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"Fluctuating cluster" equations for a polymer to $O(\epsilon^2)$. (Toward exact Flory equation?)

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Abstract. — Although Flory's approximation describes very well the radius $r$ of a polymer, it disagrees sharply with an exact $\epsilon$ expansion (derived from an analogy to $n = 0$ spins). Here the polymer is described explicitly as a critical cluster of $n = 0$ spins. While ordinary clusters resemble trees bifurcating into $b$ branches, a cluster of $n = 0$ spins constitutes a single line, weighted by $b$ phantom bifurcations. Analogous description of the polymer leads to a modified Flory-like equation, which describes $r$ and $b$, jointly, and to two more equations for $r$ and $b$ in separate. These give very accurate exponents $\nu$ and $\gamma$, correct to $O(\epsilon^2)$. In addition, the approach can be extended to many polymer systems, and to other critical clusters.

1. Introduction.

The average radius $r$ of a polymer of length $l$ (number of monomers), is expanded by excluded volume and contracted by a (neg)entropy of stretching. Flory's equation (FE) describes the free energy as a sum of two corresponding terms, $F/T = F_{ev} + F_{str}$, where in dimension $d$, $F_{ev} = l^d/r^d$ and $F_{str} = r^2/l$ [1]. From $dF/dr = 0$, follows $F_{str}/F_{ev} = Cte$, which gives at once the scaling exponent $\nu$ in $r \approx l^{\nu}$. The result for $\nu$ is quite accurate at $d = 3$ and exact at $d = 1, 2$ and 4. (The latter constitutes the upper critical dimension, $d_c$ at which $\nu$ attains the classical value of 1/2). Yet an analogy discovered by de Gennes, between the polymer to a critical system of magnetic spins of $n \rightarrow 0$ vector components, demonstrated conclusively that FE is approximate [2]. Specifically it disagrees in $d = 4 - \epsilon$ with an exact expansion for the $n = 0$ spins, to $O(\epsilon)$ already. de Gennes attributes this failure to that FE constitutes a meanfield approximation. He also points out that $F_{str}$ and $F_{ev}$ constitute each, a gross overestimate of the elastic and excluded volume energies in a real polymer [3]. The amazingly good performance he attributes to a mutual cancellation of gross errors. But such critique seems to underestimate an inherent strength of the FE approach. This claim is made out here in some detail, because it guides the subsequent study of the polymer problem. Thus, regular meanfield neglects the correlation between fluctuations. Yet FE applies meanfield to a single polymer molecule. According to
the present approach, the polymer-$n = 0$ spin analogy implies that the polymer corresponds
to a critical cluster (CC) of such spins. A CC describes (precisely [4, 5]), an instantaneously
isolated local fluctuation. Because of that we can expect the meanfield approximation of FE to
be much less severe; indeed it does yield good and nonclassical values of $\nu$. As for the mutual
cancelation of gross errors, it seems more than unlikely to occur at all $d$! Also very significant
is the fact that FE is exact at $d = 2$. We should therefore look for a reason why the errors are
not so gross after all: thus again, $F_{\text{str}}$ and $F_{\text{ev}}$ apply to an isolated fluctuation. Furthermore,
they can be regarded as representing losses of entropy, due to chain stretching and to excluded
volume, as compared to the entropy of a purely random fluctuation (viz. they do not purport
to estimate the $ev$ and elastic energies of a real polymer). Because of that we shall act on the
premise that FE constitutes a sound starting point and may be improved very significantly
and in a relatively simple way, by letting the meanfield parameters of the isolated fluctuation,
fluctuate in a correlated manner. The present article describes such a remedial treatment. The
main assumptions are first, that the polymer is analogous to a critical cluster of $n = 0$ spins,
which is linear but nonetheless weighted statistically by a phantom bifurcation $b$. Second, the
several correlations that describe the separate and joint fluctuation of $r$ and $b$, assumedly obey
a universality. The latter is utilized in order to relate one correlation to another. The result is
a set of “fluctuating cluster equations” (including a revised FE), which gives exponents $\nu$ and
$\gamma$. The approximation is accurate to $O(\epsilon^2)$, very good at $d = 3$ and exact (like FE) at $d = 2$.

