The precisely controllable growth of self-aligned single-crystal graphene grains on liquid Cu surface by ambient pressure chemical vapor deposition is reported. Large scale monolayer graphene arrays are modulated by varying growth conditions such as flow rate of carbon source, growth temperature, and growth time. Further, bilayer graphene grains are also controllably prepared under optimized growth conditions. The self-alignment mechanism of graphene is also studied and a growth model is proposed to explain that process involving surface tension of liquid phase. In all, the growth mechanism of graphene arrays is firstly probed and the grown graphene arrays show reasonable mobility and high current density, posing great potential for graphene-based electronics.