Tunable Fractal Shapes in Self-Avoiding Polygons and Planar Vesicles

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The shapes of self-avoiding continuum and lattice polygons of N monomers in a plane are studied using Monte Carlo simulations and exact enumeration. To model vesicles, a pressure increment $\Delta p = p_{\rm in} - p_{\rm out}$, is included. For $N \gg 1$ and $\Delta p = 0$, the usual universal fractal shapes appear; but for $\Delta p \neq 0$, *continuously variable* fractal shapes are found controlled by the variable $x \propto \Delta p N^{2\nu}$ where $\nu = 1/D_F = 3/4$. Thus, the ratio of principal radii of gyration $\Sigma(x) = \langle R^2_{G,\min}\rangle / \langle R^2_{G,\max}\rangle$ changes smoothly from $\Sigma(+\infty) = 1$, for circles, through $\Sigma(0) \simeq 0.39$, to $\Sigma(-\infty) \simeq 0.23$, which corresponds to branched polymers.

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Consider a statistical ensemble of geometrical objects, say, clusters, polygons, vesicles, etc., in an isotropic Euclidean space generated by a physical process which respects the isotropy. The overall ensemble-average shape of an object must, likewise, be spatially isotropic. However, as emphasized by Family, Vicsek, and Meakin,[1] the actual *mean shapes* of an object referred, say, to its own principle axes of gyration may be anything but isotropic; indeed, for the asymptotically large fractal clusters arising in branched polymers (or lattice animals), percolation, and growing percolation models in two dimensions, they demonstrated that the shapes were universal, within each class of models. More concretely, if R^2_{G1} and R^2_{G2} are the minimum and maximum eigenvalues of the radius of gyration tensor, Family, Vicsek, and Meakin[1] estimated the anisotropy ratios $\tilde{\Sigma} = \langle R^2_{G1}/R^2_{G2}\rangle$ obtaining distinct values for each problem. Thus the value of $\tilde{\Sigma}$, like the exponent ν, or fractal dimension $D_F \equiv 1/\nu$, etc., characterizes the universality class and related renormalization-group fixed point.

Following Family, Vicsek, and Meakin ϵ expansions[2] and exact lattice enumerations[3] have been performed for the shapes of percolation clusters and branched animals. In addition to the anisotropy $\tilde{\Sigma}$, defined above, the asphericity $\Delta = \langle (R^2_{G1} - R^2_{G2})^2\rangle / \langle (R^2_{G1} + R^2_{G2})^2\rangle$ has been utilized.[4] For polymer chains, modeled as random walks or, better, as *self-avoiding walks,* the question of shape has a long history[5,6] and corresponding universal shape ratios have recently been calculated by Monte Carlo simulations[6] and by analytical means[7,8] both for chains and for rings or *polygons.*

Here we ask the following: "Within a given universality class, such as self-avoiding walks, how invariant actually are the associated fractal shapes?" This question arises rather naturally from a study of the statistical mechanics of two-dimensional *vesicles* by Leibler, Singh, and Fisher[9] (LSF). LSF model vesicles as closed loops of N hard *beads* (disks) of diameter a linked by loose bonds or "tethers"[10] of length $l_0 = \frac{9}{5} a$, from bead center to center. An "osmotic pressure" difference,

$$\Delta p = p_{\rm in} - p_{\rm out} \equiv \bar{p} k_B T / a^2 , \tag{1}$$

is introduced[9] with an associated Boltzmann factor $e^{\bar{p}A}$, where A is the vesicle area. A rigidity κ also played a significant role for LSF whose main focus concerned the characteristic, *nonfractal, cytotype shapes* arising from the interplay of *deflation* ($\Delta p < 0$) and *large* rigidity. However, an extensive fractal region, evidently controlled by the self-avoiding-walk fixed point with size exponent[11] $\nu = \frac{3}{4}$, was observed as $\bar{p}$ varied with $\kappa = 0$ (or κ fixed and $N \gg 1$). For large *inflations* ($\bar{p} > 0$) the typical shapes generated in the Monte Carlo simulations[9] appeared circular; for large *deflations* the closed loops or polygons collapsed into configurations resembling branched polymers. This identification was reinforced by the size dependence in the limit $\bar{p} \to -\infty$ which corresponded to[9,12] $\nu^- \simeq 0.641$.