2. FE in terms of correlation.

To begin with let’s rewrite FE. An instantaneous radius is $r' = \sum_{i=1}^{l} \delta r_i$, where $\langle \delta r_i \rangle = 0$. Hence

$$r^2/l = l^{-1} \sum_{i,j} \langle \delta r_i \delta r_j \rangle = \xi_r$$

(1)

Ideally $r^2/l \approx 1$. Correlation $\xi_r$ causes non-ideal expansion. A corresponding exponent $\kappa \equiv 2\nu - 1$, is defined by $\xi_r \approx l^\kappa$. Flory’s $F_{\text{str}}$ is proportional to $r^2/l = \xi_r$. $F_{\text{ev}}$ however depends
upon $1/r^2 = \xi_{1/r}$, where $\xi_{1/r}$ is a correlation factor for reciprocal $r$. As is well-known, $\langle r^{-2} \rangle \approx (\langle r^2 \rangle)^{-1}$ (except for $r \to 0$). Hence $\xi_{1/r} \approx \xi_r^{-1}$ $F_{\text{str}}/F_{\text{ev}} = C_t e$ becomes

$$\xi_r e^{-d/2} \approx \xi_r^{1+d/2} \approx l^{2-d/2}$$

(2)

Equation (2), viz. rewritten FE, gives $\kappa_{\text{flory}} = (4 - d)/(d + 2)$. In $d = 4 - \epsilon$, $\kappa_{\text{flory}} = \epsilon/6+..$ This however disagrees with the formal analogy between polymers to magnetic spins of $n \to 0$
vector components [2], which gives exactly $\kappa = \epsilon/8+..$

3. Phantom bifurcation.

We adopt FE as a starting approximation and, in order to improve it, construct an explicit
analogy between the polymer and a critical cluster (CC) of $n = 0$ spins. Take first the $n = 1$
case, of up or down oriented Ising spins. A CC (which represents an isolated fluctuation),
consists of similar spins, mutually connected with a probability defined by their pairwise in-
teraction [4, 5]. The connectedness may be accorded a tree-like description [6, 7]: a CC of a
given size $s$, is reconstructed (starting from any point on it), by connecting its spins one to
another in a succession of $s$ steps. The number of continuations at an $i$’th spin is $1 + \delta b_i$. Here
$\delta b_i = -1$, 0 and $> 0$, imply, respectively, that the sequence (branch), terminates, continues
linearly, or bifurcates. For CC’s which may grow indefinitely but without blowup, we have \( \langle \delta b_i \rangle = 0 \) (asymptotically), and

\[
s^{-1} b^2 = s^{-1} \sum_{i \geq j} \langle \delta b_i \delta b_j \rangle = \xi_b \approx s^\Delta
\]

(3)

Here \( \Delta \) is a bifurcation exponent. Since \( b \) is the number of non-terminated branches (after \( s \) steps), their length \( l \) obeys \( l \approx s/b \). [True, part of \( s \) is “missing” because it belongs to branches that terminate at \( l' < l \). But an equivalent part of \( s \) is “saved”, because the terminated branches are precisely replaced by new ones (\( \langle \delta b_i \rangle = 0 \)), which start only at equivalent lengths \( l' < l \)]. Hence

\[
b \approx s^{(1+\Delta)/2} \implies l \approx s^{(1-\Delta)/2}
\]

(4)

The CC’s nonlinearity is due to a locally fluctuating number of potential continuations of a branch. There are two causes. Thermal, the concentration of similar spins fluctuates; geometric, branches form loops. We now return to our \( n = 0 \) spins, which point to “nowhere”, and are therefore uniformly similar. But since they interact, one can still define a CC, just like in the \( n = 1 \) case. Does the CC locally fluctuate? Thermal cause dissappears, but the geometric remains. Hence a linear CC reconstructed in \( s \) steps accumulates \( b \) phantom bifurcations, representing a statistical weight. “Phantom”, because a bifurcation into real mutually interacting branches would imply, erroneously, that the concentration of similar spins fluctuates. The distribution consists of CC’s of a fixed generating size \( s \), and variable \( l' \) and \( b' \) (\( l'b' = s \)). Alternatively, it consists of CC’s of a fixed length, weighted by a variable \( b' = s'/l \). These CC’s precisely will be used to describe polymers.

