Conformal invariance in two-dimensional cluster-cluster aggregation

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It is conjectured that reaction-limited cluster-cluster aggregation (RLCA) in two dimensions (2D) displays conformal invariance. In support of this hypothesis, it is found that structure functions computed for on-lattice aggregation are asymptotically invariant under conformal transformation as $t\rightarrow\infty$. Further, the fractal dimension determined for 2D RLCA is in good accord with predictions based on conformal invariance. [S1063-651X(98)05112-5]

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I. INTRODUCTION

Cluster-cluster aggregation is a model system for growth by random colloidal aggregation. Two distinct limits have been recognized [1]: diffusion limited (DLCA) in which particles bond irreversibly on contact with unit probability and reaction limited (RLCA) in which the probability of particle-particle bonding is significantly less than unity. In the former case the clusters self-organize into an ordered state due to local depletion of material from their neighborhoods, leading to an effective long-ranged repulsion [2,3]. The final gelling state emerges as the growing clusters merge into a space-filling (or perhaps percolating [4]) network [3]. However, the second case of RLCA is less well understood; we have to date no conceptual framework which seems appropriate. The system evolves, at least in two dimensions (2D), to a state which lacks any characteristic length scale: it has been suggested that it may self-organize into a critical state in the limit $t\rightarrow\infty$ [5]. We advance here the conjecture that this state possesses the symmetries of the critical state, in particular that it exhibits conformal invariance. This property of critical systems implies that at criticality the spatial correlation functions describing the system are asymptotically invariant under conformal transformation [6]. We restrict ourselves to the 2D case, where the conformal group is an infinite-parameter group, giving the symmetry considerably greater scope [6].

In the present context the $t\rightarrow\infty$ limit cannot be interpreted too strictly: any fractal growth must eventually incorporate all the matter present within a single cluster. For an infinite system this will span the system, due to the decreasing density of growing fractal objects. This will also be true of finite systems with a sufficient (system size dependent) particle density to form a spanning cluster. In such a state the system cannot show structures on all length scales as envisaged in the analogy with criticality. We thus consider the trends as t evolves towards the limit. In particular we will consider these trends for the scattering function $S(q,t)$.

There is some conflict in the literature over the form of $S(q,t)$ in RLCA[2,7]. In 2D experiments [5], as in the simulations to be described here, the form tends to a power-law extending to the smallest q values accessible: the system in the final stages of aggregation lacks any characteristic scale of length. Now, conformal invariance follows for systems which exhibit certain properties [6]:

$$\begin{aligned}&(\text{scale invariance})+(\text{translational invariance})\\&\quad+(\text{rotational invariance})\\&\quad+(\text{short-range interactions})\\&\qquad\qquad\Rightarrow(\text{conformal invariance}).\end{aligned}\tag{1}$$

RLCA involves translational, rotational [8], and, in the $t\rightarrow\infty$ limit, scale [5] invariance. Experimentally the residual electrostatic interactions between particles may include a long-range component [9], so we resort here to simulations of cluster-cluster aggregation in which bonding occurs on contact, so that the interactions are indeed short-ranged. It is these features of RLCA which lead us to consider the question of conformal invariance.

II. METHODS

Our simulations involved two-dimensional RLCA on a square 512×512 lattice with periodic boundary conditions. Particles were initially placed at random on lattice sites to achieve the desired number density or area fraction (ϕ). During the simulation clusters were selected at random and moved by one lattice spacing in a randomly chosen direction. If two particles (monomers or incorporated in clusters) arrived in neighboring lattice sites they were bonded irreversibly if a random number ($0\leqslant x\leqslant1$) was less than p, the particle-particle sticking probability. Otherwise they were left in place, free to move apart at later time. After each cluster move time was incremented by $1/N(t)$, where $N(t)$ is

