Origins of Order and Disorder in Self-Assembled Epitaxial Quantum Dots

L. H. Friedman

Pennsylvania State University, Department of Engineering Science and Mechanics, 212 Earth and Engineering Sciences Building, University Park, Pennsylvania, 16802, United States of America, email:lfriedman@psu.edu

Self-assembly of epitaxial quantum dots represents an important step in the advancement of semiconductor fabrication at the nanoscale that will allow breakthroughs in electronics and optoelectronics. Self-assembled quantum dots (SAQDs) form as the result of unstable film growth usually in the Stranski-Krastanow growth mode that is caused by elastic strain due to lattice mismatch between the film and substrate material. [1] The two most studied SAQD systems are Ge_xSi_{1-x} dots grown on a Si substrate and $In_xGa_{1-x}As$ dots grown on a GaAs substrate. The former is most interesting for electronic applications and possibly optoelectronic applications, and the latter is most useful for optoelectronic applications. An important step in developing SAQD technology is to control randomness and disorder in SAQD size and SAQD position. One would like to either a) generate large arrays of regularly spaced and/or regularly sized SAQDs or b) be able to place SAQDs in desired locations. The self-assembly process itself can be thought of as a competition or interaction between random and deterministic influences in film growth. Thus, it is critical to study this interaction to further the viability of SAQD technology. Here, this interaction is studied theoretically to shed light on factors that enhance and detract from SAQD order and to investigate a proposed method to influence SAQD formation called thermal-field directed self-assembly. [2]

SAQDs form from an initially flat film surface that is in unstable equilibrium. The initially flat surface forms undulations that finally evolve into island structures that are electronically usable, a process explained as the Asarro-Tiller-Grinfeld instability. [3] These SAQDs nucleate homogeneously, meaning that the process is an example of spontaneous symmetry breaking, and it is inherently driven by random fluctuations. While these fluctuations are useful in providing the initial perturbations required for SAQD formation, the implied cost is that the random origin of the SAQDs will lead to disorder that detracts from their electronic and optical properties or renders them useless, depending on the intended device application.

All current evidence seems to indicate that some initial randomness is maintained in the system despite any natural tendency to order or disorder. It has been suggested that under some circumstances (material parameters and growth conditions), that SAQDs have a natural tendency to order [4, 5], but in other circumstances they will tend towards long-term unstable growth [4] that is probably characterized by ripening (growth of larger dots at the expense of smaller ones). In either case, perfect arrays of SAQDs are difficult to form. The disorder from the initial stages of growth is either frozen in for the case of naturally ordering systems, or it provides an initially disordered state that only becomes

more disordered during ripening. In this light, SAQD formation and growth may be viewed as a competition/interaction between random and deterministic effects.

In order to model this interaction between random and deterministic effects in SAQD formation, a continuum approach is used to model the film growth, similar to [2,4,6] that includes random noise in either the initial conditions or in the equations governing film growth. The film surface evolves via surface diffusion giving a surface velocity

$$v_n = \vec{\nabla}_S \cdot D\vec{\nabla}_S \mu + Q + \zeta$$

where $\vec{\nabla}_S$ is the surface gradient, D is the diffusivity, μ is the diffusion potential, Q is surface deposition, and ζ is the thermal noise inherent in surface diffusion. The surface diffusion is driven by a chemical potential,

$$\mu = \mu_{\text{elastic}} + \mu_{\text{surface}} + \mu_{\text{wetting}},$$

where the *elastic* term tends to destabilize planar film growth, and the *surface* and *wetting* terms stabilize planar growth. These growth equations are solved both numerically and analytically to study the effects of initial noise and thermal noise on overall disorder and to investigate strategies for enhancing SAQD order.

This model is also used to investigate the effectiveness of thermal-field directed self-assembly. [2] Strategies to direct self-assembly of quantum dots are often based on changing the overall landscape in which SAQDs form. Some sort of spatial inhomogeneity is induced so that dots nucleate at preferential sites. The competition between randomizing influences and the ordering effect will determine the overall effectiveness of any given method to direct self-assembly. Three prior examples of directed self-assembly of quantum dots are the preferential nucleation of SAQDs above intersection points of an array of misfit dislocations [7], nucleation of quantum dots on etched mesas [8], and the ability to nucleate SAQD clusters using a FIB (focused ion beam). [9] This study shows that thermal-field directed self-assembly is promising, but must be followed up with further simulations and experiments.

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