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To cite this article: C. von Ferber and Yu. Holovatch 1997 *EPL* **39** 31

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Copolymer networks: Multifractal dimension spectra in polymer field theory

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(received 6 January 1997; accepted 25 May 1997)

PACS. 61.41+e – Polymers, elastomers, and plastics.

PACS. 64.60Ak – Renormalization-group, fractal, and percolation studies of phase transitions.

PACS. 64.60Fr – Equilibrium properties near critical points, critical exponents.

Abstract. – We explore the rich scaling behavior of copolymer networks in solution. We establish a field-theoretic description in terms of composite operators. Our 3rd-order resummation of the spectrum of scaling dimensions brings about remarkable features: Convexity of the spectra allows for a multifractal interpretation. This has not been conceived for power of field operators of ϕ^4 field theory before. The 2D limit of the mutually avoiding walk star apparently corresponds to results of a conformal Kac series. Such a classification seems not possible for the 2D limit of other copolymer stars. The 3rd-order calculation of a large collection of exponents furthermore allows for a consistency check of two complementary schemes: epsilon expansion and renormalization at fixed dimension.

Introduction. – Recently much interest focused on the relation of field theory and multifractals [1], [2] and the associated multifractal dimension spectra [3], [4] as well as non-intersecting random walks and their 2D conformal theory [5]. We present a model of multicomponent polymer networks that shows a common core of these topics and allows for a detailed study of the interrelations. The flux of diffusion onto an absorbing fractal defines a multifractal measure. Cates and Witten [3] have mapped the moments of this flux to that of a star of random walks (RW) avoiding the absorber taken to be a polymer or RW itself. Using the field-theoretic formulation of polymer theory we show that the spectrum of scaling exponents governing these problems is given by the anomalous dimensions of composite operators with appropriate symmetry.

For polymer networks consisting of polymer chains of one species it has been shown that the basic scaling exponents are connected with “stars”, polymer chains tied together at one core [6]-[8]. The number of configurations Z_{*f} of a polymer star with f arms of N monomers

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will scale for large N like

$$\mathcal{Z}_{*f} \sim N^{\gamma_f-1} \sim (R/\ell)^{\eta_f-f\eta_2}. \quad (1)$$

The second part shows scaling with the size $R \sim N^\nu$ of the isolated coil of N monomers on some scale ℓ . The exponents $\nu = 3/4, 0.58(8)$ and $\gamma_1 = \gamma_2 = \gamma = 43/32, 1.16(0)$ for space dimensions $d = 2, 3$ are known in polymer theory [9]. The exponents γ_f have been calculated analytically in perturbation theory [7], [8], [10], by exact methods in two dimensions [6], and by Monte Carlo simulations [11].

At short distance two polymer stars will repel each other. In view of the below advocated language of field theory this is described in terms of a short-distance expansion. One finds the following relation for the probability $P(r)$ to find the cores of two stars of f_1 and f_2 at short distance r [7]:

$$P(r) \sim r^\Theta, \quad \Theta = \eta_{f_1} + \eta_{f_2} - \eta_{f_1+f_2} > 0. \quad (2)$$

This is compatible with the result that the spectrum of polymer star exponents η_f is convex from below as a function of f with $\eta_1 = 0$.

On the other hand, a multifractal (MF) measure μ_x defined on the sites x of scale ℓ on some object of size R is characterized by the scaling of its moments averaged over all sites:

$$\langle \mu_x^k \rangle = \sum_x \mu_x^k \sim (R/\ell)^{y_f}. \quad (3)$$

From general inequalities for the moments of a probability distribution one may deduce that the spectrum of exponents y_f has to be convex from above. This indicates an apparent discrepancy between objects described in field theory (FT) as powers of field (see below) such as polymer stars, and the moments of a MF measure [1]. We want to resolve this by including both concepts in the same FT formalism showing that they are special cases of a more general approach, which in addition also describes the problem of non-intersecting random walks.

To this end, we study the scaling behavior of a polymer star or a general *network* of chains of *different species* and thus, within a unique formalism, include effects caused by self and mutual interactions between polymers of different species forming a network. We combine the field-theoretic formalism developed for the description of polymer stars and networks [8] with the corresponding theory which describes multicomponent polymer solutions [12].

