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Generic Phase Diagram of Binary Superlattices[1] ALEXEI TKACHENKO, Brookhaven National Laboratory — Emergence of a large variety of self-assembled superlattices is a dramatic recent trend in the fields of nanoparticle and colloidal sciences. Motivated by this development, we propose a model that combines simplicity with a remarkably rich phase behavior, applicable to a wide range of such self-assembled systems. Those include nanoparticle and colloidal assemblies driven by DNA-mediated interactions, electrostatics, and possibly, by controlled drying. In our model, a binary system of Large and Small hard sphere (L and S)interact via selective short-range ("sticky") attraction. In its simplest version, this Binary Sticky Sphere model features attraction only between 'S' and 'L' particles, respectively. We demonstrate that in the limit when this attraction is sufficiently strong compared to kT, the problem becomes purely geometrical: the thermodynamically preferred state should maximize the number of S-L contacts. A general procedure for constructing the phase diagram as a function of system composition f, and particle size ratio r, is outlined. In this way, the global phase behavior can be calculated very efficiently, for a given set of plausible candidate phases. Furthermore, the geometric nature of the problem enables us to generate those candidate phases through a well defined and intuitive construction. We calculate the phase diagrams both for 2D and 3D systems, and compare the results with existing experiments. Most of the 3D superlattices observed to date are featured in our phase diagram, while several more are yet to be discovered.

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