Supplementary Information

Antibacterial activity of large-area monolayer graphene film manipulated by charge transfer

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1. Experimental details

Graphene transfer

Graphene films were transferred from the Cu and Ge substrates onto TEM grids by a PMMA-assisted wet-transfer method. In detail, a thin layer of polymethyl methacrylate (MicroChem 950 PMMA C, 3 % in chlorobenzene) was spin-coated onto the substrate to protect the graphene film and to act as a support which was then cured at 180 ºC for 10 min. Afterwards, the substrate was etched away by a mixture of HNO_3 and HF (1:1, aq) for Ge or by 1 mol/L $(NH_4)_2S_2O_8$ (aq) for Cu, allowing the PMMA/carbonic layer to float on top of the solution. After placing the layer on a filter paper, it was washed with deionized water. The PMMA/graphene layer was subsequently transferred to TEM grids, followed by annealing at 50 ºC for 90 min to improve adhesion. The PMMA was then dissolved gradually with acetone and deionized water. Finally, the graphene sample was washed with isopropanol. (After the removal of the PMMA film in acetone, the graphene films can be transferred onto any substrate for analysis and characterization subsequently.)

2. Supplementary figures

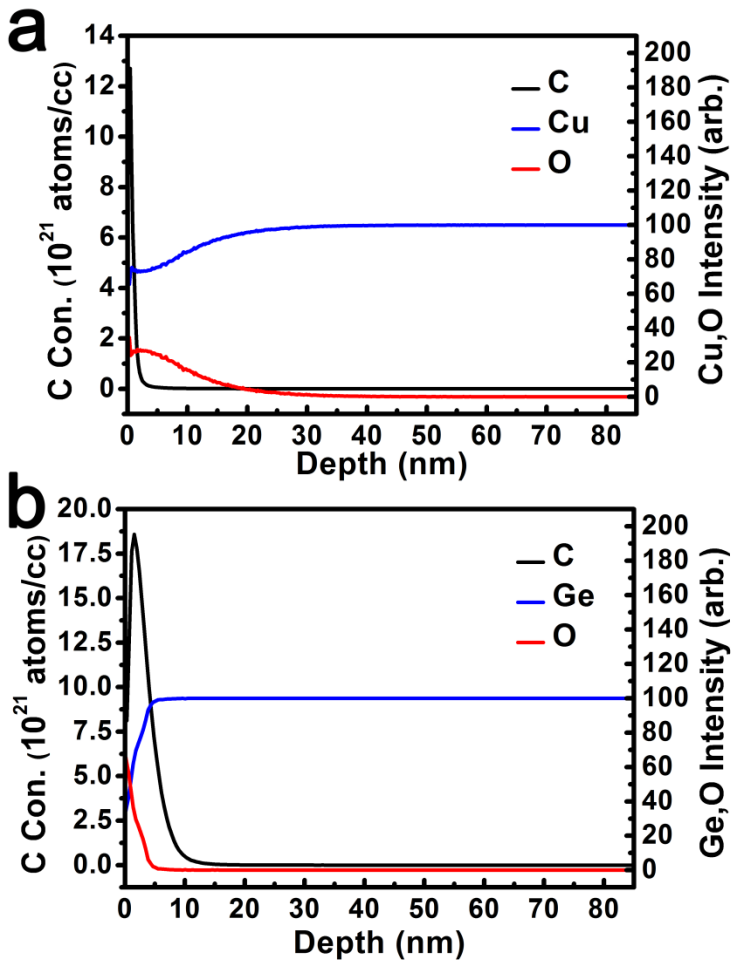

Figure S1. SIMS depth profiles showing the carbon distribution in Cu (**a**) and Ge (**b**) after the growth of graphene films.

The SIMS depth profile in **Figure S1a** indicates the limited carbon dissolution and diffusion in the Cu substrate after the growth of graphene film, which is similar to that of the Ge substrate in **Figure S1b.** Hence, the Cu and Ge substrates show a huge similarity. And the obtained results agree with the XPS analysis.