Theory. – We introduce a Landau-Ginsburg-Wilson Lagrangian \mathcal{L} of f interacting fields ϕ_b each with n components, *i.e.* $\phi_a^2 = \sum_{\alpha=1}^n (\phi_a^\alpha)^2$, with an interaction matrix $u_{aa'}$ and mass parameters m_a :

$$\mathcal{L}\{\phi_b, m_b\} = \frac{1}{2} \sum_{a=1}^f \int d^d r (m_a \phi_a^2 + (\nabla \phi_a(r))^2) + \frac{1}{4!} \sum_{a,a'=1}^f u_{aa'} \int d^d r \phi_a^2(r) \phi_{a'}^2(r). \quad (4)$$

In this theory the star exponents are given in terms of the anomalous dimensions of composite operators $\prod_{a=1}^f \phi_a$ [8]. We define vertex functions Γ^{*f} with insertion of this operator by

$$\delta(q_0 + \dots + q_f) \Gamma^{*f}(q_0 \dots q_f) = \int \prod_{k=0}^f e^{i(q_k r_k)} d^d r_k \left\langle \prod_{a=1}^f \phi_a(r_0) \phi_1(r_1) \dots \phi_f(r_f) \right\rangle_{1\text{pi}, n=0}^{\mathcal{L}}. \quad (5)$$

As in standard polymer FT, this is evaluated with respect to the Lagrangian (4) keeping only contributions which correspond to one-particle irreducible (1pi) graphs which have non-vanishing tensor factors in the $n = 0$ limit. In the single-component case the theory may also be

described in terms of one $O(n)$ symmetric field ϕ with $n > f$, where the corresponding operator is $N^{\alpha_1 \dots \alpha_f} \phi^{\alpha_1} \dots \phi^{\alpha_f}$ with a traceless tensor $N^{\alpha_1 \dots \alpha_f}$ in the formal limit $n = 0$ [8], [13].

We apply RG theory to make use of the scaling symmetry of the systems in the asymptotic limit to extract the universal content and at the same time remove divergences which occur for the evaluation of the bare functions in this limit [14]. Several asymptotically equivalent procedures serve to the purpose of renormalization. In the present study we use two somewhat complementary approaches: zero-mass renormalization with successive $\varepsilon = (4 - d)$ -expansion [14] and the massive RG approach at fixed dimension [15]. Application of both approaches will enable us to check the consistency of approximations and the accuracy of the results obtained. We pass from the theory in terms of the initial bare variables to a renormalized theory. This can be achieved by a controlled rearrangement of the series for the vertex functions (5) introducing renormalizing Z -factors for fields (Z_{ϕ_a}), couplings (Z_{ab}) and mass. Then, for instance the bare couplings u_{ab} are given in terms of their renormalized dimensionless counterparts g_{ab} by

$$u_{ab} = \kappa^{4-d} Z_{\phi_a} Z_{\phi_b} Z_{ab} g_{ab} . \quad (6)$$

The scale parameter κ represents the mass at which the massive scheme is evaluated and the scale of external momenta in the massless ε -expansion scheme. We define the Z -factors in (6) as to renormalize the correlators $\langle \dots \rangle^{\mathcal{L}}$ in each RG procedure (see, *e.g.*, [14]). The polymer limit $n = 0$ of zero-component fields leads to essential simplification. Each field ϕ_a , mass m_a and coupling u_{aa} renormalizes as if the other fields were absent. The renormalization of the couplings u_{ab} involves only the fields ϕ_a, ϕ_b [12]. The renormalized couplings g_{ab} defined by relations (6) depend on the scale parameter κ . Thus the renormalization Z -factors also depend implicitly on κ . This dependence defines the RG functions and exponents: $\kappa \frac{d}{d\kappa} g_{aa} = \beta_{aa}(g_{aa})$; $\kappa \frac{d}{d\kappa} g_{ab} = \beta_{ab}(g_{aa}, g_{bb}, g_{ab})$; $\kappa \frac{d}{d\kappa} \ln Z_{\phi_a} = \eta_{\phi_a}(g_{aa})$. The function η_{ϕ_a} defines the pair correlation critical exponent. The set of scaling exponents η_{*f} for general copolymer stars is defined by the renormalization factors Z_{*f} for the star vertex functions Γ^{*f} :

$$\prod_{a=1}^f Z_{\phi_a}^{1/2} Z_{*f} \Gamma^{*f}(u_{bb'}(g_{bb}, g_{b'b'}, g_{bb'})) = \kappa^{\delta_f}, \text{ with } \eta_{*f}(g_{ab}) = \kappa \frac{d}{d\kappa} \ln Z_{*f} . \quad (7)$$

$\delta_f = d + (1 - d/2)f$ is the engineering dimension of the corresponding bare vertex function.