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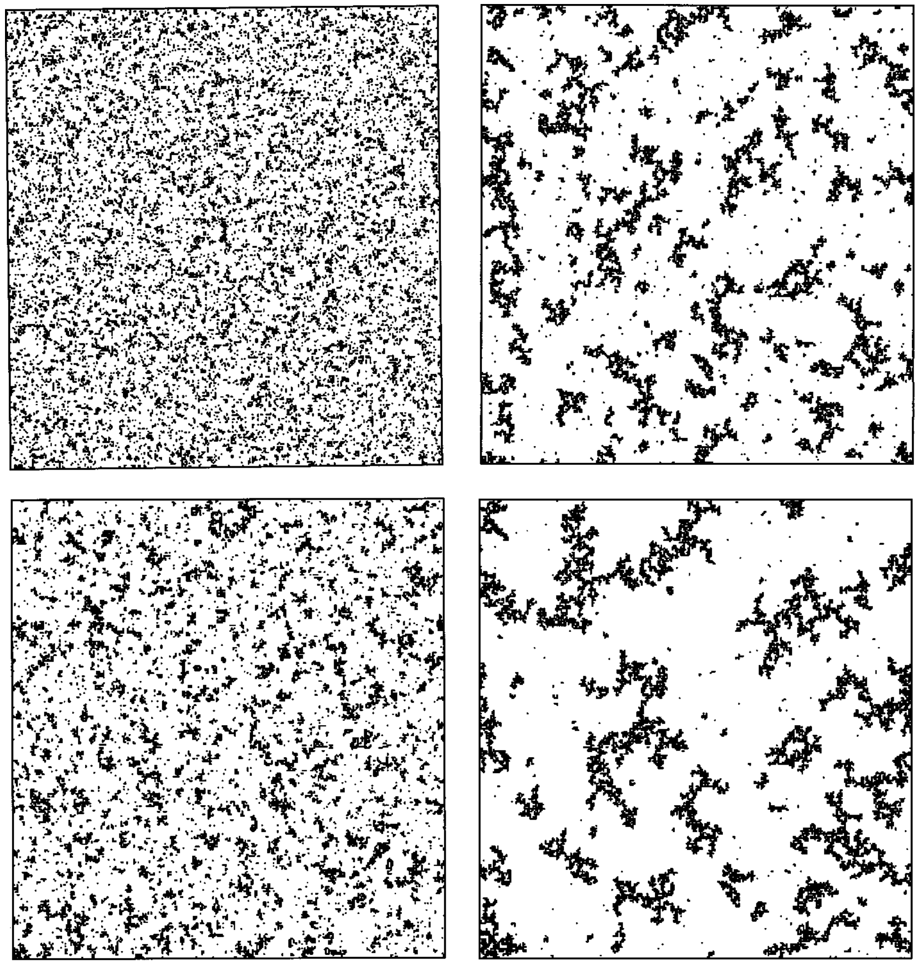

FIG. 1. Selected images ($t=1501$, 4480, 10 652, and 16 566 time steps) from a 512×512 2D RLCA simulation with $p=0.001$ and $\phi=0.1$.

the number of clusters in the system at time t. Thus one time step roughly corresponds to all clusters in the system having moved. Simulations were run until all particles had aggregated to form a single cluster.

In our simulations we used various sticking probabilities and area fractions. The data to be presented all involve $p=0.001$ and $\phi=0.1$, the latter being comparable with experimental values. The evolution of a typical simulation, which ran for a total of 56 174 time steps, is illustrated in Fig. 1.

III. RESULTS AND DISCUSSION

$S(\boldsymbol{q},t)$ was computed at various stages in the aggregation process as

$$S(\boldsymbol{q},t)\propto\left|\int\rho(\boldsymbol{r},t)e^{i\boldsymbol{q}\cdot\boldsymbol{r}}d\boldsymbol{r}\right|^2, \qquad (2)$$

where $\rho(\boldsymbol{q},t)$ is the local density. Circularly averaged data [$S(\boldsymbol{q},t)$ shows no azimuthal structure, demonstrating the rotational invariance mentioned above] are shown in Fig. 5(a) below. At early times a peak is apparent in $S(q,t)$ at $q\approx 2\pi/2\langle R_g\rangle$, where $\langle R_g\rangle$ is the average radius of gyration of the clusters (R_g of a cluster is defined as the trace of its diagonalized inertia tensor). This peak arises from conservation of mass as the growing clusters deplete the local concentration of monomers and small clusters. However, as t increases this peak becomes washed out until eventually in the $t\rightarrow\infty$ limit $S(q,t)$ falls as a power law from the lowest q. In fact, as noted experimentally [5], the $S(q,t\rightarrow\infty)$ limit diverges as q^{-D} as $q\rightarrow 0$, where D ($=1.56\pm0.02$) is consistent with the fractal dimension of RLCA [10]. However, this scaling is *not* associated with the individual clusters, but rather with the entire system: the aggregation leads to self-organization into a state in which the spatial disposition of the clusters is such that the system as a whole lacks any characteristic scale of length.

