Supporting Information

Strongly Interacting C_{60}/Ir(111) Interface: Transformation of C_{60} into Graphene and Influence of Graphene Interlayer

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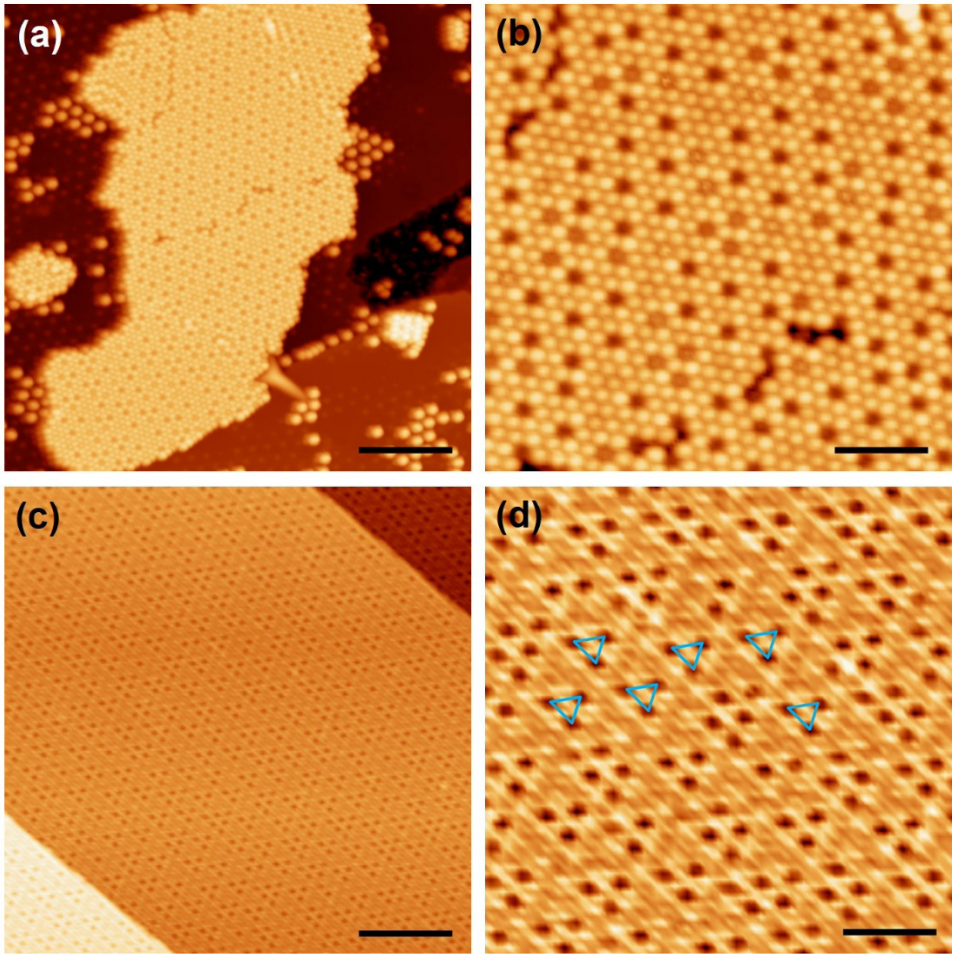
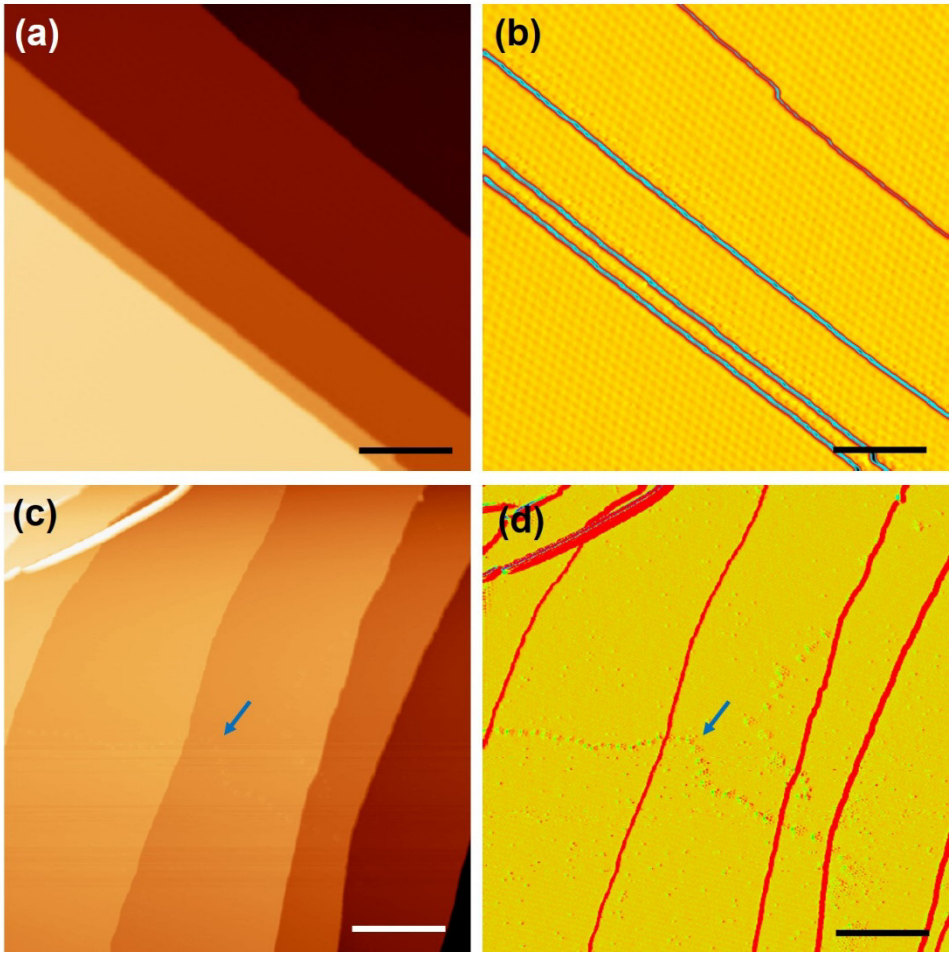
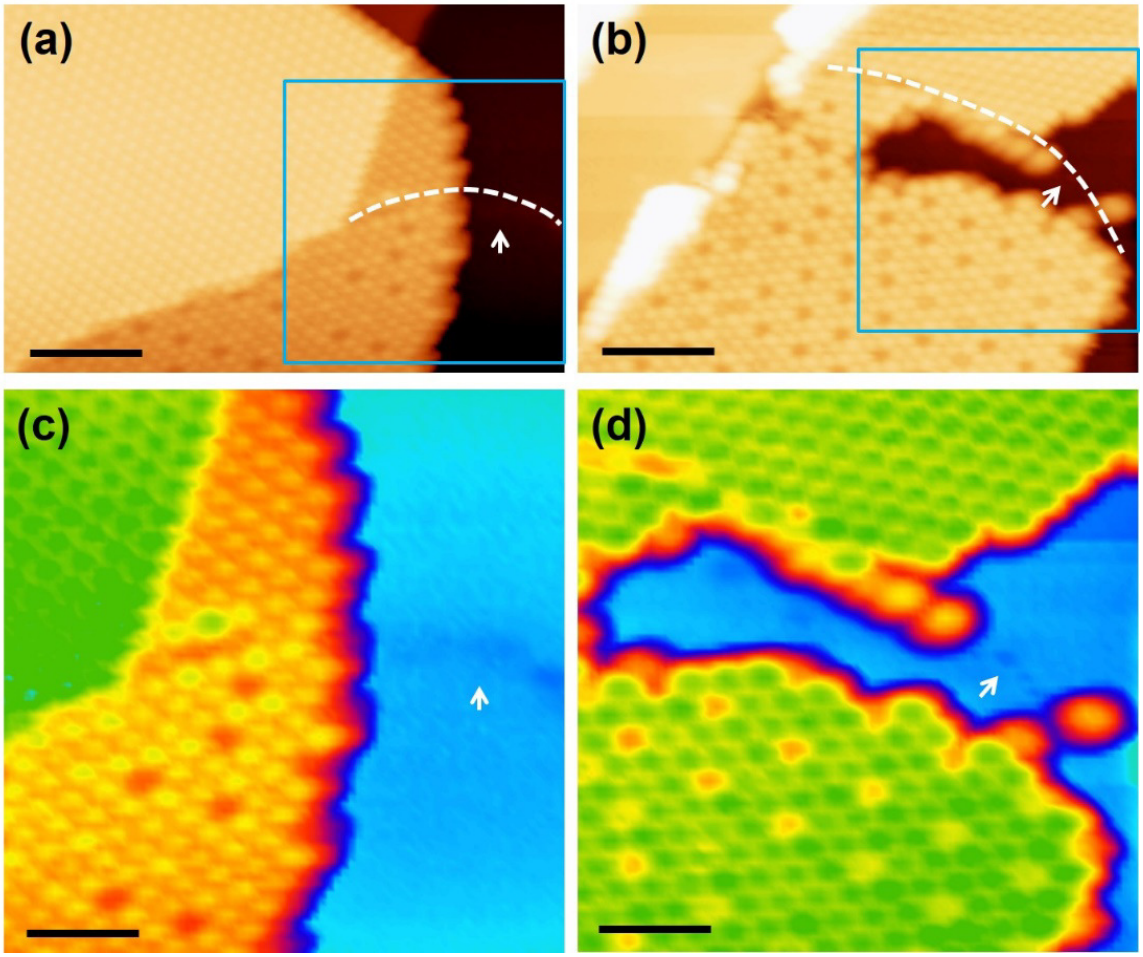