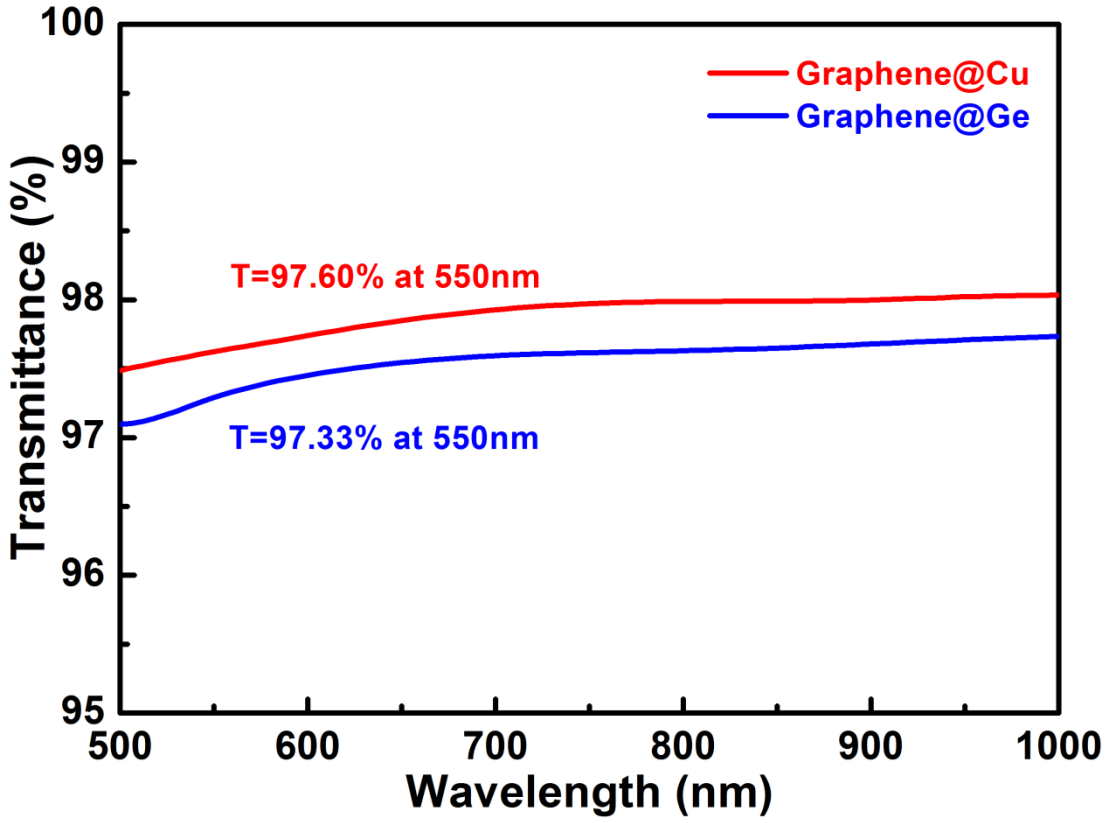

Figure S2. Transmittance of monolayer graphene film measured by visible-absorption spectroscopy on quartz substrate at 550 nm. The testing graphene films are transferred from those grown on Cu and Ge substrates.

As shown in **Figure S2**, the transmittance at 550 nm was measured from the graphene film transferred onto glass slide. From this figure, one can see that a high transparency of ~ 97 % was obtained. Considering an absorbance of ~2.3 % for an individual graphene layer[1], the graphene film can be inferred to have only one layer.

