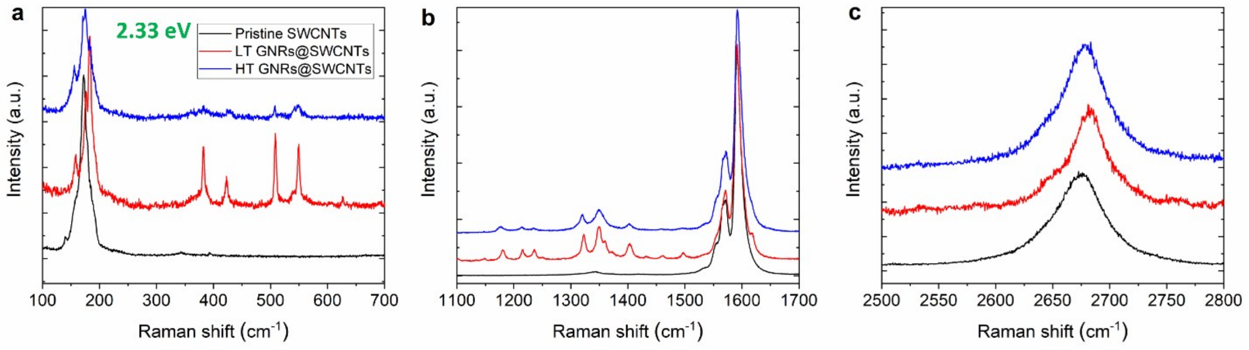
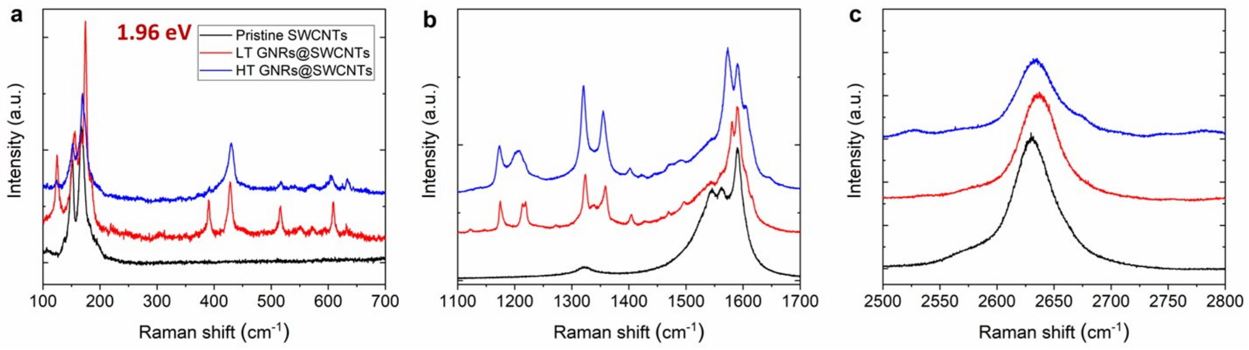
Electronic Supplementary Material (ESI) for Nanoscale.

# •Supporting Information Description



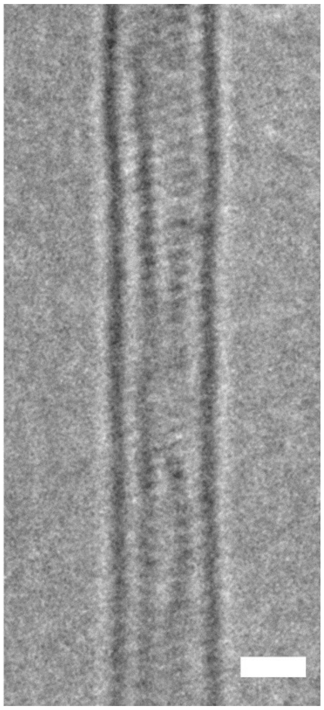
S1 Raman spectra of the pristine SWCNTs (black), low-temperature-grown GNRs inside SWCNTs (red) and high-temperature-grown GNRs inside SWCNT (blue). Excitation wavelength 2.33 eV.



S2 Raman spectra of the pristine SWCNTs (black), low-temperature-grown GNRs inside SWCNTs (red) and high-temperature-grown GNRs inside SWCNT (blue). Excitation wavelength 1.96 eV.

Raman measurements were performed with two excitation wavelengths on LabRAM HR Evolution spectrometer. Samples were in the solid form. One can note the differences between pristine nanotubes and two types of GNR@SWCNTs samples, although the signals of SWCNTs and GNRs overlap. For measurements performed at 2.33 eV excitation (Fig. S1), the main difference between the pristine and filled samples is in the appearance of new components, in example in the ranges 350-600 cm$^{-1}$ and 1150-1500 cm$^{-1}$. For HT GNRs@SWCNTs we detect the decrease of intensity of those components. One additional feature is the shift of the main RBM component of the nanotubes, positioned at 183 cm$^{-1}$ for LT GNR@SWCNT sample. The shift is around 10 cm$^{-1}$ compared to the high temperature sample and also to pristine nanotubes. According to this observation one may assume that for low temperature samples the filling causes the extra change in the vibrational properties of the nanotubes, which is relaxed after the higher temperature treatment. That is in accordance with the PL data obtained in the work. The G band (Fig. S1b) doesn't demonstrate the change in the position after the formation of GNRs.

As previously was reported [1] entirely different spectrum is observed for HT@SWCNTs at 1.58 eV. In our case we use 1.96 eV excitation and also are able to detect changes between low temperature polymerized (LT) and high temperature (HT) samples. Additional peaks corresponding to HT GNRs around the G band of the nanotubes become more prominent, namely 1573 and 1604 cm$^{-1}$ (Fig. S2b), together with the component placed around 1300 cm$^{-1}$. More importantly only one peak is detected at 430 cm$^{-1}$, that is placed in the region for the RBLM of GNRs. Similar to what is reported for atomically precise GNRs [2]. The possibility of the DWCNT formation is ruled out by two reasons: (i) the used temperatures for polymerization are much less than reported for the growth of DWCNTs; (ii) for SWCNTs with diameter distribution 1.4-1.6 nm as used in the experiment, the formed inner tubes should demonstrate the Raman response around 300 cm$^{-1}$.



S3 High temperature grown GNR inside SWCNT, scale bar – 1 nm.

The TEM image was taken by a JEM 2010F electron microscope with a CEOS spherical aberration corrector operated at 120 keV.

[1] H. E. Lim, Y. Miyata, M. Fujihara, S. Okada, Z. Liu, Arifin, K. Sato, H. Omachi, R. Kitaura, S. Irle, K. Suenaga and H. Shinohara, *ACS Nano*, 2015, **9**, 5034–5040.

[2] B. V. Senkovskiy, M. Pfeiffer, S. K. Alavi, A. Bliesener, J. Zhu, S. Michel, A. V. Fedorov, R. German, D. Hertel, D. Haberer, L. Petaccia, F. R. Fischer, K. Meerholz, P. H. M. van Loosdrecht, K. Lindfors, and A. Gruneis, *Nano Lett.*, 2017, **17**, 4029-4037.