How do these changes of shape take place as $\bar{p}$ varies? For $\Delta p = 0$ we should observe shape parameters in accord with previous results for self-avoiding rings.[6] Indeed we estimate,[4,13] for $N \to \infty$,

$$\Sigma_0 = \langle R^2_{G1}\rangle / \langle R^2_{G2}\rangle \,|_{\bar{p}=0} = 0.393 \pm 15 , \tag{2}$$

fully consonant with the $\Sigma_0 = 0.405 \pm 9$ found for $N = 32$ and larger values observed for $N < 32$.[6] However, when Δp becomes positive and $N \to \infty$, does Σ first stick at the value Σ_0 only ultimately jumping discontinuously to $\Sigma^+ = 1$ (for circles) at $\bar{p} = +\infty$? Or does Σ switch immediately to the value 1 as soon as Δp exceeds 0? See the dash-dotted and dotted lines in Fig. 1. The analogous questions arise when Δp becomes negative with, now, the branched polymer values, which we estimate as[4,13]

$$\Sigma^- = 0.230 \pm 10 \quad \text{or} \quad \tilde{\Sigma}^- = 0.275 \pm 10 , \tag{3}$$

being expected for $\bar{p} = -\infty$. (The Σ^- value is new; $\tilde{\Sigma}^-$ accords with previous studies yielding[1] 0.29 and[3] 0.280 ± 3.)

Alternatively, can the fractal vesicle shapes vary *continuously* as $\bar{p}$ is changed? We argue here that this last possibility is actually realized as $N \to \infty$ *provided* Σ, and the other shape parameters,[4] are examined on the scale

$$x = D\bar{p} N^{\varphi\nu} \text{ with } \varphi = 2 , \tag{4}$$

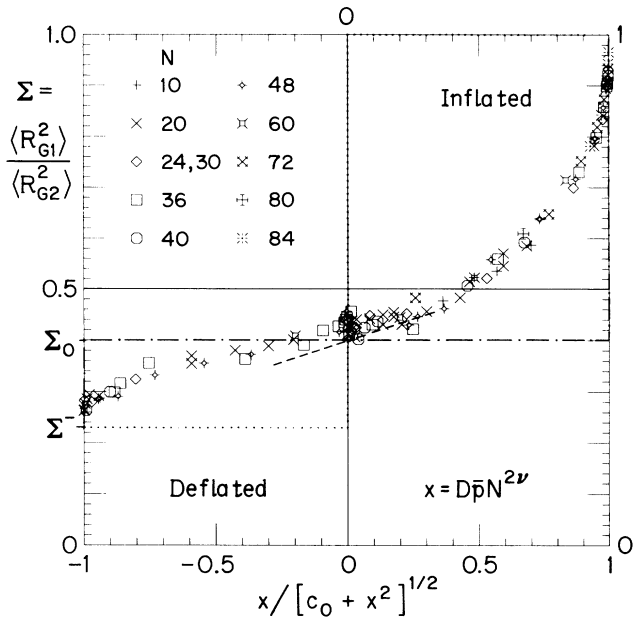

FIG. 1. Variation of the shape parameter Σ with the scaled variable x on a nonlinear scale set by $c_0 = 0.30$. Note that Σ_0 and Σ^- represent asymptotic ($N \to \infty$) values for self-avoiding ring polymers and branched polymers, respectively; $\Sigma^+ \equiv 1$ describes circles.

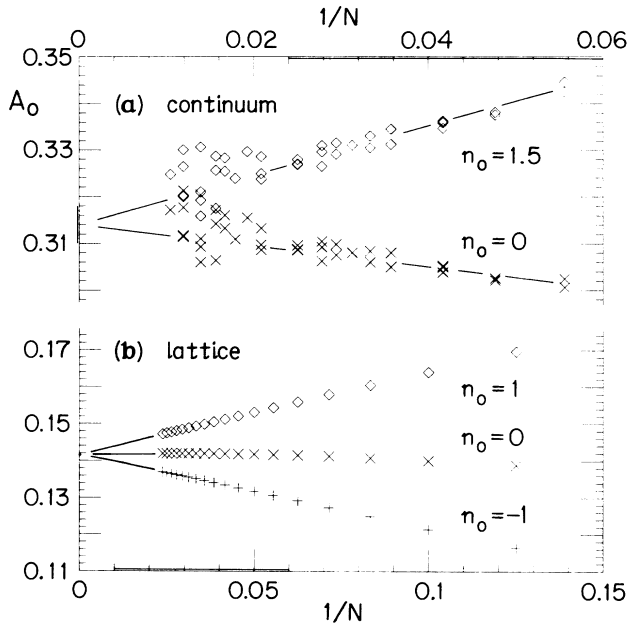

FIG. 2. Estimates $A_{0,N} = \langle A \rangle_N / (N - n_0)^{2\nu}$ with $\nu = \frac{3}{4}$, for the area amplitude A_0 for continuum and lattice polygons (in units of a^2, at $\Delta p = 0$). The lines indicate reasonable extrapolations to $1/N = 0$.

where D is a suitable nonuniversal metrical factor. Monte Carlo simulations and exact series data[14] confirm this proposition. Thus we have vesicles (or "pressurized polygons") with *tunable fractal shapes* all of which, however, have the *same* fractal dimension $D_F = 1/\nu = 4/3$ (at fixed x). Figure 1 portrays the variation of Σ with x; as $x \to \pm\infty$ we find that specific power laws characterize $\Sigma(x)$.

