Supplementary information

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Ultra-high vacuum Raman setup

Figure S1 depicts a sketch of the UHV Raman experiment. The laser path, the inverted flange with the optical elements and the alkali metal getter is indicated.

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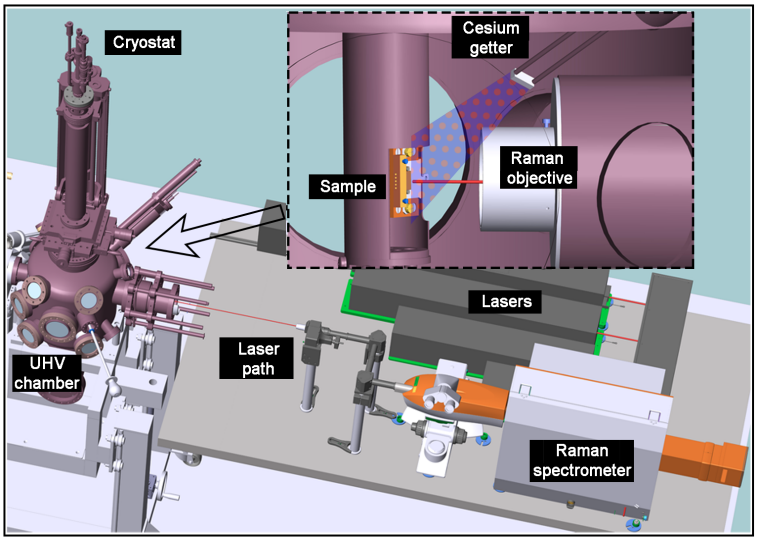

Figure S1: A sketch of the experimental setup that we used for UHV Raman spectroscopy of Cs doped graphene.

Low energy electron diffraction

Electron diffraction (LEED) of all Cs/graphene phases was checked for each experimental method (UHV-Raman, ARPES, STM) separately. LEED as a function of Cs coverage is shown in Figure S2.

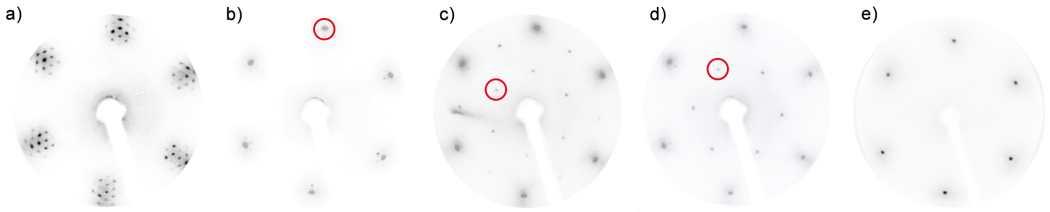

Figure S2: Low energy electron diffraction (LEED) of graphene/Ir(111) with increasing amounts of Cs deposited. (a) pristine graphene/Ir(111), (b) 1×1 phase, (c) 2×2 phase, (d) $\sqrt{3} \times \sqrt{3}$ phase and (e) the overdoped phase.

Angle-resolved Photoemission Spectroscopy

ARPES of the largest doping level achieved is shown in Figure S3.

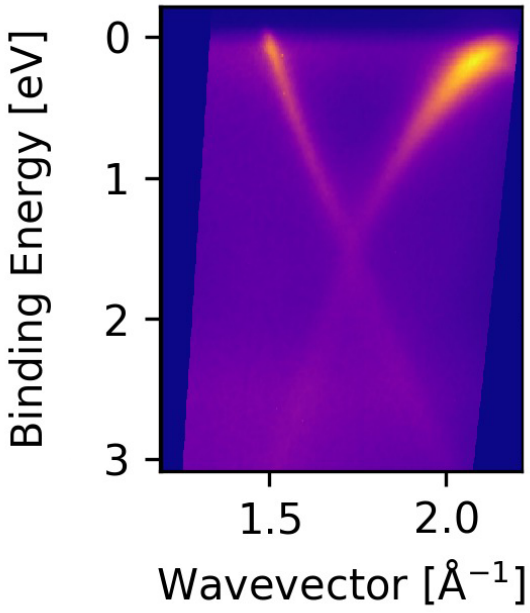

Figure S3: ARPES spectrum Cs doped graphene/Ir(111) corresponding to the highest carrier concentration of $n = 4.4 \times 10^{14}\text{cm}^{-2}$.

Fourier Transform Scanning Tunneling Spectroscopy

Figure S4 depicts scanning tunneling microscopy images together with their Fourier Transforms. Analysis of the scattering vectors allowed us to extract the Fermi surface contours and subsequently the carrier concentrations that are reported in the main paper.

Dopant stability under laser irradiation

Figure S5 depicts UHV Raman spectra ($T = 5$ K) of Cs doped graphene/Ir measured with 2 different laser powers and after exposure to the beam for 3 hours. Since all measurements yield identical spectra we conclude that the sample is stable in the laser beam and that the laser power per unit area is sufficiently low to prevent deintercalation by heating.