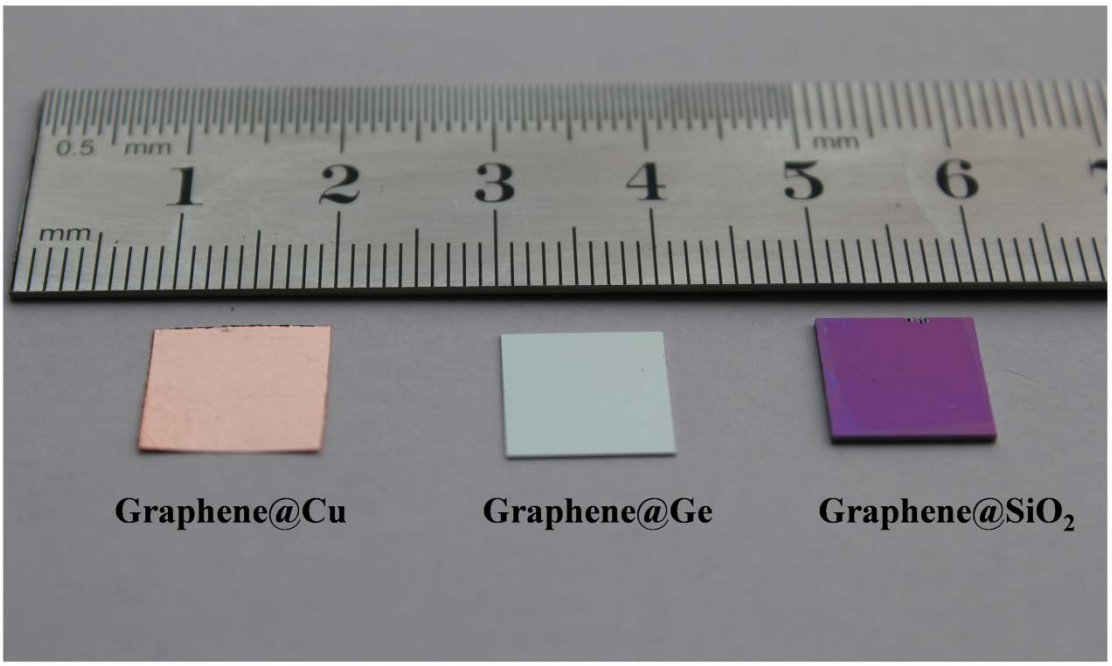

Figure S3. Photographs of the large-area graphene films on conductor Cu (Graphene@Cu), semiconductor Ge (Graphene@Ge) and insulator SiO_2 (Graphene@SiO_2) substrates, respectively.

