Supplementary information

Enhancing optoelectronic properties of SiC-grown graphene by a surface layer of colloidal quantum dots

Oleg Makarovsky, Lyudmila Turyanska, Nobuya Mori, Mark Greenaway, Laurence Eaves, Amalia Patané, Mark Fromhold, Samuel Lara-Avila, Sergey Kubatkin and Rositsa Yakimova

SI1 Calculations of the electrostatic potential landscape

Our electrostatic model assumes that only QDs in direct contact with the SLG layer are involved in the charge transfer process and that each QD can transfer one electron to the SLG. This assumption is confirmed by the measured doping effect induced by the QD layer and is used to calculate the 2D QD density from the measured Hall concentration $n = N_{imp} + N_{QD} =$ 12×10^{12} cm⁻², where the 2D density of QDs is $N_{QD} = 6.8 \times 10^{12}$ cm⁻². For the calculation of the local electrostatic potential we sum up electrostatic potentials of all impurities and QDs using a dielectric constant $\varepsilon = 10$ [1] and an off-plane distance $z_0 = 1$ nm. Fluctuations of the electrostatic potential (Fig. 3) are numerically calculated as a 2D standard deviation.

SI2 Analysis of 2D charge distribution

The spatial correlation between charged impurities is generated by Coulomb interaction and is described using the minimum distance between impurities, r_0 . Maximum correlation

corresponds to $r_0 = r_i$, where r_i is the maximum possible value of r_0 . In an idealised hexagonal closed packed (*hcp*) 2D lattice, the value of r_0 is $r_{hcp} = \sqrt{\frac{2}{\sqrt{3}}} \frac{1}{\sqrt{N}} \approx 1.075 \frac{1}{\sqrt{N}}$ with

filling factor F = 91%. For the random (uncorrelated) case, the 2D distribution of impurities can be described by a nearest-neighbour distance, r_{nn} , with statistical distribution given by [2]:

$$P(r_{nn}) = 2\pi r_{nn} N \exp\left[-\pi r_{nn}^2 N\right], \qquad (1)$$

where the most probable r_{nn} is $r_{nn}^{\max} = \frac{1}{\sqrt{2}\sqrt{N\pi}}$ and the mean value of r_{nn} is

$$r_{nn}^{mean} = \int_{0}^{\infty} P \, r dr = \frac{1}{2\sqrt{N}}$$
. The red line in Figure S1 illustrates the nearest-neighbour statistics (1).

For numerical modelling of the impurity distribution, we first consider SLG only and place impurities one by one on the randomly generated XY-coordinates within a 1 μ m x 1 μ m area at a distance larger than r_0 from the nearest impurity (blue points in Figure S1). Our calculations demonstrate that an increase of r_0 leads to a narrowing of the nearest neighbour distribution indicating increasing ordering (spatial correlation) and a corresponding increase of the r_{nn}^{max} .



Figure S1. Calculated distribution of the nearest neighbour distance for $N_{imp} = 12 \times 10^{12} \text{ cm}^{-2}$ over the area of 1µm x 1µm for different values of r_0 . The red line is calculated using Equation (1).

For the SLG decorated with a layer of colloidal QDs, we first consider randomly distributed impurities in the SLG ($r_0 = 0$) with concentration $N_i = 5.2 \times 10^{12}$ cm⁻² and then add QDs with $N_{QD} = 6.8 \times 10^{12}$ cm⁻². For the uncorrelated QD distribution (see Figure 3b in the main paper), the QD coordinates were generated randomly using the same algorithm as that used for the SLG impurities. A correlated distribution of QDs (Figure 3c in the main text) was achieved by placing them one-by-one in the minima of the electrostatic potential. The resulting nearest neighbour statistics for both cases is shown in Figure S2. A distinct narrowing of the r_{nn} distribution is observed and is accompanied by the shift from $r_{nn}^{max} = 1.15$ nm for the uncorrelated case to $r_{nn}^{max} = 2.4$ nm for the correlated case, approaching the values expected for the hexagonal packing ($r_{nn}^{max} = 3.2$ nm). This result confirms a significant increase of the degree of spatial correlation in our devices and is consistent with the results of the electrostatic potential calculations (Figure 3 in the main text).



Figure S2. The nearest neighbour statistics calculated in a circular area with radius 1 μ m for randomly distributed impurities with $N_i = 5.2 \times 10^{12}$ cm⁻² and QDs with $N_{QD} = 6.8 \times 10^{12}$ cm⁻² distributed in uncorrelated (black circles) and correlated (blue squares) arrangement. The red line is calculated using Equation (1).

- [1] L. Patrick and W.J. Choyke, Phys. Rev. B 2, 2255 (1970)
- [2] S. Chandrasekhar Rev. Mod. Phys. 15 (1943)