More generally, this type of continuous variation of fractal shapes at fixed fractal dimension should apply to many analogous problems where there exists a natural relevant variable similar to p, e.g., near-critical Ising-model clusters, appropriately defined,[15] with a magnetic field allowed for. However, the crossover exponent $\phi \equiv \varphi\nu$ may not always be simply related to other exponents as in (4).

To justify these conclusions,[16] recall first that LSF[9] studied the mean area $\langle A \rangle$ and the radius of gyration $\langle R_G^2 \rangle = \langle R_{G1}^2 \rangle + \langle R_{G2}^2 \rangle$ and verified the scaling laws

$$\langle A \rangle \approx A_0 N^{2\nu_A} Y(x), \quad \langle R_G^2 \rangle \approx R_0^2 N^{2\nu} X(x), \tag{5}$$

with $\nu_A = \nu$,[17] where we have adopted the normalizations $Y(0) = X(0) = 1$. Further, since the scaling functions should be *analytic* for small x, we may define D in (4) by setting $Y_1 \equiv (dY/dx)_0 = 1$. Then all other derivatives $Y_2, \ldots, X_1, X_2, \ldots$ should be *universal*.

Actually LSF found $2\nu_A \simeq 1.52$, $2\nu \simeq 1.51$, and $\varphi \simeq 2.13$. We attribute the deviations from[17] $\nu_A = \nu = \frac{3}{4}$ and $\varphi = 2$ (which follows[9] from A scaling as $R_G^2 \sim N^{2\nu}$) mainly due to finite-size effects ($N \lesssim 100$). Indeed, the amplitude estimates $A_{0,N} \equiv \langle A \rangle_0 / N^{2\nu}$, etc., should vary as $1 + c_\theta / N^\theta + c_1 / N + \cdots$. If the leading correction exponent satisfies $\theta \gtrsim 1$, or if c_θ is relatively small, the dominant corrections can be effectively gauged in plots versus $1/N$ by replacing N by $N - n_0$ for various choices of "n shift," n_0. Experience with series extrapolations for lattice critical phenomena suggests that small values of n_0 ($\simeq \pm 1$) should suffice for first cumulants of the underlying distributions while successively larger values are needed for higher cumulants. Surprisingly, perhaps, we find closely similar behavior for our continuum simulations (which extend LSF); see, e.g., Fig. 2 which compares the estimation of A_0 for continuum and lattice models.[14] Note that lower statistical precision at large N (aggravated by longer equilibration times[9]) tends to cancel the desirable advantage of small $1/N$. Estimation of R_0^2, Σ_0, etc., proceeds similarly.[18]

For a square lattice (of spacing a) and for the continuum model we find $A_0/a^2 = 0.1415 \pm 3$ and 0.314 ± 3, respectively, and

$$\tilde{\pi} \equiv A_0 / R_0^2 = 2.52 \pm 4 \text{ and } 2.55 \pm 5, \tag{6}$$

which exemplifies the anticipated universality for $\Delta p = 0$.[18] Of course, $\tilde{\pi} = \pi$ for circles but, in fact, aligned but *superposed* vesicle configurations, as presented by LSF, yield a fuzzy, roughly elliptical "cloud," less dense at the center, with semiaxes in the ratio $\bar{a}/\bar{b} \simeq 0.64$. Notice that $(0.64)^2 \simeq 0.41$ which agrees, somewhat surpris-

ingly, with the corresponding anisotropy estimate (2).

