Comparing Au-seeded and self-seeded nucleation of GaAs nanowires using in situ TEM

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When modeling growth and geometry of nanowires grown via the VLS mechanism, the diameter of the nanowire is determined by the nanowire-seed particle interface. Modeling the process usually requires an already formed crystal whose radial geometry does not significantly change as the crystal grows vertically. Less research has been done on the very first stages of crystallite nucleation which eventually leads to formation of the nanowire. Nucleation of a new crystallite in a free droplet or in a droplet on an amorphous substrate differs importantly from surface nucleation on a particle-crystal interface. Nanowires grown on crystalline substrates generally follow the substrate epitaxy. Without a crystalline substrate, droplet seeded growth of crystalline nanowires is certainly possible, but the nucleation kinetics are likely to be different and a single droplet may even exhibit multiple nucleation sites.

In this study, we use in situ ultra-high vacuum transmission electron microscopy (UHV-TEM) to study the initial stages of Au-seeded and self-seeded nucleation of GaAs nanowires. To the microscope, arsine (AsH3) and trimethylgallium (TMGa) were attached as precursor gases. Aerosol Au particles or thin Au films were deposited on Si_3N_4 membranes to act as Au seed particles; to form Ga droplets, we used beam induced in situ formation of Ga droplets from TMGa on the same type of membranes. The Si_3N_4 membranes act as an inert, electron transparent window and allow nucleation studies without chemical reaction between substrate and particle. After nucleation of GaAs crystallites, we could observe subsequent growth into GaAs nanowires.

For Ga droplets, we will show nucleation of single crystallites of GaAs within several minutes of opening the As source. For Au particles, we observe initial alloying with Ga, supplied from the decomposition of TMGa. The AuGa alloy has a lower melting point than Au, and by tuning the amount of Ga available to form AuGa, a liquid AuGa forms. This reaction to form liquid takes place at the surface of the Au particle and progresses inwards to leave a solid Au core that eventually disappears. Only after the complete seed particle has transformed into liquid AuGa does GaAs nucleation take place. We will discuss these observations based on the bulk equilibrium phase diagrams and discuss nanoscale and non-equilibrium effects. Furthermore, we find a narrower temperature range for nucleation compared to subsequent growth, from both AuGa and Ga droplets. When a nucleus has formed, crystal growth can continue at higher temperatures at which nucleation appears to be suppressed. We will discuss the lower temperature required for nucleation in terms of supersaturation and surface kinetics.