3.1 Relationships \( \Delta \) to \( \gamma \) and \( \gamma \) to \( \nu \). — The polymer of length \( l \), weighted by an average phantom bifurcation \( b \), can be related to a familiar concept. We identify \( b \) with the number of polymer configurations in standard theory, \( b = W_i \approx l^\gamma \). It follows at once from equation (4),

\[
\gamma - 1 = 2\Delta/(1 - \Delta)
\]

(5)

We shall now relate \( \Delta \) to \( \kappa \). This and following scaling estimates, are based on an assumption of universality of correlations (which has been already applied to CC’s of \( n = 1 \) spins [7]). It postulates that it is immaterial what pairs of random variables cause a particular correlation; e.g. whether \( \delta r_i \delta r_j \) in \( \xi_r \), or \( \delta b_i \delta b_j \) in \( \xi_b \). The correlations do depend however on the effective number of participating pairs. Thus, all \( \delta r_i \delta r_j \) contribute to the summation over \( i; j = (1, l) \) in equation (1). But not all \( \delta b_i \delta b_j \) contribute to the summation over \( i; j = (1, s) \) in equation (3). The \( b \) phantom branches do not interact mutually because they “live in a nowhere land” [8]. Consequently only pairs such that \( i \) and \( j \) belong to the same branch, contribute. They constitute a fraction \( b^{-1} \) of the total number of pairs. Therefore, the double sum has to be multiplied by \( b^2 \), to make it equivalent to that in equation (1). Or, \( (b^2)^\Delta \approx l^\kappa \), whence (by Eq. (4)), \( 2\Delta(1 + \Delta)/(1 - \Delta) = \kappa \). Substituting \( \kappa = 2\nu - 1 \) and \( \gamma - 1 = 2\Delta/(1 - \Delta) \) (Eq. (5)) we get,

\[
(2\nu - 1)/(\gamma - 1) = 1 + \Delta \approx 1 + (2\nu - 1)/2
\]

(6)

which agrees with the \( e \)-expansion to \( O(e^2) \). Thus, we have derived a novel approximation, relating \( \nu \) and \( \gamma \) (and ipso facto \( \Delta \)). In order to find all three exponents in separate, we return to FE.
3.2 Fluctuating Flory Equation (FFE). — The left hand side of equation (2), is interpreted as an average, of a product of fluctuating correlations viz. \(\langle x_i^2 (x_i^1)^{-d}\rangle_s\), for a CC of fixed generating size \(s\). This is at first approximated by a product of averages \(\langle x_i^2 \rangle_s \langle (x_i^1)^{-d}\rangle_s\). It is also more convenient to consider polymers of fixed \(l\) and a variable number of branches \(b' = s'/l\). A given \(r^2\) reappears \(b'\) times; viz. \(\langle x_i^2 \rangle_s \equiv \langle r^2/l'\rangle_s \approx \langle (b'^2 r^2)/s'\rangle_i\). Hence

\[
\langle x_i^2 \rangle_s \approx \langle s'^{-1} \sum_{i \geq 1} \sum_{m \geq n} \delta r_i \delta r_j \delta b_m \delta b_n \rangle \approx \xi_r \xi_{r,b}
\]

Similarly \(\langle x_i^2 \rangle_s \approx \xi_1 \xi_{1/r} \xi_{1/r,b}\). Cross-factor \(\xi_{r,b}\) is marked by a "tilde", while \(\xi_1 / r \) is not, to indicate that the first and second represent, respectively, an anti-correlation and a correlation. The reason is as follows. The two respectively, are coupled to the two deviations from an ideal ground state, \(\xi_r\) and \(\xi_{1/r}\). In a stable system, each of these cross-factors should damp an increase of its coupled deviation, by a simultaneous decrease of the statistical weight \(b\). Hence \(\xi_r\) and \(\xi_{1/r}\) are indeed anti-correlated. By the same token \(\xi_1 / r \) and \(\xi_{1/r,b}\) are anti-correlated too; however since \(\xi_1 / r \approx \xi^{-1}\) (see Eq. (2)), \(\xi_r\) and \(\xi_{1/r,b}\) are instead correlated. Extending the hypothesis of universal scaling of correlations to the present case, we assume \(\xi_r \approx \xi_{1/r,b}\) \(\approx (\xi_{r,b})^{-1} \approx l^d\). The net effect of the two opposing tendencies determines a total cross-factor \(Y_{cross}\) in FFE,

\[
(\xi_1 / r \xi_{1/r,b}[= Y_{cross}) = \xi_r^{d/2-1}
\]