Taking $\Delta p \neq 0$ and performing further simulations[19] enables us to check the proposed shape scaling relation

$$\Sigma_{\bar{p},N} \approx \Sigma(x) = \Sigma_0(1 + W_1 x + \cdots). \quad (7)$$

The good data collapse for $N = 10$–84 seen in Fig. 1 supports the hypothesis. The estimation of the scaling function $\Sigma(x)$ is improved by extrapolation to $1/N = 0$ at fixed x (as in Fig. 2). For $x \lesssim 0$ merely using the values $n_0 \simeq -0.4$ and 0.7, found optimal for $\langle R_{G1}^2 \rangle_N$ and $\langle R_{G2}^2 \rangle_N$, sharpens the collapse significantly; for large positive x, however, bigger values of n_0 are needed. If (7) is valid, the universal amplitude W_1 can be estimated by extrapolating data for $\Delta p = 0$ via

$$D\Sigma_0 W_1 \approx \frac{1}{N^{2\nu}} \left(\frac{\partial}{\partial \bar{p}} \Sigma \right) \Bigg|_{\bar{p}_0}$$

$$\approx \frac{\langle R_{G1}^2 A \rangle_0 \langle R_{G2}^2 \rangle_0 - \langle R_{G2}^2 A \rangle_0 \langle R_{G1}^2 \rangle_0}{(N - n_0)^{2\nu} \langle R_{G2}^2 \rangle_0^2}, \quad (8)$$

as shown in Fig. 3, and similarly for DW_1. The fairly well defined finite value indicated by Fig. 3 supports the scaling hypothesis. By making analogous estimates for Y_1 ($=1$) and X_1 [see Eq. (5) *et seq.*] we find $D = 0.0175 \pm 15$ and then $X_1 = 0.65 \pm 20$ and $W_1 = 0.78 \pm 10$. Crosschecks on the value of W_1 (and Σ_0) follow from estimates for the amplitudes $R_{1,0}^2$, $R_{2,0}^2$, and scaling function derivatives $X_{1,1} = 1.21 \pm 12$ and $X_{2,1} = 0.41 \pm 6$, for $\langle R_{G1}^2 \rangle$ and $\langle R_{G2}^2 \rangle$. The slope corresponding to W_1 is shown as a dashed line in Fig. 1; it agrees well with what might be concluded by interpolating for small x ($\simeq \pm 0.2$).

Note that when $N \to \infty$ at fixed $\bar{p} \neq 0$, the scaling (7) implies that vesicles should rapidly inflate to circular form for $\bar{p} > 0$ or deflate to branched polymers for $\bar{p} < 0$. *In this sense* the shape change may, indeed, be considered asymptotically as a discontinuous function of Δp (as originally suggested by the dotted lines in Fig. 1). More specifically, however, it follows that the limiting behavior of $\Sigma(x)$ when $x \to -\infty$ should be controlled by the correction-to-scaling exponent, $\theta^- = 0.87 \pm 7 < 1$, for branched polymers.[20] This conclusion can be checked by plotting Σ (and $\tilde{\Sigma}$, Δ, and $\tilde{\Delta}$) (Ref. 4) versus $x^{-\theta^-/2\nu} \sim N^{-\theta^-}$. On discounting data points for $x < -5.6$, since equilibration is inadequately fast for large $|\bar{p}|$, linear behavior is found. Indeed the expression

$$\Sigma(x) \approx \Sigma^{\pm}(1 \mp W_1^{\pm} / |x|^{\psi^{\pm}}) \quad (x \gtrless 0), \quad (9)$$

with $\psi^- = \theta^-/2\nu \simeq 0.58$ and $W_1^- \simeq 0.36$, provides a good fit in the deflated regime up to $x \lesssim -\frac{1}{2}$.

In the *inflated* regime a vesicle becomes close to a circle of mean radius $\bar{R}(N,\bar{p})$ but with transverse (radial) fluctuations, $u(\theta)$. Then the shape ratio will vary as $\Sigma \approx (\bar{R} - c\Delta R)^2/(\bar{R} + c\Delta R)^2 \approx 1 - 4c(\Delta R/\bar{R})$ when $\bar{R} \to \infty$, where $\Delta R = \langle u^2 \rangle^{1/2}$ and $c = O(1)$. To estimate $\langle u^2 \rangle$ we regard the vesicle as having a surface tension γ related to the pressure, as usual, via $\Delta p = (d-1)\gamma/\bar{R}$ (with $d = 2$ here). Now the low-amplitude transverse thermal vibrations of a string of length L ($= 2\pi\bar{R}$) under a tension γ obey $\langle u^2 \rangle = k_B T L/\gamma \sim k_B T/\Delta p$.[21] Thus we expect $1 - \Sigma \sim (k_B T/\Delta p \bar{R}^2)^{1/2}$. Last, one can show[22] that $\bar{R}$ varies as $\bar{p}^{\omega} N^{\nu^+}$ in the inflated regime with $\omega = (1-\nu)/(2\nu - 1) = 1/2$ and $\nu^+ = \nu/(2\nu - 1) = 3/2$. Substitution yields (9) for $x \to +\infty$ with $\psi^+ = 1/2 \times (2\nu - 1) = 1$. The Monte Carlo data (up to $N \simeq 150$) confirm this; indeed, (9) with $W_1^+ \simeq 0.32$ fits well.[23]

