Capillary driven strain-free synthesis of epitaxial PbTe/CdTe quantum dots for min-infrared devices

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Self-assembled semiconductor quantum dots are commonly produced by strain-driven Stranski-Krastanow growth of strained-layer heteroepitaxy. The resulting quantum dots have been widely employed for visible and near-infrared photonics but efficient mid-infrared emission has been difficult to achieve due strain-induced type II band alignments. As a solution, we have invented a strain-free synthesis method that is based on capillary driven phase separation and nano-precipitation rather than on epitaxial strain [1]. This method has been applied to the PbTe/CdTe model system, which is a combination of a narrow gap IV-VI semiconductor with a wide gap II-VI semiconductor, featuring identical lattice constants but different crystal structures. As shown by Fig. 1, due to phase separation, 2D PbTe/CdTe epilayers grown by molecular beam epitaxy onto GaAs (001) transform into isolated quantum dots during post annealing. The resulting QDs show highly symmetric shapes and their size is well controlled. This leads to an exceedingly wide tunability of emission over the whole 1.5 to 4 µm wavelength region [1], well suited for optoelectronic device applications.

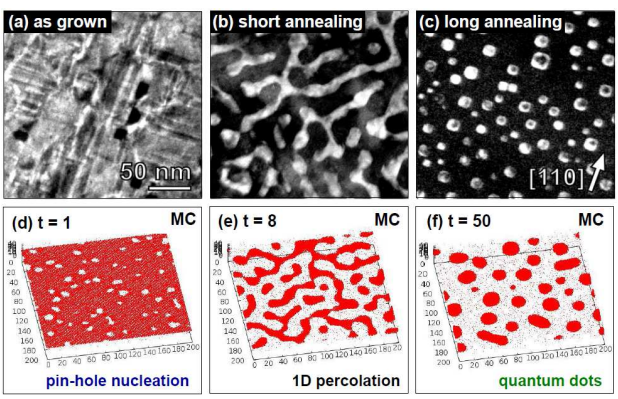

Figure 1: Formation of PbTe/CdTe nanocrystals by capillary driven nanoprecipitation from intially 2D epitaxial PbTe layers embedded in CdTe.
Top: Pan-view TEM images for (a-c) as grown, short and long annealed sample.
Bottom: Coarse grain Monte Carlo simulation of the topological transition, revealing the three characteristic transformation stages.

To unravel the mechanism involved in this synthesis method, in the present work we have developed a Monte Carlo model that accounts for thermodynamics by appropriate choice of the nearest and next-nearest neighboring bond energies. As demonstrated by Fig. 1, this model captures all essential features observed by *in situ* TEM annealing experiments, namely, the initial nucleation of interpenetrating pin-holes in the 2D epilayers that rapidly grow in size to form a one-dimensional percolation network, which then subsequently split up further into isolated QDs (*cf.* lower part of Fig. 1). Further annealing leads to coarsening by Ostwald ripening. As demonstrated by our calculation, this *topological transition* is driven by interface-energy minimization of the two phase system and the key kinetic parameters of this process are identified. Quantitative comparison of the size distribution, density and shapes shows excellent agreement between calculations and experimental data, including also the results from optical measurements. Based on the energetics, we propose a generic analytic model that predicts the size of the quantum dots as a function of the initial structure parameters that can be applied to arbitrary material systems.

[1] H. Groiss *et al*., APL **91** 222106(2007). A. Hochreiner *et al*., APL **98** 021106(2011); APL **100**, 113112 (2012).
[2] H. Groiss, I. Daruka *et al*., submitted.