We have investigated the effects of a conformal transformation upon the simulated system. Such a transform is most clearly described by considering the 2D lattice in the complex plane: $z=x+iy$. Any analytic function $z'=f(z)$ then defines a conformal transform [6]. The effects of various transforms were studied, with essentially identical results. Here we present data for

$$z'=z+\frac{z^2}{10}. \qquad (3)$$

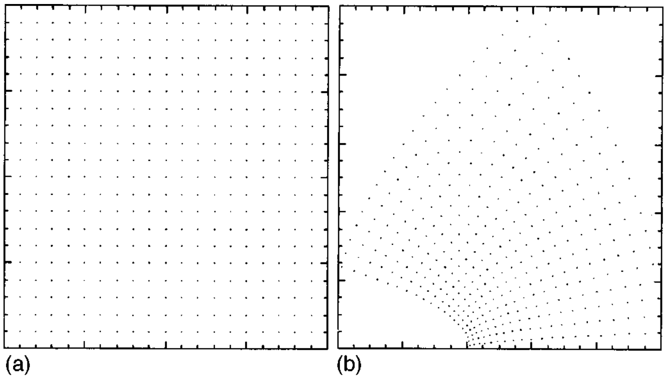

FIG. 2. The effect of the conformal transform of Eq. (3) upon a 20×20 square lattice.

Figure 2 illustrates how this transform *locally* corresponds to a combined translation, rotation, and dilatation, also the local preservation of angles due to the absence of any shear components in $f(z)$.

Figures 3 and 4 show the result of applying the transformation of Eq. (3) to RLCA systems at two different times. While the resultant image in Fig. 3(b) is, apart from the distorted boundary, qualitatively similar to the original, it is not obvious that the conformally transformed system is indeed statistically unchanged (in fact it is not unchanged—see below). However, this image is for an intermediate time, not really at the $t\rightarrow\infty$ limit, so this may not be entirely surprising. The effects upon the system at much later time (Fig. 4) are more convincing. However, such visual inspection can only give a qualitative impression of similarity; we should rather compare the statistical information implicit in correlation functions or (as here) scattering functions.

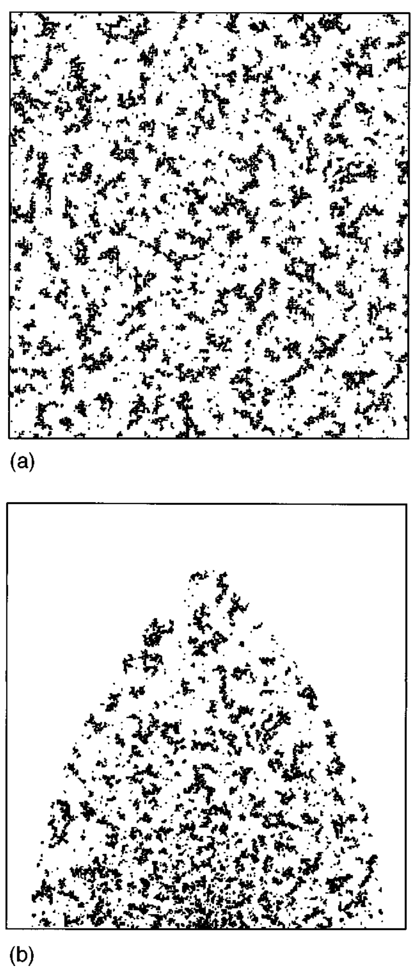

FIG. 3. A typical RLCA image at $t=7686$ time steps (a) and as conformally transformed by Eq. (3) (b).

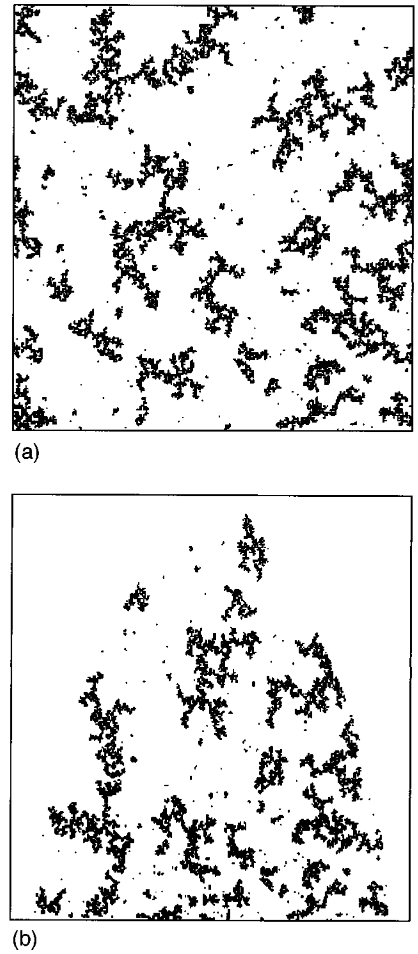

FIG. 4. As Fig. 3, but for $t=16\,566$ time steps.