One may also enquire as to how $\tilde{\pi}(x)$ [see (6)] approaches π when $x \to \infty$. For near-circular vesicles one has $1 - \tilde{\pi}/\pi \approx 2\langle u^2 \rangle/\bar{R}^2 \sim x^{-2\psi^+}$. The data for $x > 1$ are quite consistent with this predicted x^{-2} approach (although a decay as slow as $x^{-1.8}$ cannot be excluded).

Finally, we note that the rigidity κ is an irrelevant variable at the self-avoiding-walk fixed point. Thus our results for $\Sigma(x)$, which should be a *universal* function, are independent of κ provided $l_\kappa \equiv \kappa/k_B T \ll Na$; only the metrical factor D will change with κ. Conversely, for $l_\kappa \gtrsim Na$ one enters the regime of *nonfractal,* cytotype shapes.[9,16]

In conclusion, we have shown that the fractal shapes of two-dimensional vesicles or polygons of fixed fractal dimension $D_F = \frac{4}{3}$ can be tuned continuously from circular to branched polymeric by varying the pressure differential Δp on a scale $x/N^{3/2}$. The universal scaling function $\Sigma(x)$ for the shape ratio $\langle R_{G1}^2 \rangle / \langle R_{G2}^2 \rangle$ has been estimated numerically for all x. When $x \to \pm\infty$, $\Sigma(x)$ exhibits characteristic power laws; for $x < 0$ the exponent is determined by the correction-to-scaling exponent for branched polymers; for $x > 0$ one has $1 - \Sigma \sim 1/x$.

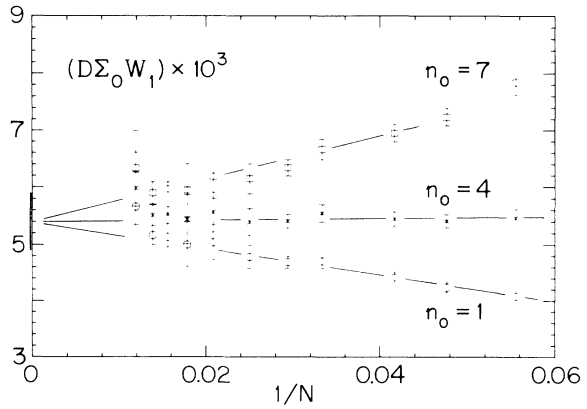

FIG. 3. Monte Carlo estimates of the parameter $D\Sigma_0 W_1$ for continuum polygons; this determines the slope of the universal shape scaling function $\Sigma(x)$ at $x = 0$.

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[13]Uncertainties quoted refer to the last decimal place of the central estimate. To Eq. (2) we may add $\tilde{\Sigma}_0=0.425\pm 15$ with associated dispersion $\sigma(\tilde{\Sigma}_0)=0.185\pm 5$, $\Delta_0=0.220\pm 10$, $\tilde{\Delta}_0=0.215\pm 10$ with $\sigma(\tilde{\Delta}_0)=0.155\pm 5$; and to Eq. (3), for branched polymers, $\sigma(\tilde{\Sigma}^-)=0.180\pm 8$, $\Delta^-=0.463\pm 15$ [compared with 0.466 ± 2 from series (Ref. 4)], and $\tilde{\Delta}^-=0.380\pm 10$ with $\sigma(\tilde{\Delta}^-)=0.22\pm 1$. As noted by Family, Vicsek, and Meakin (Ref. 1) for $\tilde{\Delta}^-$, the dispersions vary remarkably little with N.

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[18]We also estimated the Kuhn amplitude r_0^2 for the end-to-end distance squared $\langle R_N^2\rangle$ for open self-avoiding lattice and continuum chains obtaining $R_0/r_0=0.273\pm 3$ and 0.27 ± 1, further confirming lattice-continuum universality. For open chains we also observed $\langle R_G^2\rangle/\langle R_N^2\rangle=0.141\pm 10$ in agreement with 0.140 ± 1 obtained via exact enumeration by C. Domb and F. T. Hioe, J. Chem. Phys. **51**, 223 (1969).

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[23]If (9) is rewritten for $x>0$ as $\Sigma\simeq 1/(1+W_1^+/x^{\psi^+})$, the fit is good down to $x\gtrsim\frac{1}{2}$.