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Exact results for the jammed state of binary mixtures of superdisks on the plane

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ABSTRACT

By analytical and numerical methods we investigate the late stage deposition of binary mixtures of oriented "superdisks" on a plane. Superdisks are objects bounded by Lamé curves $|x|^{2p} + |y|^{2p} = 1$, where deformation parameter p controls their size and shape. For deposition of single-type superdisks, the maximum packing and jamming densities are known to be nonanalytic at $p = 0.5$. For binary mixtures of superdisks, we discover that nonanalyticities form a locus of points separating "phase diagram" of shape combinations into regions with different excluded-area constructions. An analytical expression for this phase boundary and exact constructions of the excluded-areas are presented. The corresponding saturation coverages are obtained by extensive numerical Monte Carlo simulations.

Covering of planar surface with differently shaped, non-overlapping geometrical objects is the subject of perennial interest to mathematicians, physicists, and engineers for both its theoretical and experimental importance. Theoretical studies have generally focused on topics such as maximum packing density [1], stacking and clustering [2,3], space filling [4–6], and other collective arrangements influenced by geometric features of basic building units. Almost all studies of this kind have considered convex units, usually circles, spheres, polyhedra, ellipsoids, etc.

On the experimental side, however, it is now possible to synthesize and manufacture particles (objects), ranging in size from sub-micrometer to nanometer scale [7], with a wide assortment of well defined shapes and morphologies [8,9], including concave, star-like, and a variety of other irregular forms. Note that these particles are synthesized under nonequilibrium conditions and their shapes are usually not that of equilibrium crystals; yet they remain thermodynamically stable in shape and size over time scales of experimental interest.

Particles prepared in this way are often deposited on variously modified surfaces to improve and/or achieve new functionalities in applications in metallurgy, biomedicine [10], optoelectronics [11] and other emerging fields of high technology [12]. In many applications it is often advantageous to use the *mixture* of particles to be deposited. For instance, the process of sintering is improved with the use of binary mixture of particles with different sizes [13]. Similarly, in inkjet printing technology [9], nanosize silver particles are deposited on a supporting surface to form a film which is subsequently sintered to produce electrically conducting structure with desired properties. This experiment [9] also revealed that the conductivity of the final product is improved if the pre-sintered film is formed using the *mixture* of particles of two different sizes (the size ratio of about 1 : 8), instead of the single-size units.

Thus, the large scale properties of the deposited film, e.g., surface coverage, jamming limit, or late stage kinetics of deposition, and their dependence on shapes and sizes of depositing particles, have stimulated renewed interest recently. For

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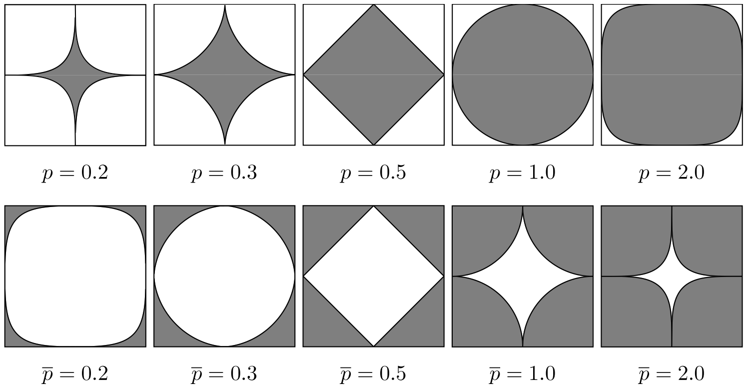

Fig. 1. Families of Lamé shapes, $\mathcal{L}$, (top panel, gray areas) and corresponding Lamé-like shapes, $\overline{\mathcal{L}}$, (bottom panel, white areas) for several values of deformation parameter p. Note the change in convexity at $p = 0.5$ in both families. By construction, the binary mixtures of gray shapes from top panel, and white shapes from bottom panel perfectly match to cover the plane when $p = \bar{p}$.