Equation (2) revised with the help of equation (7), and equation (8) lead to

\[
\xi_r^{d/2+1} Y_{cross} \approx \xi_r^d \approx l^d \approx \xi^{d/2}
\]

giving \(\kappa_1 = (4 - d)/2d\). We note that our FFE, differs from FE in the inclusion of \(Y_{cross}\), which increases the combined power of correlations from \(d/2 + 1\), to \(d\). But due to that FFE becomes very plausible: the net result of excluded volume, viz. a product of \(d\) reciprocal correlations \(\xi_i^{-d}\), is proportional to (square root of) the number of random increments causing each of them in separate, raised to power \(d\), viz. \(\propto l^{d/2}\), the latter being normalized by \(l^{d/2}\) at which correlations vanish. This provides a weighty support to our derivation of \(Y_{cross}\); the unmodified FE for example equates in an inconsistent manner \(\xi_i^{d/2+1}\) to \(l^{d/2}\). Another way of looking, is that our approach determines \(d_c = 4\)! At \(d = 2\), the power \(d/2 - 1\) vanishes and \(Y_{cross} = 1\) (Eq. (8)). Consequently our FFE reverts to FE and both alike yield the exact result [9]. Since the dependence of \(F_{EV}/F_{EV} = Cte\) upon \(d\) is the same for all values of \(n\), the result \(Y_{cross} = 1\) at \(d = 2\), should hold for the \(n\)-vector model in general. It is intriguing to speculate that this may be the cause of the relative simplicity of the case \(d = 2\), which manifests itself in the existence of exact solutions at all \(n\) [9]. At \(d = 1, b \equiv 1\), hence \(Y_{cross} = 1\) obtains trivially. A next approximation returns to that, actually FFE considers an average of a product of instantaneous correlations (cf. before Eq. (7)), \(\langle x_i^2 (x_i^1)^{-d}\rangle_s \approx \langle (x_i^2)^{d/2+1} \rangle_l \approx \xi_r^{d/2+1} Y_{cross}\) \(\approx \xi_r^{d/2+1} \xi_0^{d/2+1} Y_{cross}\). But pairs that are thus already correlated, are not effective in the product. To estimate that we utilize again the trick of equation (6) and write \(l_{eff}(Y_{cross})^{d_{tot}} \approx \rho_{eff}(\Delta \kappa) \approx l_{eff}/l \approx l^{-1/(1 + \Delta \kappa)^{-1}}\) Here \(\Delta \kappa = d_{tot}(d/2 - 1)\kappa_1\) (cf. Eq. (8)). More precisely, since the division by 1 + \(\Delta \kappa\) corrects \(\Delta \kappa\) itself, the divisor is a continuous fraction, viz. \(\Delta \kappa = [d_{tot}(d/2 - 1)\kappa_1]/[(1 + \Delta \kappa)/(1 + ..)]\). Introducing the result into equation (9), we write

\[
[l_{eff}/l]^{1/d} \xi_r^{d/2} \approx l^{\kappa_1 d}
\]
Table I. — Non-ideality exponent $\kappa(= 2\nu - 1)$.

