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Supporting information for

Hole doping, hybridization gap, and electronic correlation in graphene on a platinum substrate

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1. CVD method versus carbon segregation method

When graphene is prepared by the segregation of carbon atoms from single-crystal Pt(111) ^{1,2}, graphene exhibits nearly freestanding nature. On the other hand, when using the CVD method with an external carbon source^{3,4} on the same substrate, single-crystal Pt(111), strong hybridization is observed between graphene and platinum bands, similar to our results obtained from graphene prepared using the CVD method on a polycrystalline platinum foil. These are summarized in Table SI. As a result, crystallinity of Pt does not play a crucial role in the observed hybridization between graphene and Pt.

For the samples prepared using the segregation method (first two panels in Table SI), one can find that two results show somewhat different ARPES intensity maps, i.e., observation of platinum bands which is measured as non-uniform background intensity near the stronger intensity denoted by dotted lines corresponding to the graphene band¹ vs. absence of platinum bands². The difference between the two results is the annealing temperature of the substrate, which allows thermal activation of carbon atoms so that they diffuse out of the bulk. When above 1000 °C data (first panel) is compared to the CVD results (third and fourth panels in Table SI), the platinum bands become weaker and fuzzy. When providing higher energy, i.e., 1600 °C (second panel), platinum bands completely disappear. As a result, when the better quality sample is prepared with the higher temperature segregation method, the quality of the electron band structure of the platinum surface becomes worse, which naturally leads to the absence of the hybridization.

Table SI. Comparison of crystallinity and hybridization for graphene on platinum substrates prepared with different methods.

	Carbon segregation from the bulk Pt		Chemical vapor deposition on Pt	
Crystallinity	Single crystal ¹	Single crystal ²	Single crystal ^{3,4}	Poly crystal (Our work)
ARPES data	(a) 0 - (5 - 1 - 10 - 0.5 - 0 0.5 - 10 - 10 - 0.5 - 0 0.5 - 10 - 10 - 10 - 10 - 10 - 10 - 10 - 1	0 (a) 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7	a graphene/P((111) (A) 2 (A) Abaue Bully	b π 0 -2 -2 -3 -4 -3 -4 -8 -10 Γ Κ
Hybridization	No		Yes	
Preparation temperature	Above 1000 °C	1600 °C	877 °C	950 °C
Observation of Pt bands	Yes	No	Yes	Yes

2. Crystallinity of a polycrystal Pt foil

Typically, the (111) orientation has lower energy so that *fcc* metals such as nickel, copper, and platinum tend to have the (111) orientation facing up after thermal treatment. For example, after being hot-rolled, nickel⁵ and platinum⁶ foils are crystallized mostly into (111) orientation. In addition, as the thickness of a platinum foil decreases from 0.5 mm to 0.2 mm, more than 30 % of the surface was found to have (111) orientation⁷. In our case, the

platinum foil is 0.1 mm thick and the grain is as wide as a few hundreds microns as shown in Fig. S1. The observation of both Pt(111) bands and graphene bands suggests a possibility that the ARPES measurements have been done for a relatively big piece of CVD-grown graphene (two major graphene pieces within the probing area of $40\times80~\mu\text{m}^2$ as discussed in Fig. 2a of our manuscript) on a relatively wide platinum grain with the (111) orientation.

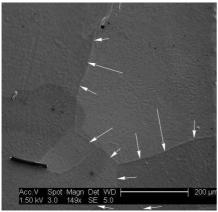


Figure R1. An SEM image of the Pt foil used in our experiments. The white arrows indicate Pt grain boundary.

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