This work investigates III-V nanowires synthesized via the vapor-liquid-solid method, whereby a catalyst droplet promotes one-dimensional growth. By combining molecular beam epitaxy experiments, structural characterization and theoretical analyses, I study and clarify several critical issues.

One of them is the control of the crystal phase, which is frequently found to be a mix of cubic and hexagonal segments. By performing a probabilistic analysis of the stacking sequence of InP nanowires, I show that phase selection is determined not only by growth conditions but also by interactions between layers. I highlight and discuss the role of the edge energy of the nucleus that mediates the formation of each monolayer.

Another important problem is the formation of axial heterostructures, which interface sharpness is severely limited by material accumulation in the droplet (‘reservoir effect’). To this end, I study the formation of such heterostructures in Ga-catalyzed GaAs nanowires using either a second group V element (P) or a second group III element (Al). The composition profiles of the ternary insertions are analyzed with monolayer resolution. The interface widths are found to be larger [Ga(As,P)] or narrower [(Al,Ga)As] than expected, and the morphology of the growth front depends on supersaturation. In both cases, I demonstrate that the interface width can be reduced to a few monolayers and suggest further improvements.

Attempts to achieve ultrathin GaAs and GaP nanowires that would permit lateral quantum confinement are presented. Finally, I consider the possibility of minimizing the stochastic character of nucleation ultimately to control the growth of single monolayers.

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