Supporting information for:

Electrical annealing and temperature dependent transversal conduction in multilayer reduced graphene oxide films for solid-state molecular devices

Jonas Rahlf Hauptmann,^{a†} Tao Li,^{b†} Søren Petersen,^b Jesper Nygård,^a Per Hedegård,^a Thomas Bjørnholm,^b Bo W. Laursen^{*b} and Kasper Nørgaard,^{*b}



Figure S1: AFM image of GO flakes deposited on a SiO₂ wafer.



Figure S2: Electrical annealing of junction 5. Measurements of the current as a function of bias voltage at 14 K. In these measurements the voltage is swept initially at low voltages between $(\pm 0.5V)$ and subsequently increased in steps of 0.1 or 0.2 V until the sweep voltage is $\pm 7V$. At each voltage the sweep is repeated typically 10 to 20 times. a) Shows 10 IV-curves swept between $\pm 3.3V$ just below the threshold for current increase. b) 50 voltage sweeps between $\pm 3.5 V$. An increase in current is observed and increases with the number of voltage sweeps. c) The first and last sweep in each series of sweeps at increasing voltages.



Figure S3: IV curves measured before and after electrical annealing of the rGO film. a) IV-curve on five different junctions measured at 10K before electrical annealing. The inset is an enlarged view of the least conducting junction. b) IV-curves of the same five junctions as in a) after electrical annealing of the rGO films (measured at 14K - note the very different scale on the y-axis). The inset is an enlarged view of the least conducting junction.



Figure S4. Resistance dependence as a function of temperature measured for the electrically annealed junctions. a) Resistance as a function of temperature for the five junctions on a logarithmic scale, fitted with the equation $G(T) = G_1 \exp(-B/T^{1/4}) + G_t$ describing 3D VRH plus an extra term describing temperature independent transport [1]. b) Resistance as a function of temperature for the three most conducting junctions plotted on a linear scale the lines are linear fit to the scatter points. The inset shows the four most conducting junctions on linear scale.

It is observed that the data points, for the four most conducting junctions, can be fitted linearly after the electrical annealing. Prior to annealing, an exponential increase in the resistance as a function of temperature was observed, indicating an isolating behavior at low temperatures, which, after electrical annealing, have changed to a linear behavior. After annealing, the samples show no indication of insulating behavior at temperatures down to 10 K. The actual conduction mechanism is best described by a semiconducting behavior since it is neither isolating nor metallic.

References [1] A. B. Kaiser, C. Gómez-Navarro, R. S. Sundaram, M. Burghard, K. Kern, *Nano Lett.* 2009, **9**, 1787.