We thus compare the structure functions $S(q,t)$ for the original and transformed systems (Fig. 5). To permit computation of Fourier transforms of the latter a square section of the transformed image (as large as possible) was interpolated onto a square 512×512 lattice. For early times $S(q,t)$ for the transformed system differs somewhat from that for the original, but as we approach the $t\rightarrow\infty$ limit (e.g., $t=16566$ in the figure) these differences diminish, until finally the two functions are qualitatively very similar indeed. This similarity of $S(q,t)$ is particularly marked at low q, where essentially the same power-law divergence is found, while at large q the transform changes $S(q,t)$ due to the distortion of the underlying lattice. These features reflect the asymptotic nature of conformal invariance.

Thus for RLCA the $q\rightarrow 0$ divergence of $S(q,t)$ as $t\rightarrow\infty$ is unaffected by conformal transformation of the system. Such conformal invariance is not to be expected in DLCA, which self-organizes into a state with a characteristic length scale,

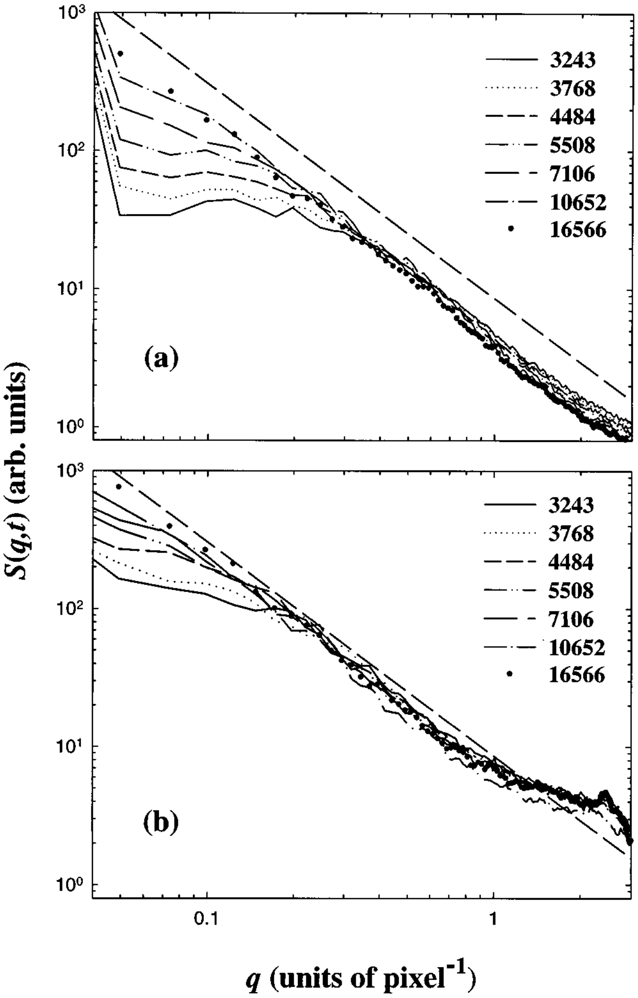

FIG. 5. Structure functions (a) for the original system and (b) for the conformally transformed system (see text for details).

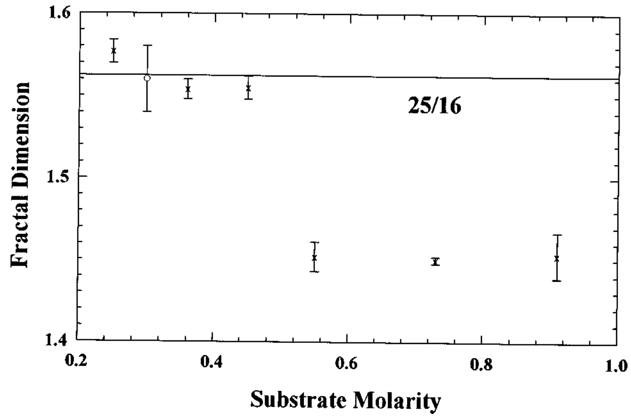

FIG. 6. Fractal dimensions found in 2D cluster-cluster aggregation compared to the predictions of Eq. (3) for $n=5$ (line). Data are for the present simulations (○) and for 2D experiments (×) on aqueous $CaCl_2$ solutions of varying concentrations [13]: note the crossover to DLCA above 0.5M.

so that scale invariance fails [3], and for which (at least in 2D) rotational invariance is broken, as shown by the development of a hexagonal symmetry in $S(\mathbf{q})$[8]. For DLCA the peak in $S(q,t)$ at q_m[3] which reflects the characteristic length scale is destroyed by conformal transformation of the system, as that length scale is altered in a spatially nonunique fashion.