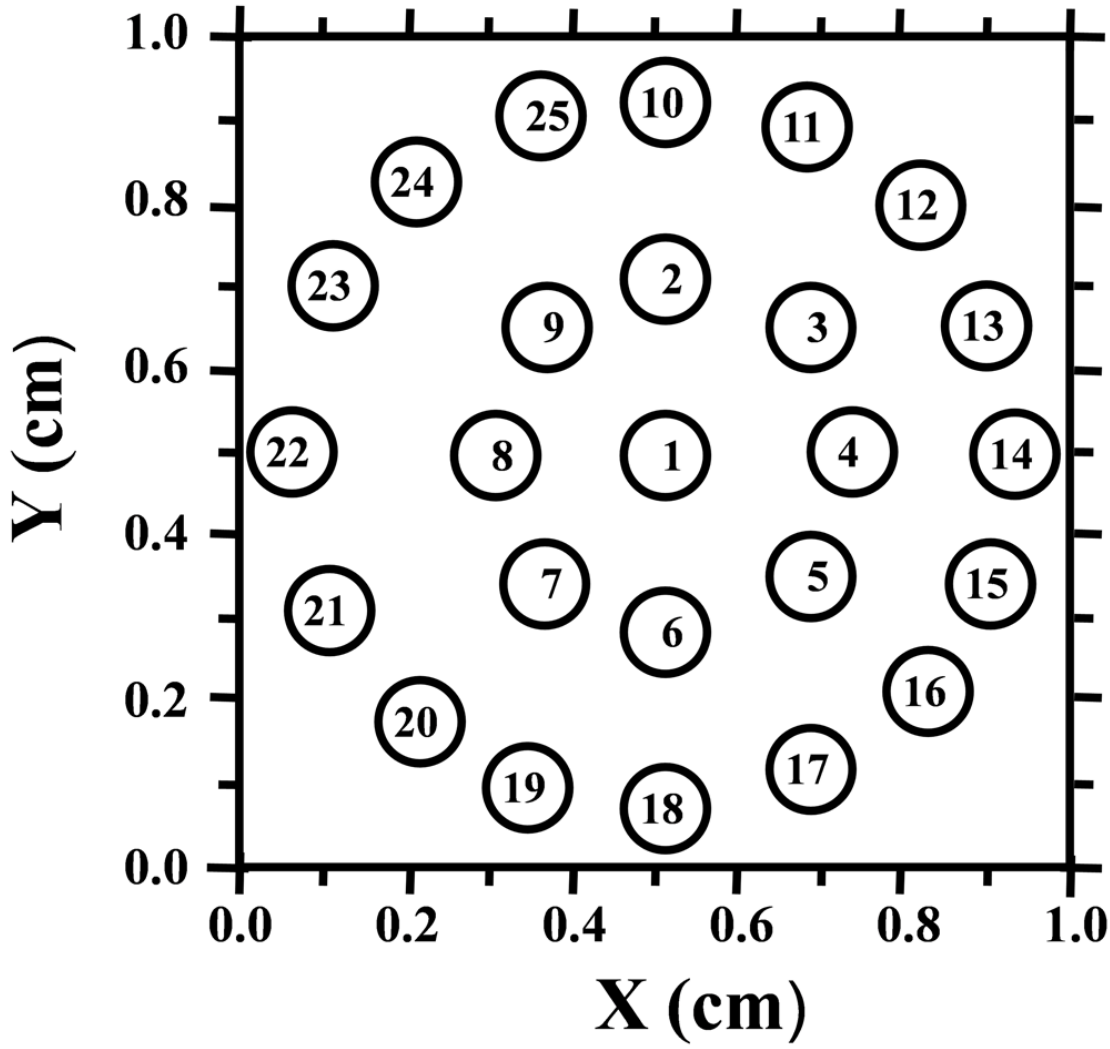

Figure S4. Schematic illustration for the Raman mapping measurements of graphene films on conductor Cu, semiconductor Ge and insulator SiO_2 substrates.

To estimate the uniformity and the coverage of the graphene films on Cu, Ge and SiO_2 substrates, 25 points uniformly distributed over the area of 1 × 1 cm^2 have been analyzed by Raman scattering, as illustrated in **Figure S4**. In the experiment of antibacterial ability evaluation, before seeding the bacteria onto the graphene films on substrates, all the testing samples were selected by the Raman mapping method in order to make sure the uniformity and coverage of graphene films on substrates with an area of 1 × 1 cm^2 used in the test.

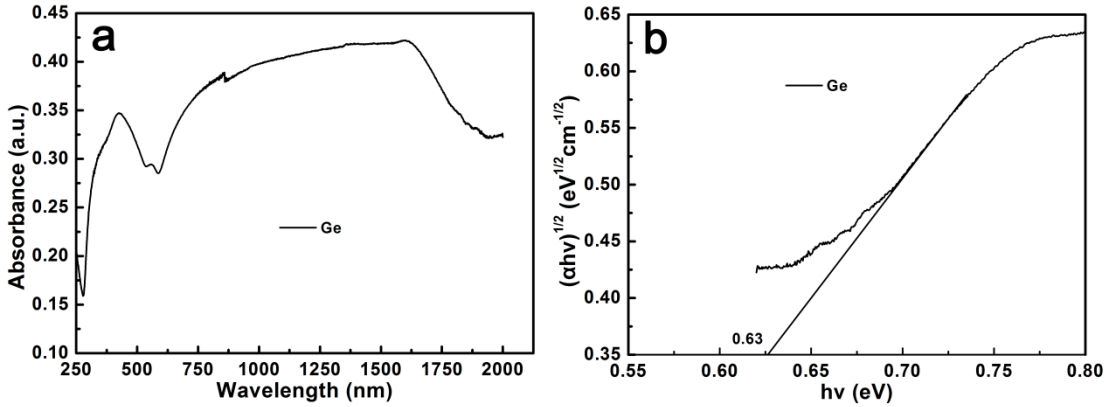

Figure S5. UV-Vis diffuse reflectance spectrum of the pristine Ge substrate, accompanied by the corresponding Kubelka–Munk transformed reflectance spectrum.

Figure S5a shows the UV-Vis diffuse reflectance spectrum of the pristine Ge substrate. The Kubelka–Munk function is utilized to convert diffuse reflectance measurement into the equivalent absorption coefficient[2].

$$\alpha = \frac{(1-R)^2}{2R} \tag{1}$$

Where α is the optical absorption coefficient near the absorption edge for indirect interband transition, R is the reflectance of semiconductor, $R = 10^{-A}$, and A is the optical absorbance.

$$\alpha h\nu = C_1(h\nu - E_g)^2 \tag{2}$$

$$h\nu = \frac{1240}{\lambda} \tag{3}$$

Where C_1 is the absorption constant for indirect transition, $h\nu$ is the photon energy, E_g is the indirect bandgap energy (eV), and λ is the wavelength (nm).

Figure S5b shows $(\alpha h\nu)^{1/2}$ versus $h\nu$ and the vertical segment is extended to intersect the $h\nu$ axis to obtain the E_g value of the measured sample. The roughly estimated E_g value is 0.63 eV for the pristine Ge substrate.