<table>
<thead>
<tr>
<th>$d$</th>
<th>$\kappa_{\text{flory}}$</th>
<th>$\kappa_1$</th>
<th>$\kappa_2$</th>
<th>$\epsilon - \text{expansion}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 - $\epsilon$</td>
<td>$\epsilon/6$</td>
<td>$\epsilon/8$</td>
<td>$3.75\epsilon^2/64$</td>
<td>$\epsilon/8 + 3.75\epsilon^2/64$</td>
</tr>
<tr>
<td>2</td>
<td>1/5</td>
<td>1/6</td>
<td>0.180</td>
<td>0.14 (0.180 best)</td>
</tr>
<tr>
<td>2</td>
<td>1/2</td>
<td>1/2</td>
<td>1/2</td>
<td>0.13 (1/2 exact)</td>
</tr>
<tr>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>$-0.3$ (1 exact)</td>
</tr>
</tbody>
</table>

where $\kappa_2$ does, while $\kappa_1$ does not, take account of the correction. Equation (10) gives, $\kappa_2 = \kappa_1[1 - d^{-1}[1 - 1/(1 + \Delta\kappa)]^{-1}$ Let us summarize the results for $\kappa(= 2\nu - 1)$; FE gives,

$$\kappa_{\text{flory}} = (4 - d)/(d + 2) ; \quad (11)$$

the present first approximation is,

$$\kappa_1 = (4 - d)/2d ; \quad (12)$$

while an improved one is,

$$\kappa_2 = \kappa_1/[1 - d^{-1}[1 - 1/(1 + \Delta\kappa)] , \quad (13)$$

here $\Delta\kappa$ is a continuous fraction, $\Delta\kappa = (3d/2 + 1)(d/2 - 1)\kappa_1/[1 + \Delta\kappa/(1 + ...)$. Finally, the non-approximate equality of equation (6) gives,

$$\gamma - 1 = [-B + (B^2 + 4\kappa)^{1/2}]/2 , \quad \text{where} \quad B = 1 - \kappa/2 \quad (14)$$

Table I compares, at $d = d_c - \epsilon$, 3, 2 and 1, $\kappa_{\text{flory}}$, and present $\kappa_1$ and $\kappa_2$, with exact [9] or best [10] results. The $\epsilon$ agreement improves successively, from none, to $O(\epsilon)$, and $O(\epsilon^2)$ respectively. The value of $\gamma - 1$ is also accurate to $O(\epsilon^2)$, and accurate, but not exact at $d = 2$; it is determined by an equation which, in contrast to those for $\kappa$, does not become exact when $d/2 - 1 = 0$.

3.3 POLYMER AT A FINITE CONCENTRATION. — An important support for the present description of a polymer as a CC with $b$ pseudo-bifurcations, comes from a somewhat unexpected source. Once again lets refer to the useful reconstruction of an isolated CC, or polymer, in a sequence of steps [6, 7]. Lets suppose that the probability $p$ for an $i$'th "spin" to become connected to any of its available (viz. non-visited) neighbors is $p = p_c$, such that $\langle \delta b_i \rangle = 0$. In that critical case the polymer may grow indefinitely and without blowup, precisely as we have required in equation (3). Clearly the value of $p_c$ just suffices to overcome the loss of available neighbors due to imminent self-intersections of the polymer with itself. We now pass to the case of a finite concentration of polymers. The average number of available neighbors of an $i$'th spin
is now smaller, because there now exists a probability $c$ that our polymer encounters an obstacle, viz. a neighbor that belongs already to one of the other polymers. How to ensure that the polymer will still grow indefinitely, or at least to any desired length $l$, despite $c > 0$? A naive answer is, let's simply increase the probability to $p + \Delta p(c)$. But this is quite unsatisfactory; it spoils badly the analogy to magnetic spins where a $p > p(c)$ implies the creation of an infinite CC. But no problem arises if the polymer is described as possessing $b$ pseudo-bifurcation. The obstacles merely decrease the number of bifurcations that the polymer accumulates on its way. The accumulation of obstacles is at most proportional to $l$ itself (assuming a many-“blob” length [3]). But $b \approx l^{(1+\Delta)}/(1-\Delta)$ (Eq. (4)). Hence there will be always enough bifurcations to guarantee the polymer an indefinite growth at arbitrary $c > 0$, and still at $p \neq p(c)$!