In a study devoted to ternary polymer solutions the RG flow given by the above-defined β -functions has been calculated [12], [16] to third loop order. The equations for the fixed points of the β -functions were found to have the following non-trivial solutions: $\beta_{aa}(g_S^*) = 0$ and for $a \neq b$: $\beta_{ab}(0, 0, g_G^*) = 0$, $\beta_{ab}(g_S^*, 0, g_U^*) = 0$, $\beta_{ab}(0, g_S^*, g_U^*) = 0$, $\beta_{ab}(g_S^*, g_S^*, g_S^*) = 0$, corresponding to all combinations of interacting and non-interacting chains.

We evaluate the exponents for two general arrangements of the fixed-point matrix: the ternary case of two mutually interacting species of polymer chains in solution, and the mutual avoiding walk case of essentially f *only mutually* interacting species. In the first case we describe polymer stars made of f_1 chains of species 1 and $f_2 = f - f_1$ chains of species 2. Either both species are non-self-interacting and

$$\eta_{f_1 f_2}^G \equiv \eta_{*f}(g_{ab} = 0 \text{ if } a, b \leq f_1 \text{ or } a, b > f_1; \text{ else } g_{ab} = g_G^*) , \quad (8)$$

or species 1 self-interacts and species 2 does not, such that

$$\eta_{f_1 f_2}^U \equiv \eta_{*f}(g_{ab} = g_S^* \text{ if } a, b \leq f_1; g_{ab} = 0 \text{ if } a, b > f_1; \text{ else } g_{ab} = g_U^*) . \quad (9)$$

For $f_2 = 0$ this includes the homopolymer star with $\eta_f = \eta_{f,0}^U$ in eq. (1). The mutually avoiding walk case reads

$$\eta_f^{\text{MAW}} \equiv \eta_{*f}(g_{ab} = 0 \text{ if } a = b; \text{ else } g_{ab} = g_G^*) . \quad (10)$$

Results. – We give the results for the exponents in $\varepsilon = (4-d)$ -expansion. The corresponding more lengthy expressions obtained by fixed $d = 3$ RG may be found in [16]:

$$\begin{aligned} \eta_{f_1 f_2}^G(\varepsilon) = & -f_1 f_2 \frac{\varepsilon}{2} + f_1 f_2 (f_2 - 3 + f_1) \frac{\varepsilon^2}{8} - \\ & -f_1 f_2 (f_2 - 3 + f_1) (f_1 + f_2 + 3\zeta(3) - 3) \frac{\varepsilon^3}{16}, \end{aligned} \quad (11)$$

$$\begin{aligned} \eta_{f_1 f_2}^U(\varepsilon) = & f_1 (1 - f_1 - 3f_2) \frac{\varepsilon}{8} + f_1 (25 - 33f_1 + 8f_1^2 - 91f_2 + 42f_1 f_2 + 18f_2^2) \frac{\varepsilon^2}{256} + \\ & + f_1 (577 - 969f_1 + 456f_1^2 - 64f_1^3 - 2463f_2 + 2290f_1 f_2 - 492f_1^2 f_2 + 1050f_2^2 - \\ & - 504f_1 f_2^2 - 108f_2^3 - 712\zeta(3) + 936f_1 \zeta(3) - 224f_1^2 \zeta(3) + \\ & + 2652f_2 \zeta(3) - 1188f_1 f_2 \zeta(3) - 540f_2^2 \zeta(3)) \frac{\varepsilon^3}{4096}, \end{aligned} \quad (12)$$

$$\begin{aligned} \eta_f^{\text{MAW}}(\varepsilon) = & -(f-1)f \frac{\varepsilon}{4} + f(f-1)(2f-5) \frac{\varepsilon^2}{16} - \\ & -(f-1)f(4f^2 - 20f + 8f\zeta(3) - 19\zeta(3) + 25) \frac{\varepsilon^3}{32}. \end{aligned} \quad (13)$$

