Surface Chemical Choreography of Semiconductor Nanowire Synthesis

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Semiconductor nanowires are emerging as indispensable nanoscale building blocks for next generation energy conversion, electronic, and photonic devices. The bottom-up vapor-liquid-solid (VLS) mechanism – whereby a liquid eutectic "catalyst" droplet collects precursor molecules (or atoms) from the vapor and directs crystallization of the solid nanowire – is a nearly ubiquitous method for nanowire synthesis. The ability to control morphology, composition, and/or crystal phase along the nanowire length is a key benefit of VLS growth. However, except for a small number of cases, the mechanistic understanding of this process remains insufficient to *a priori* program nanowire structure and function. To this end, we combine *in situ* infrared spectroscopy and electron microscopy to probe the heterointerfacial chemical processes that choreograph growth. Our studies of Si and Ge nanowires reveal the dramatic influence of various species adsorbed on the sidewall. We find, for example, that changes to hydrogen atom coverage can modify catalyst phase and nanowire crystal orientation. These results demonstrate that changes to surface bonding are critical to understand nanowire synthesis and provide new guidelines for rationally selecting catalysts, forming heterostructures, and controlling dopant profiles. Our insights also permit the design of novel precursors with which to engineer a range of structures, including defect, kinking, doping, and diameter-modulated superstructures.