4. Discussion and conclusions.

Successive estimates of critical exponents $\nu$ and $\gamma$, are derived with the help of (closed) fluctuating-cluster equations. They attain agreement with $\epsilon$ expansion to $O(\epsilon^2)$ at $d = d_c - \epsilon$, converge much faster than $\epsilon$ expansion at other $d$, and reproduce the exact results at $d = 2$ and 1. The faster convergence is explained by that higher $O(\epsilon)$ terms enter due to two reasons. New physical phenomena, but also mere expansion of series to higher order. The second reason disappears of course in a description with the help of closed equations. Yet, the present $\epsilon^3$ term is not accurate. Possibly this could be remedied by a $\kappa_3$ approximation, which would somehow correct for that, the summation of $\delta b_i\delta b_j$ in equation (7) refers to constant $l$ (variable $s'$), whereas the definition of the correlation between the $\delta b_i\delta b_j$ pairs in equation (3) refers to constant $s$ (variable $l'$). However it is difficult to judge whether this will help to extend the agreement to $\kappa_3$. Another shortcoming is the long list of assumptions. First and foremost, the linear polymer is accorded a phantom bifurcation, which leads to the count of correlated increments in equation (6); second, the universal scaling of correlations is assumed in equations (6) and (8); third is the assumption in equation (8) that $\xi_{1/\tau,b}$ and $\xi_{\tau,b}$ correspond to a correlation and anti-correlation, respectively; finally comes the justification of $\kappa_2$ by the existence of $\Delta \kappa$. But most serious is the lack of a systematic scheme of successive approximations; indeed the assumptions made here have been guided by prior knowledge of the $\epsilon$-expansion series, and of the result at $d = 2$. To a degree this may be attributed to a lack of experience with a conceptually novel approach. Let’s therefore in conclusion recapitulate it briefly. Flory’s equation is viewed as a fortunate meanfield approximation, describing a balance of correlations at the level of a single CC, viz. an isolated fluctuation. Further improvement is achieved by letting the correlations fluctuate jointly. This leads to our revised, fluctuating Flory equation, FFE. The latter is strongly supported by that, the total power of coupled correlations increases from Flory’s $d/2 + 1$, to $d$. It makes the equation pass from an inconsistency, to a very plausible, almost self-evident, $d$-dependence in equation (9). That dependence constitutes also a unique way to determine the upper critical dimension $d_c$. Equations (8) and (9) also explain why present FFE and FE alike become exact at $d = 2$. FFE and its associated equations give a complete set of critical exponents, viz. static $\nu$ and $\gamma$, and bifurcation $\Delta$. The latter exponent does not appear to be of immediate physical significance in connection to the polymer problem; this in contrast to the Ising problem where $\Delta$ (together with static $\nu$ and $\gamma$) determines the dynamic exponent $z$ [7]. One might wonder therefore why introduce at all $\Delta$ i.e. the bizarre phantom bifurcation in equation (4), when in equation (5) already the two are substituted, respectively, by $\gamma$ and the well recognized number of polymer configurations. The justification is that it is not clear how the derivation of the equivalence leading to equation (6), and the evaluation of $Y_{cross}$ in equation (8) can be achieved without recourse to phantom bifurcation. One more aspect of bifurcation, phantom and not, merits comment. For polymers, viz. for the
n = 0 phantom case we find \( \Delta > 0 \). However for the Ising \( n = 0 \) non-phantom case \( \Delta < 0 \) [see Ref. [7], but taking into account that, in order to define positive exponents, the notation therein is \( b \approx s(1-\Delta)/2 \) ]. The reason for that appears to be the following. Simultaneous bifurcation of (adjacent) real branches' ends is associated with an anti-correlation because of \( \varepsilon_v \); hence \( \Delta < 0 \) for the non-phantom case. But the phantom branches do not interact mutually (Eq. (6)). What remains is "correlated information". If local condition are such that branch \( i \) bifurcates (viz. low \( \varepsilon_v \)), this indicates that the selfsame local condition promotes bifurcation of \( j \); viz. correlation for the \( n = 0 \) case, viz. \( \Delta > 0 \). In conclusion let it be added that the present approach also helps to describe the case of finite polymer concentration within the context of the polymer-\( n = 0 \)-spin analogy, and that it should also be applicable to other critical systems; indeed it has been already so applied [7], and is being currently reapplied [11], to statics and dynamics of the \( n = 1 \) Ising spins.

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