Supplementary Figure S1: Spatial distribution of vacancies in C_{60} islands on the graphene/Ir(111) surface. (a) and (b): Hexagonal vacancy superlattice in which each lattice

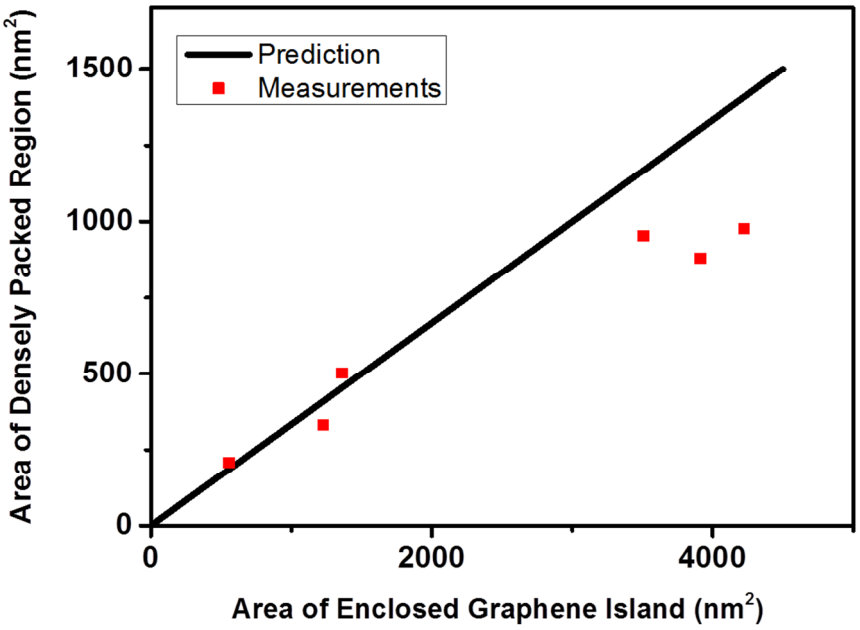
point corresponds to one single-molecule vacancy. (c)-(d): Hexagonal vacancy superlattice in which each lattice point corresponds to three single-molecule vacancies. Scanning Parameters: (a) and (b) V_{bias} = 3 V, I = 50 pA; (c) and (d) V_{bias} = 3 V, I = 10 pA. Scale Bars: (a) 15 nm; (b) 5 nm; (c) 20 nm; (d) 6 nm.



Supplementary Figure S2: Graphene domains on the Ir(111) surface. Graphene was prepared by thermal decomposition of C_2H_4 on the Ir(111) surface. The most predominant domain is R0, as shown in (a) and (b), where the graphene lattice is strictly aligned with the Ir(111) lattice. Different graphene domains coexist on the surface. The domain boundaries are indicated by the arrows in (c) and (d). Images in (b) and (d) are the derivatives of (a) and (c), respectively. Scanning Parameters: (a) and (b) V_{bias} = 50 mV, I = 5 nA; (c) and (d) V_{bias} = 3 V, I = 50 pA. Scale Bars: (a) and (b) 20 nm; (c) and (d) 60 nm.



