Graphene Growth Dynamics on Ir(111): A LEEM Study of Nucleation Kinetics and Morphology

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The morphology of 2D-materials grown by chemical vapor deposition (CVD) is strongly influenced by the choice of substrates. Transition metal surfaces provide enhanced catalytic activity that enable precise growth control like the self-limitation to exactly a single layer of 2D-material. In general, the growth process is governed by competing microscopic processes like adsorption and dissociation of precursors, diffusion, attachment and detachment of elements or molecules of the desired 2D-material, and nucleation, growth, coarsening, and percolation of 2D-islands that ultimately determine the optimum growth parameters. The kinetic limitations on diffusion and nucleation lead to change of 2D-island density $n(T_g,p_{dos})$, which can be controlled by tuning the growth temperature T_g and dosing pressure p_{dos}. In addition, during the deposition process adsorbed species can dissolve into the bulk, reducing the surface concentration. Upon cool down at reducing temperature, the decreasing solubility causes segregation of the dissolved atoms to the surface, influencing the overall quality of the already grown 2D layer. The temperature-dependent solubility of the adsorbed species, combined with optimized growth parameters, thus plays a crucial role in controlling layer uniformity and minimizing defects. Understanding the interplay of diffusion, nucleation of 2D-islands, and segregation is thus essential for tailoring growth conditions to achieve high quality 2D materials with desirable structural and electronic properties. Using *in-situ* and *in-operandi* low-energy electron microscopy (LEEM), we investigated the kinetics of nucleation and growth of graphene island during CVD of ethylene on Ir(111) as function of growth temperature T_g in the range of 750°C to 1050°C at various dosing pressures p_{dos}. We observed heterogeneous nucleation of small graphene islands at pre-existing step edges of the Ir(111) substrate at low T_g or high p_{dos} (cf. Fig.1). At higher T_g or lower p_{dos}, we found a transition towards homogeneous nucleation of large islands. The temperature- and pressure-dependent variations in island density $n(T_g,p_{dos})$ can be described within Venables nucleation theory with a near-linear dependence on dosing pressure $n \sim p_{dos}^{i^*/(i^*+2)}$ attributed to a critical nucleus size $i^* = 5$, i.e., a single carbon hexamer is stable. Dissolution of carbon into the bulk of the Ir substrate causes a transition from the regime of complete nucleation to the regime of initially incomplete nucleation with a much stronger dependence of island density $n \sim p_{dos}^{i^*/2}$ as function of dosing pressure.

Figures

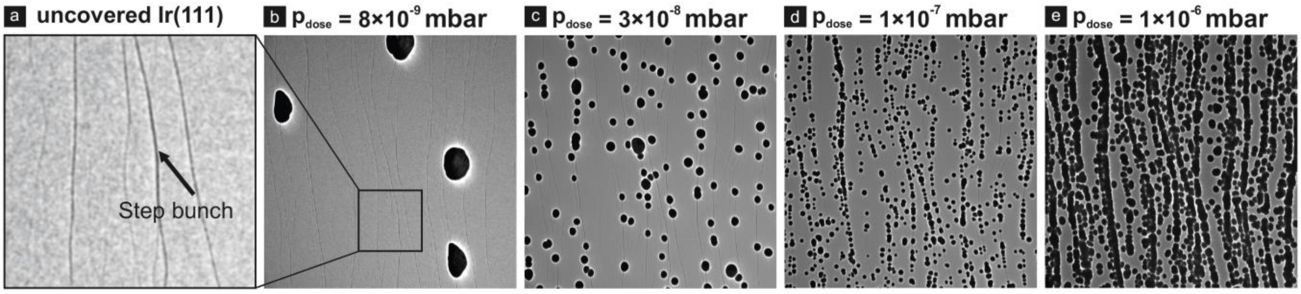

Figure 1: LEEM morphology showing variation in graphene island density on Ir(111) surface depending on the ethylene dosing pressure at the same growth temperature of 890°C. The field of view is 10 micron for (a) and 40 micron for (b)-(e)