NANOPARTICLE NETWORKS ON SILICON: SELF-ORGANISED OR DISORGANISED?

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A structural motif that appears very frequently not only in a wide range of nanostructured systems but on mesoscopic to macroscopic length scales is the *cellular network*. We present a quantitative analysis of the morphology of cellular networks formed by thiol-passivated Au nanoparticles, and, for comparison, organometallic molecules, spin cast onto native oxide-terminated silicon substrates. The structural parameters determined from Voronoi tessellation, and Minkowski functional analyses of the experimental data are compared to those extracted from Monte Carlo simulations of nanoparticle network formation. The key result of this comparative study is that although the cell positions are spatially correlated, i.e., they deviate strongly from those expected for a Poisson point set, this correlation arises simply from a coalescence of neighbouring cells during network formation. Complex non-linear processes such as spinodal decomposition or Marangoni convection are therefore not *always* a prerequisite for the formation of spatially correlated networks.[1] Recent results have indicated that it is possible to selectively and locally modify the spatial order in nanoparticle assemblies on H-terminated silicon substrates. Lithographic oxidation by an electrically biased AFM tip has been used to produce nanoscale variations in surface chemistry, which interfere with the far-from-equilibrium assembly processes. This ability to tune the pattern forming mechanisms on sub-micron and nanometre length scales has particular potential in the development of novel percolating pathways in nanoparticle systems.

[1] Martin, C. P.; Blunt, M. O.; Moriarty, P. J. *Nano Letters* **2004**, *4*, 2389-2392

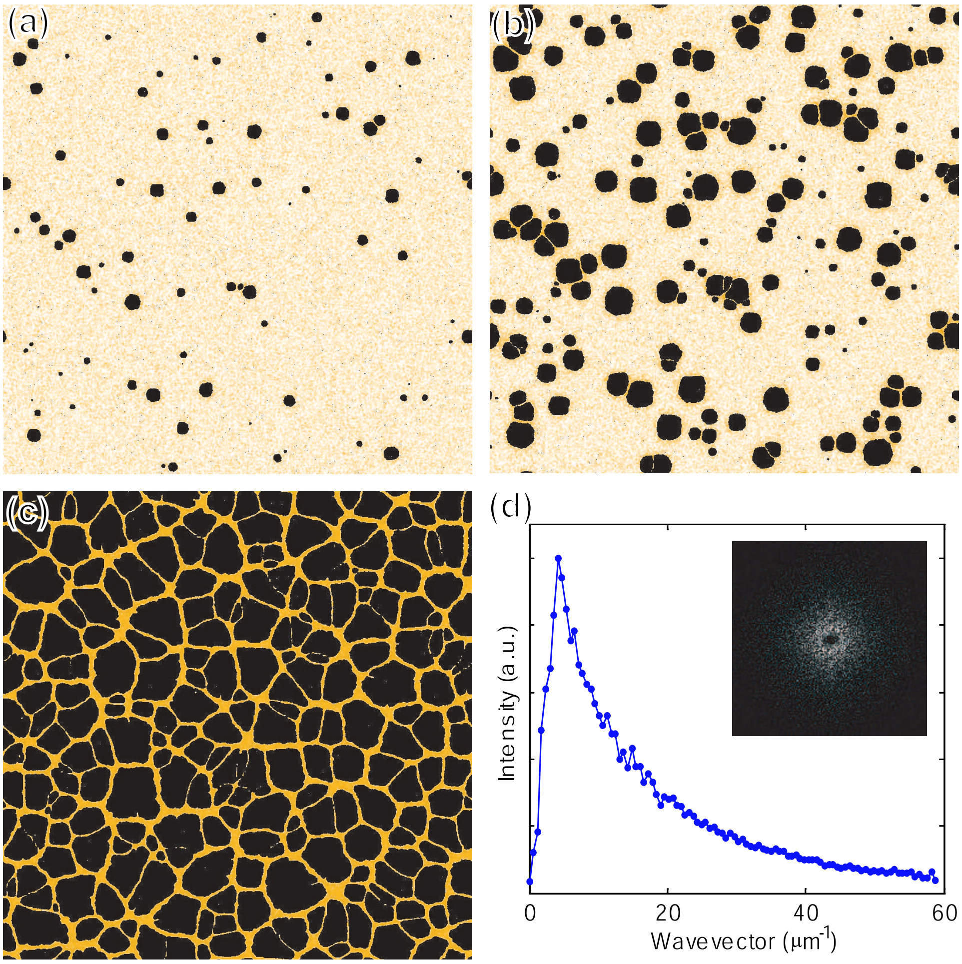

Figure 1: Results from the heterogeneous (nucleation) regime, with $k_BT = \epsilon_l/4$, $\epsilon_n = 2\epsilon_l$, $\epsilon_{nl} = 1.5\epsilon_l$, $\mu = -2.25\epsilon_l$, MR=30, and a nanoparticle coverage of 20%, showing the early stages of pattern formation in a 4008x4008 pixel system after (a) 99 MC steps, with a distribution of nucleation sites that is clearly uncorrelated, (b) 199 MC steps, illustrating coalescence of neighbouring nucleation sites, and (c) 899 MC steps, the stable end result, which is best described as a cellular network. Pane (d) shows a radially averaged two-dimensional Fourier transform of (c), with a clear peak resulting from the ring visible in the inset.