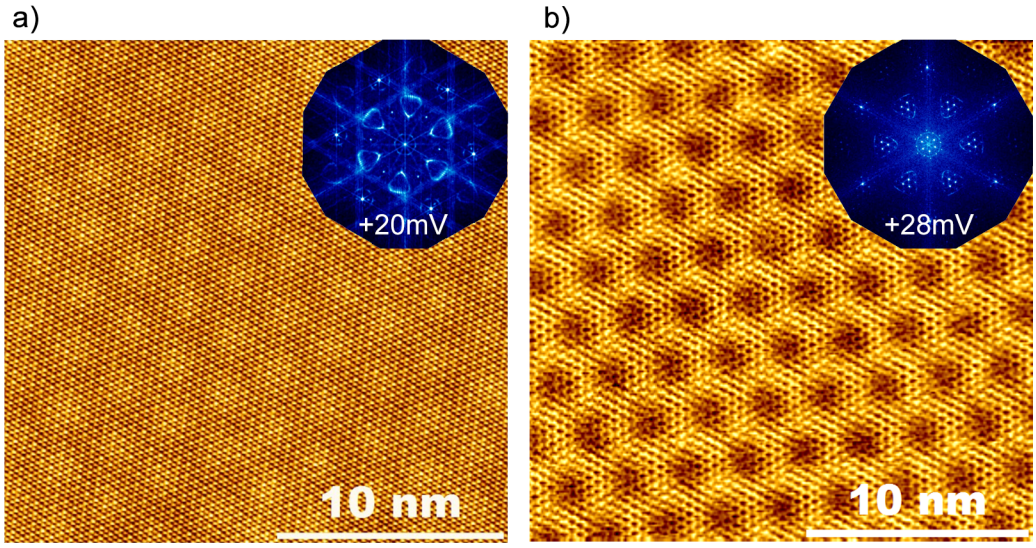

Figure S4: Scanning tunneling microsopy images of (a) 2×2 and (b) $\sqrt{3} \times \sqrt{3}$ phases. The tunneling parameters for the 2×2 image are 128 mV and 31 pA and for the $\sqrt{3} \times \sqrt{3}$ image 28 mV und 100 pA. The insets show Fourier transforms of scanning tunneling spectroscopy (FT-STS) maps. The parameters for the FT-STS maps were 20 mV und 100 pA (2×2) and 28 mV und 50 pA ($\sqrt{3} \times \sqrt{3}$).

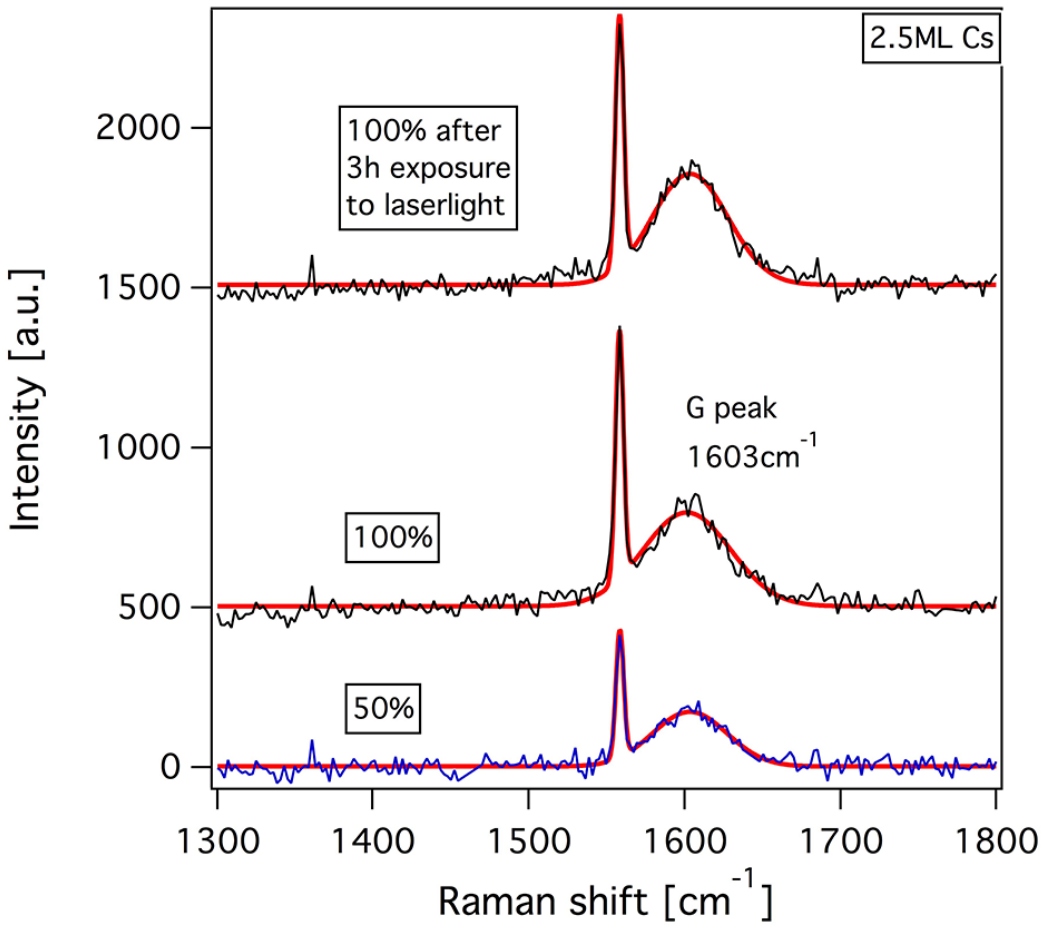

Figure S5: From bottom to top: Raman G band spectra measured at 50% and 100% laser power and after exposure to the laser beam for 3 hours. All measurements were done using the 325 nm excitation line and at T=5 K.