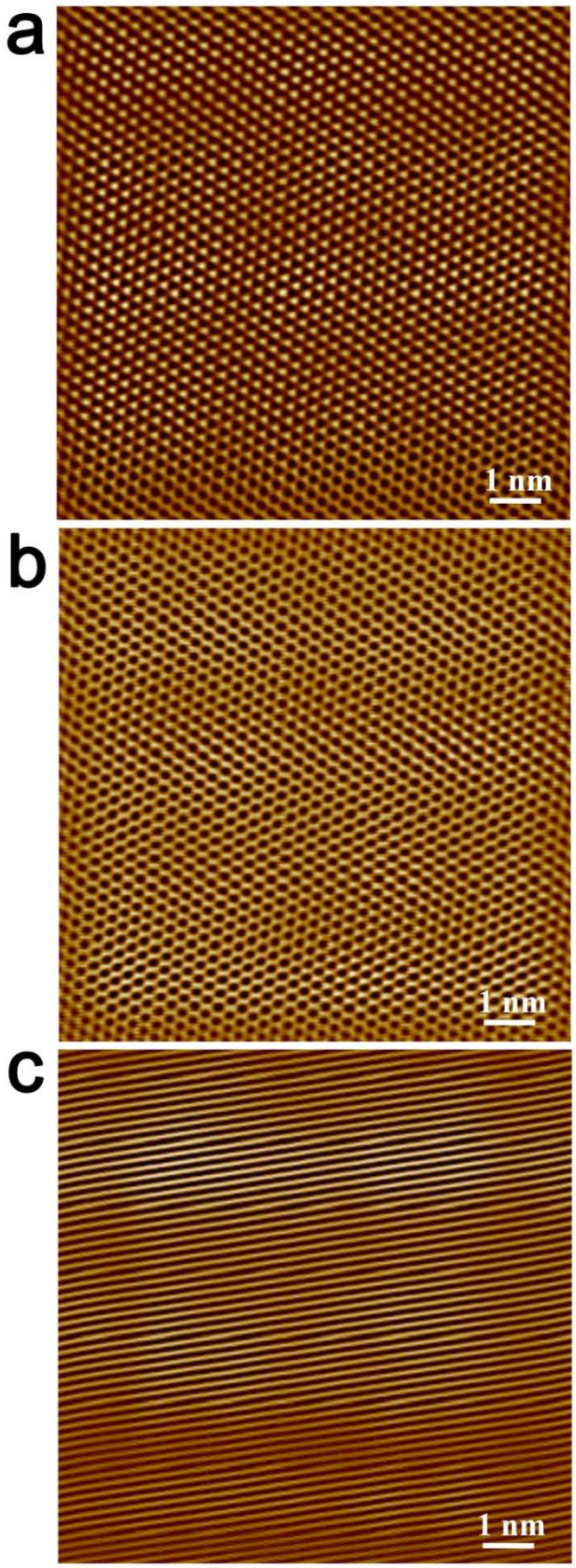

Figure S6. Scanning tunneling microscope (STM) images of graphene films on conductor Cu (**a**), semiconductor Ge (**b**) and insulator SiO_2 (**c**) substrates.

STM analysis was carried out on a MultiMode8 STM base in ambient condition. Based on the operation principle of STM, when the probe tip is brought very near to the graphene film on conductor Cu substrate (**Figure S6a**), the tunneling current can be produced to obtain the microscope image of hexagonal graphene lattice. This is also true for the detecting of graphene film on semiconductor Ge substrate (**Figure S6b**) by STM. However, for the graphene film on insulator SiO_2 substrate, the tunneling current cannot be produced. As a result, the hexagonal lattice of graphene cannot be observed by STM, as shown in **Figure S6c**. The STM results may provide a potential support for the observed electron transfer related bacterial responses toward the large-area monolayer graphene films on different substrates.

References

1. Nair, R. R. et al. Fine structure constant defines visual transparency of graphene. *Science* **320**, 1308 (2008).

2. Serpone, N. et al. Size effects on the photophysical properties of colloidal anatase TiO_2 particles: size quantization versus direct transitions in this indirect semiconductor? *J. Phys. Chem.* **99**, 16646-16654 (1995).