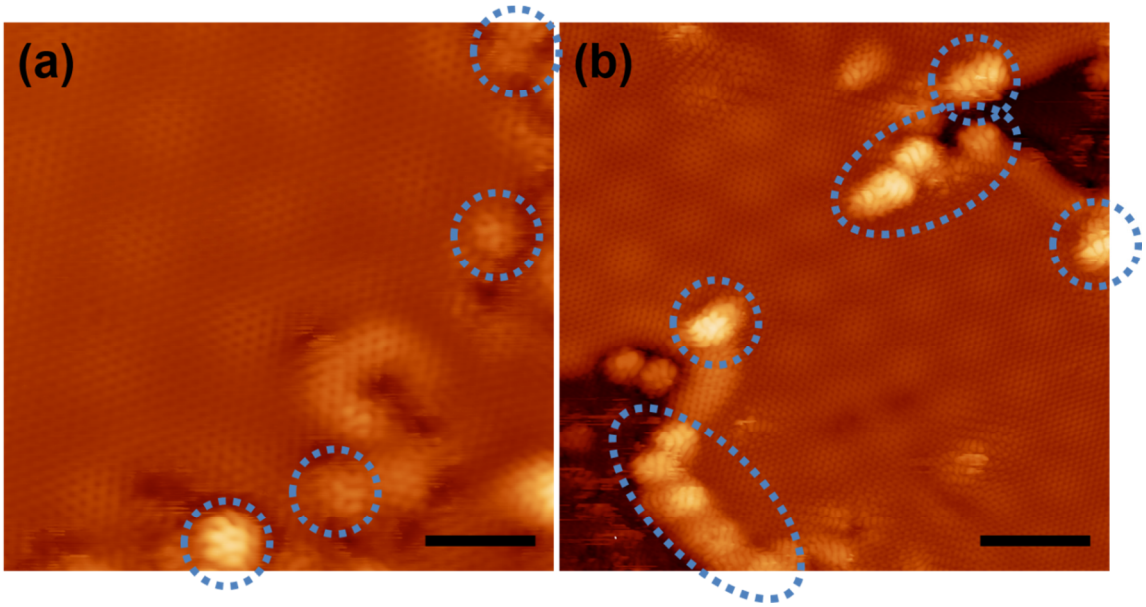
Supplementary Figure S3: Correlation between molecular arrangements and the domains on the underlying graphene/Ir(111) surface. In both (a) and (b), there are two types of molecular arrangements, one with vacancies and the other with no vacancies. The boundaries between the two domains of molecular arrangements are always aligned with the domain boundaries (indicated by arrows) on the underlying graphene/Ir(111) surface. These observations suggest that the formation of vacancies in molecular islands is correlated to the domains on the underlying graphene/Ir(111) surface. Most C_{60} islands have vacancies, which indicates that, in most domains on the graphene/Ir(111) surface, molecular assembly process is modulated by the surface and vacancies are formed to adapt to the underlying moiré superstructures. Scanning Parameters: (a) – (d) V_{bias} = 3 V, I = 500 pA. Scale Bars: (a) and (b) 6 nm; (c) and (d) 3 nm.



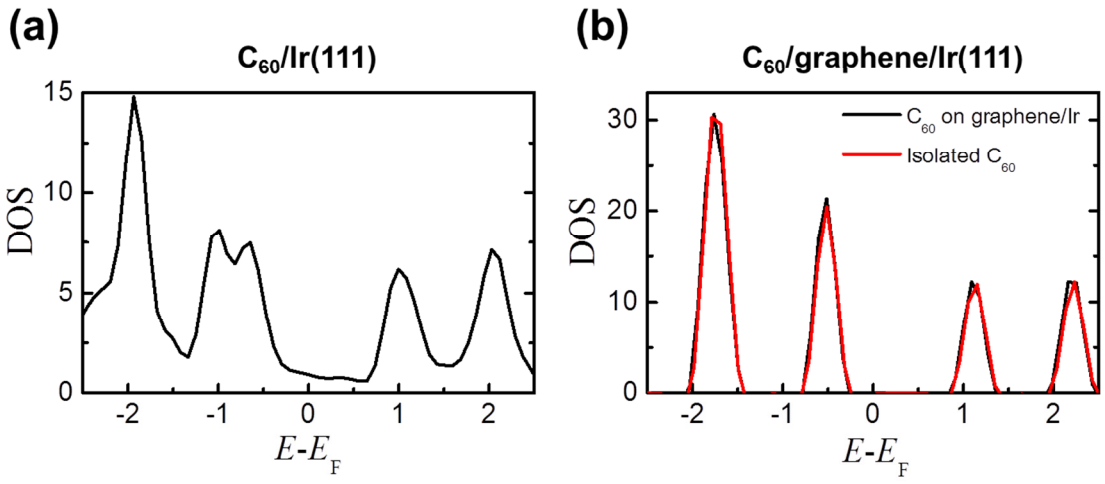
Supplementary Figure S4: Quantitative analysis of the dependence of the area of densely packed regions on the area of enclosed graphene islands. This analysis is for the sample shown in Figures 3a and 3b. The solid black line is the predicted dependence by assuming that the coverage of C_{60} molecules on graphene/Ir(111) is the same as that on Ir(111), 0.25 ML, and that the appearance of densely packed regions results from the diffusion of C_{60} molecules from graphene/Ir(111) to Ir(111). The predicted dependence is:

$$Area\ of\ Densely\ Packed\ Region = \frac{Area\ of\ Enclosed\ Graphene\ Island}{3}$$

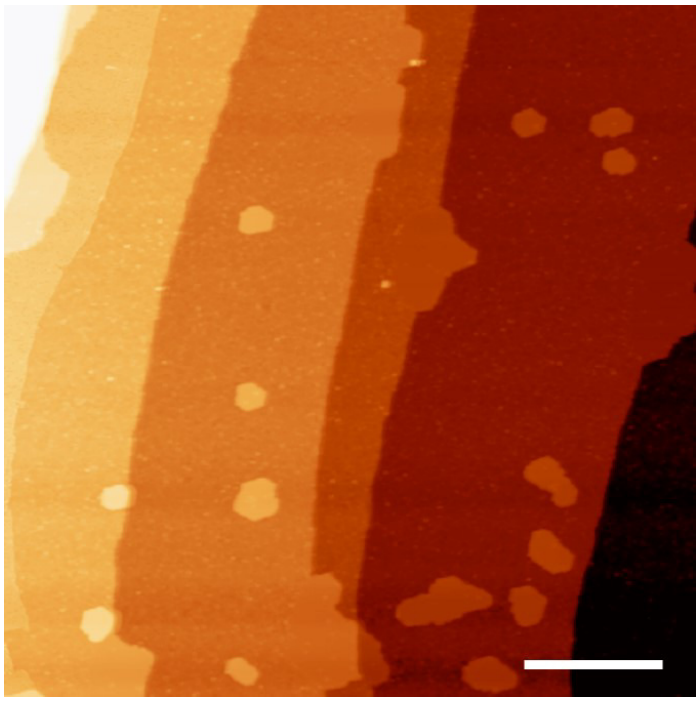
The six data points in red were obtained by analyzing STM images of six different graphene islands on this sample surface. The distribution of these data points is largely in agreement with the prediction. This quantitative analysis corroborates the diffusion process, as described in Section 3.3, of C_{60} molecules on the sample surface under thermal annealing at ~330 K.



Supplementary Figure S5: Intermediate structures between C_{60} and graphene during the growth of (a) single-layer graphene and (b) bilayer graphene through thermal annealing of the C_{60}/Ir (111) surface. Scanning Parameters: (a) V_{bias} = -200 mV, I = 5 nA; (b) V_{bias} = 20 mV, I = 500 pA. Scale Bars: (a) 2 nm; (b) 4 nm.



Supplementary Figure S6: Calculated DOS of the C_{60} molecule on the Ir(111) and graphene/Ir(111) surfaces. The DOS around the E_F is zero for C_{60} on the graphene/Ir(111) surface but not for C_{60} on the Ir(111) surface.



Supplementary Figure S7: Graphene patches obtained by thermal decomposition of C_2H_4 on the Ir(111) surface. In preparing this sample, a clean Ir(111) surface was first exposed to 30 Langmuir C_2H_4 at room temperature, flash annealed to 1350 K, and then annealed at 1170 K for 10 min. The size of the smallest graphene patches is ~10 nm. Scanning Parameters: V_{bias} = 3 V, I = 50 pA. Scale Bar: 60 nm.