the purpose of such studies, two-dimensional Lamé objects (superdisks), defined as the set of points in the plane bounded by the curves $|x|^{2p}+|y|^{2p} = 1$, with $p \in (0, \infty)$, that include large family of shapes, from concave ($p < 0.5$) to convex ($p > 0.5$), are particularly suited (see Fig. 1, top panel). As indicated above, recent experimental advances have made it possible to synthesize thermodynamically stable nano and microsized particles of almost any shape. This makes studies of unusually shaped particles, such as Lamé objects, more relevant and timely. Apart from theoretical interest in this problem, it finds applications in studies of design and control of prepatterned surfaces with special properties. New capabilities to pattern surfaces at the nanoscale, and use nanosize particles, have promise for development of novel biosensors and detectors.

In recent study, Gromenko and Privman[14] have examined Random Sequential Adsorption (RSA) model [15] for deposition of superdisks on planar surface and have shown that the jamming limit (i.e. the instance of coverage beyond which no further additions are possible) exhibits nonanalytic behavior when object's shape changes convexity, at $p = 0.5$ (see below). Additionally, it was shown [16] that consequent nonanalyticities are also present in the late-stage kinetics of deposition at the same value of *deformation parameter* p. An important recent work on optimal packing of superdisks [1], for all values of p, reported similar results for nonanalyticities of densest packings. In the above studies, the superdisks used were all of the single type (shape), with a specified and fixed value of deformation parameter p.

However, as mentioned, the use of *mixtures* of shapes (or sizes) for deposition or packing is attracting considerable experimental and theoretical interest. For instance, in recent work by Conway et al. [6], space-filling by Platonic solids is only achieved by using the appropriate mixture of tetrahedra and octahedra. Further, deposited mixtures may sometimes exhibit mixture-specific behaviors, like spontaneous ordering, or size segregation, as reported in experiments with nanoscopic gold particles [17], which makes mixture depositions an interesting object of study in its own right (within the simple RSA model, considered in this work, segregation behavior cannot be captured since deposited particles are not allowed to migrate). For depositions of mixtures, what limited theoretical work there is confined to the studies of objects of the same shape but different sizes. These include mixture of circles (spheres) of different radii [18–21] (but see also [22]), or mixture of line segments of different lengths [23,24]. These studies were primarily devoted to kinetics of the late-stage approach to saturation (jamming).

In this work we report exact and numerical results for Random Sequential Adsorption of binary mixtures of oriented (aligned) convex/concave objects on planar surface, and study new instance of plane tiling by appropriate combination of Lamé and Lamé-like shapes (see below). The motivation is to investigate the origin of the singularity at $p = 0.5$, seen in previous studies [1,14,16]. Namely, it is unclear if this singularity is caused by (i) the change of convexity of depositing objects at $p = 0.5$, or (ii) the fact that $p = 0.5$ superdisks (squares) perfectly cover the plane. We focus on this particular point, $p = 0.5$, because with the advent of nanotechnology, and with the proliferation of experiments on deposition of proteins, one expects that situations will be realized when the particle shapes on the surface change between concave and convex depending on the physical and chemical conditions of the environment. This change might affect the asymptotic approach to the jamming coverage, an issue that calls for a separate study. As demonstrated in this and other works [14,16], the change in the concavity also results in specific nonanalytic behavior of the jamming coverage.

A note is in order regarding the choice of oriented (aligned) superdisks in our simulations. Namely, such a choice may appear somewhat artificial and of modest significance for real experiments. While most earlier experimental setups study deposition of objects with random orientation [15], the use of prepatterned substrates in new technologies has made it increasingly important to deposit objects with parallel (aligned) orientation, as demanded in, e.g., plasmonics applications [25]. Theoretical interest in RSA of aligned objects has been stimulated by the hypothesis of Palasti [26] that the jamming coverage of squares in two dimensions exactly equals the square of the maximum coverage of segments in one dimension. Palasti hypothesis was tested by extensive numerical simulations [27] and was shown to be only approximately correct, with the error of about 0.5%. Additionally, these simulations have revealed that the difference between maximum