Here $\zeta(3) \simeq 1.202$ is the Riemann ζ -function. The above formulas reproduce the 3rd-order calculations of $\gamma_f - 1 = \nu(\eta_{f,0}^U - f\eta_{2,0}^U)$ [8] as well as the 2nd-order exponents $\lambda^{(\text{xx})}$ defined in equations (xx) of [3], $\lambda^{(29)}(n) = -\eta_{2,n}^G$, $\lambda^{(47)}(n) = -\eta_{2,n}^U + \eta_{2,0}^U$, $\lambda_e^{(48)}(n) = -\eta_{1,n}^G$, $\lambda_e^{(49)}(n) = -\eta_{1,n}^U$, correcting a missprint in eq. (49) of [3]. Also the 2nd-order results for the exponents $x_{L,n} - x_{L,1} = -2(\eta_{L,n}^G - \eta_{L,1}^G)$ of [1] and $\sigma_L = 1/2\eta_L^{\text{MAW}}$ defined in [5] find their 3rd-order extension by the above expansions.

With these exponents we can describe the scaling behavior of polymer stars and networks of two components, generalizing the relation for single-component networks [7]. In the notation of (1) we find for the number of configurations of a network \mathcal{G} of F_1 and F_2 chains of species 1 and 2

$$\mathcal{Z}_{\mathcal{G}} \sim (R/\ell)^{\eta_{\mathcal{G}} - F_1 \eta_{20} - F_2 \eta_{02}}, \text{ with } \eta_{\mathcal{G}} = -dL + \sum_{f_1+f_2 \geq 1} N_{f_1 f_2} \eta_{f_1 f_2}, \quad (14)$$

where L is the number of loops and $N_{f_1 f_2}$ the number of vertices with f_1 and f_2 arms of species 1 and 2 in the network \mathcal{G} . To receive an appropriate scaling law we assume the network to be built of chains which for both species will have a coil radius R when isolated.

To obtain reliable numerical values from the ε -expansions (11)-(13) in and from the series obtained in the fixed- d scheme [16] we apply Borel resummation using the technique of conformal mapping [17] which has proven to yield good results for many critical exponents. We use information about the higher-order behavior [17], [12] of the series (11)-(13) derived from the instanton analysis of the appropriate field theory. The results for $d = 3$ are given in table I. The data show consistency and stability of the results, while deviations grow for large number of arms, as may be expected. Note that the above expansions are in fact series in $f\varepsilon$, not ε alone.

Conclusion: multifractals and field theory. – Does the data answer the question of convexity? A close study of the matrix of values reveals that for fixed f_1 both $\eta_{f_1 f_2}^G$ and $\eta_{f_1 f_2}^U$ are convex from above as a function of f_2 , thus yielding “MF statistics”. The relation to a MF

have the appropriate short-distance behavior. This is also illustrated in fig. 1, showing the spectrum of exponents $\eta_{f_1 f_2}^U$ in the 2D limit [16]. The opposite convexity along the two axes is clearly seen for these unsymmetric combinations of a polymer f_1 -star and a random walk f_2 -star which mutually interact.

2D copolymer stars. – The 2D exponents for polymer stars have been shown to belong to a Kac series of exponents of conformal FT with $\gamma_f - 1 = (4 + 27f - 9f^2)/64$ [6]. There are strong indications that this is the case also for MAW stars with $\eta_f^{\text{MAW}} = (1 - 4f^2)/12$ [5]. Already in view of fig. 1, though, such a simple 2nd-order polynomial seems not to describe the 2D limit of general copolymer star exponents. In 2D, however, each chain of a star will interact only with its direct neighbors. A star described here by η_{ff}^G will behave like a MAW $2f$ -star if each species-1 chain has two neighbors of species-2 whereas it will behave differently if the chains are ordered in such a way that each species is in one bulk of chains. The 2D copolymer stars in this sense reveal an even richer behavior. Thus, the copolymer generalization of the MAW star adds another problem, for which a rigorous formulation in terms of an exactly solvable 2D model is yet to be found.

We thank L. SCHÄFER for valuable discussions. Supported in part by Deutsche Forschungsgemeinschaft SFB 237 and Minerva Gesellschaft.

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