFE layer can be identified (in purple color) not only at the top of the cavity like in Fig. 4 d, e (in the manuscript), but also along the sidewalls and at the edge where sidewall and the bottom of the cavity meet. It demonstrates the ability of Raman tomography to analyze the material and chemical composition inside the device, e.g., under a graphene layer.



Figure S1: A data set for a Raman tomography of a graphene covered cavity. a) Sequence images of cavity 4 for the 3D scan taken at different z-positions; b) 3D tomography image of a cavity. Here a polymer ring could be identified not only on the top of the cavity, but along the sidewalls and at the bottom of the cavity. Here, yellow is FSG, turquoise SSG, red is the polymer and blue the Si.

#### S2: Description of K-means cluster analysis

The K-means cluster analysis is often assigned to the partial clustering methods. It differs from the hierarchical cluster analysis in that the number of clusters must be selected prior to the clustering. However, the method can also then be applied to the sub-clusters in order to sort them further into sub-clusters and thus generate a pseudo-hierarchical cluster tree (see for example Dieing et al.1). Once the number of clusters (N) is defined for the K-Means cluster analysis, the algorithm first defines N centers in the M-dimensional space, where M is the number of pixels on the CCD camera, and assigns each point (spectrum) the center closest to it. Then the centroid (one might also call it an average spectrum) for each group is computed. Following this the spectra are again sorted according to their distance to the calculated centroids and then the procedure is repeated. The algorithm is typically stopped once the assignment of the points (spectra) to their group ceases to change. While this method needs somewhat more supervision than hierarchical clustering and is heavily dependent on the selection of the N initial centers, it requires much less processing power and can easily be run on standard PCs or Laptops.

### **References:**

(1) Dieing, T.; Hollricher, O.; Toporski, J. *Confocal Raman Microscopy*; Springer Science & Business Media, 2011.