At this point we digress from the main thread to emphasize that this conclusion is independent of the exact transform used. Transforms which just involve global translations, rotations, or dilatations (or any combinations of these) were not investigated, as they are trivial. Transforms which are not one-to-one for all z were used in such a way as to ensure a one-to-one mapping for the x, y domain of the lattice used in the simulations [e.g., for $z'=\sin(z)$, z was redefined to lie $\leqslant \pi/2$]. The transforms studied included $z'=z+z^2/n$, $10\leqslant n\leqslant 50$, $z'=z^2$, $z'=\exp(\pi z/512)$, and $z'=\sin(\pi z/1024)$. It would be tedious to display all of the data. In all cases the results were qualitatively similar to those shown here, quantitative agreement being found for the slope of $S(q,t\rightarrow\infty)$ in the asymptotic limit of low q. The present results suggest universality in terms of the transforms used.

The consistency of RLCA as $t\rightarrow\infty$ with the conjecture of conformal invariance, encourages us to pursue this conjecture. A major success of the application of conformal invariance to the study of critical systems has been the quantitative prediction of exact critical exponents [6]. This has been extended to the prediction of fractal dimensions: Larsson [11] has suggested that percolation in 2D can be treated by conformal invariance, leading to fractal dimensions given by

$$D=\frac{100-n^2}{48}, \tag{4}$$

n being integer. The theory is nonminimal in requiring half-integer labels in the Kac table [11], leading to difficulties in deducing conformal dimensions *ab initio*, but can be deduced from twisted $N=2$ supersymmetry [12], apparently overcoming these difficulties. Here we accept Eq. (4) as it stands. Clearly in 2D we require $0<D<2$, restricting $2\leqslant n\leqslant 9$. Could this be applicable to RLCA? As noted above, any fractal growth process must ultimately lead to a percolating cluster provided the system holds enough monomers. However, the percolation which occurs for DLCA in sufficiently dense systems entails fractal clusters growing into each other [3,4]. But RLCA is quite different, in that in sufficiently dense systems (exact ϕ depending on L) a single system-spanning cluster may arise in the presence of smaller clusters on all length scales so that a prediction based on a percolation model may be appropriate. Indeed it is just this presence of all length scales in RLCA which underlies the attribution of self-organized criticality in this case.

The fractal dimensions observed in our simulations and in experimental studies [13] of 2D RLCA (which agree with literature values) agree essentially exactly with Larsson's predictions for $n=5$: $D=25/16$. Figure 6 illustrates this agreement. [The crossover apparent in the figure as the salt concentration (c) increases through 0.5M corresponds to a change in structure from RLCA-like at low c, where the charge on the experimental particles is only partially screened, to DLCA-like at high c, where the screening is essentially complete [13].] The closest alternatives for D, 1.75 (for $n=4$) and 1.33 ($n=6$), do not correspond at all to the established fractal dimensions for cluster-cluster aggregation. While conformal invariance allows the prediction of scaling exponents, it offers no guidance as to which statistical model they might apply to. While the exact significance of this accord is thus not immediately apparent, it again suggests that RLCA can be associated with conformal invariance. We note that the only other quantitative prediction for cluster-cluster aggregation (apparently for DLCA) $D=1.39$, based on the fixed scale transformation approach [14], is in very much poorer agreement with the accepted value of 1.44 for DLCA.

IV. CONCLUSIONS

We have conjectured that in RLCA a colloidal system self-organizes to a state closely resembling a critical state, which in particular is invariant under conformal transformation. The asymptotic statistical invariance of $S(q,t)$ under conformal transformation of the system as $t\rightarrow\infty$, and the agreement of the fractal dimension of RLCA clusters with predictions based on conformal invariance suggest that this conjecture may be sound. Apart from advancing our understanding of one of the fundamental models for fractal growth, the attractiveness of this conjecture lies in the provision of a theoretical framework within which reaction limited cluster-cluster aggregation can be explored in more detail than heretofore. For example, conformal invariance makes further demands on correlation functions which are testable, and provides a framework for treatment of finite size effects [6]. Again, Eq. (3) offers several fractal dimensions which may be appropriate to different subsets of RLCA, defined perhaps as in the percolation case [15].

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