Supporting Information:

Graphene on Ru(0001) Moiré Corrugation Studied by Scanning Tunneling Microscopy on Au/graphene/Ru(0001) Heterostructures

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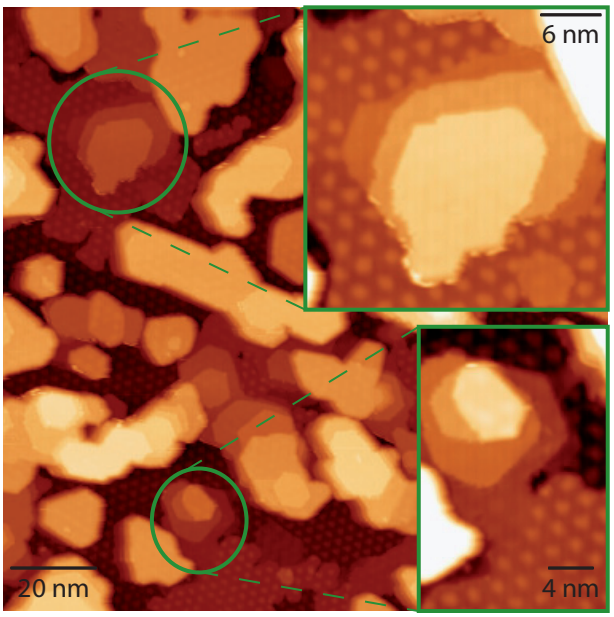

Figure S1: Overview STM image of room temperature deposited Au on monolayer graphene/Ru(0001), observed by UHV STM at 77 K (V_{sample} =-0.27 V, I =0.46 nA). Circles mark islands shown in the close-up views whose terraces are partially or completely flat.

Equation for $A(h)$

The general equations of elasticity for the case of zero body forces can be written in terms of the displacement field $\mathbf{u}$ as:[1]

$$\nabla(\nabla \cdot \mathbf{u}) + (1 - 2\nu)\triangle \mathbf{u} = 0, \tag{1}$$

where ν is the Poisson ratio of the material. Equation (1) admits solutions of the Papkovitch-Neuber form,[2–4] *i.e.*, with the displacement components u_i $(i = 1, 2, 3)$ expressed in terms of a scalar field ϕ and a vector field $\psi \equiv (\psi_1, \psi_2, \psi_3)$ as

$$u_i = \psi_i - \frac{1}{4(1-\nu)} \frac{\partial}{\partial x_i}(\phi + x\psi_1 + y\psi_2 + z\psi_3), \tag{2}$$

where ϕ and ψ_i $(i = 1, 2, 3)$ are harmonic functions to be determined. The boundary conditions that we impose on the film which extends indefinitely in the x and y directions consist in prescribed displacements on the lower surface of the film, and zero applied stress on the upper surface:

$$\mathbf{u} = (0, 0, A_0 \sin \frac{x}{\lambda}), \text{ on the lower boundary } (z = 0) \tag{3}$$

$$\sigma_z = 0, \text{ on the upper boundary } (z = h). \tag{4}$$

Using Hooke's law and the definition of normal strains in terms of displacements (*e.g.*, $\epsilon_1 = \partial u_1/\partial x$), Eq. (4) can be cast as

$$\frac{\partial u_1}{\partial x} + \frac{\partial u_2}{\partial y} + \frac{1-\nu}{\nu} \frac{\partial u_3}{\partial z} = 0, \text{ on } z = h. \tag{5}$$

Since the film extends indefinitely in the (x, y) plane, the requirement of bounded, spatially periodic solutions implies that ψ_1 and ψ_2 should vanish everywhere, which leads to

$u_2(x,y,z) = 0$. The other two harmonic functions, ϕ and ψ_3, can now be uniquely chosen as combinations of exponential and sinus functions:

$$\phi = (C\mathrm{e}^{\frac{z}{\lambda}} + D\mathrm{e}^{\frac{-z}{\lambda}})\sin\frac{x}{\lambda} \quad (6)$$

$$\psi_3 = (E\mathrm{e}^{\frac{z}{\lambda}} + F\mathrm{e}^{\frac{-z}{\lambda}})\sin\frac{x}{\lambda} \quad (7)$$

Substituting Eqs. (6,7) in the boundary conditions Eqs. (3) and (5) leads to a linear system for the coefficients C, D, E, and F:

$$\begin{pmatrix} 1 & 1 & 0 & 0 \\ -1 & 1 & \lambda(3-4\nu) & \lambda(3-4\nu) \\ \mathrm{e}^{h/\lambda} & -\mathrm{e}^{-h/\lambda} & (h-(1-2\nu)\lambda)\mathrm{e}^{h/\lambda} & (-h-(1-2\nu)\lambda)\mathrm{e}^{-h/\lambda} \\ \mathrm{e}^{h/\lambda} & \mathrm{e}^{-h/\lambda} & (h-2(1-\nu)\lambda)\mathrm{e}^{h/\lambda} & (h+2(1-\nu)\lambda)\mathrm{e}^{-h/\lambda} \end{pmatrix} \begin{pmatrix} C \\ D \\ E \\ F \end{pmatrix} = \begin{pmatrix} 0 \\ 4A_0\lambda(1-\nu) \\ 0 \\ 0 \end{pmatrix}. \quad (8)$$

With the solutions of the linear system (8) in place, we can substitute back into Eqs. (6, 7) and (2) to determine the u_3 component of the displacement field. At $z = h$, the amplitude $A(h)$ of this component, *i.e.*, of the the corrugation of the upper surface, works out to be:

$$A(h) = 4A_0(1-\nu)\frac{2(1-\nu)\cosh(h/\lambda) + (h/\lambda)\sinh(h/\lambda)}{2h^2/\lambda^2 + 5 - 4\nu(3-2\nu) + (3-4\nu)\cosh(2h/\lambda)}. \quad (9)$$

When using Eq. (9) to compare to experimental data pertaining to Au/graphene/Ru(0001) heterostructures, several things have to be noted. Firstly, the theory is derived for the Au film alone in absence of any graphene/Ru substrate. The perturbation is imposed on the lowest Au atomic layer, and the results should hold (at least qualitatively) for the Au film and not for the graphene underneath. As described in the text of the article, graphene can have either chemical or physical bonding with the Au, which cannot be captured by elasticity equations. We therefore derived Eq. (9) with displacements imposed on the lowest surface of the Au film and under the assumption that those imposed displacements are not changed

during deposition of Au in any way.

Secondly, when dealing with a bilayer of Au which only has a thickness of 3 Å, there is a possibility that the numerical value of the Poisson ratio used in Eq. (9) changes because the Au film is ultra-thin, which can lead to changes in the predictions based on the elasticity equations. In order to test the sensitivity of the result of Eq. (9) to the value of ν, we have first used DFT calculations to compute the Poisson ratio as a function of the thickness of the Au layer from 2 to 6 ML. The parameters of these DFT calculations are the same as those described in the main text, except for the k-point grid which has been increased to $11 \times 11 \times 1$. We have found that for a bilayer of Au, the Poisson ratio is 0.347, which differs significantly from the bulk value of 0.42. However, it turns out that the normalized amplitudes $A(h)/A_0$ given by Eq. (9) do not change by more that 4 % when the Poisson ratio varies in the range $0.35 \leq \nu \leq 0.42$. Therefore, at least qualitatively, it is warranted to use Eq. (9) for comparison with experiments.

References

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