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Dilute and dense systems of random copolymers in the equilibrium state

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Dilute and dense systems of macromolecules with quenched compositional disorder along the chain and the short-range interaction between monomers are studied by numerical Monte Carlo (MC) simulations. The equilibrium structural properties of such systems are characterized by the mean-square end-to-end distance and the static structure factor of the polymer chains. Both temperature and density determine the structure (collapse, swelling, or screening regime). A description of this behavior is given using generalized scaling arguments and proven by the numerical MC simulations.

I. INTRODUCTION

Various concepts of statistical mechanics of random systems, such as those of a spin glass[1] or of neural networks,[2] are used to describe the complex structural conformation of biological macromolecules. In the first case, the strength and the sign of the interaction between pairs of monomers are chosen at random;[3,4] i.e., we have $N(N-1)/2$ different interacting pairs whose interaction is statistically distributed. In the second example the properties of neural networks are used, where the interaction results from a learning process, using the information from known structures.[2]

In the present paper we investigate a model in which the interaction of each pair of monomers is well defined by a bilinear function of the characteristic values of both monomers (usually called charges) and a unique potential.[5–8] The randomness is determined by the quenched disorder of the monomeric charge sequences $\{q_i\}$.[9] Therefore, the interaction potential is given by the general expression

$$U_{ij}(r_i-r_j)=w(r_i-r_j)+b(r_i-r_j)[q_i+q_j]+v(r_i-r_j)q_iq_j, \tag{1}$$

with q_i being the charge of the monomer i. The first term contains nonstochastic interactions (for example the excluded volume interaction), which in most cases is of short-range order, $w(r)=w_0\delta(r)$. The influence of the second term is discussed in Ref. 9. In the case of invariance against a charge inversion $q_i\rightarrow -q_i$, $b=0$ follows immediately. In the present paper we restrict ourself to this case; i.e., we discuss the influences of the potentials w and v only.

A single chain with a short-range stochastic interaction $v(r-r')=-v_0\delta(r-r')$ and vanishing excluded volume effect $(w_0=0)$ shows a collapse regime for long chains and a strong monomer interaction $v_0\Delta_0$ (with $\langle q_iq_j\rangle=\Delta_0\delta_{ij}$), whereas a swollen regime with a swelling exponent $\nu=1+\epsilon^2/8$, $\epsilon=d_c-d$ was predicted[10] for short chains and a weak interaction $v_0\Delta_0$ (and with an upper critical dimension $d_c=2$) (see also the numerical results of Ref. 6). On the other hand, the presence of a nonvanishing excluded volume interaction $w_0\neq 0$ leads to a usual swelling regime for a weak stochastic interaction $v_0\Delta_0$ or short chains and a collapse regime $R\sim N^{1/d}$ for a strong stochastic interaction $v_0\Delta_0$.[11] It should be remarked that a long-range stochastic interaction $[v(r)=v_0r^{2-d}]$ leads to a general collapse transition for all dimensions $d<d_c=4$.[12]

However, these briefly discussed results hold for isolated chains only. Therefore, the equilibrium behavior of dense and semidilute systems of such stochastic copolymer chains is an interesting question, which will be discussed in the present paper by using numerical Monte Carlo simulations and generalized scaling arguments.

II. MODEL

The present work is devoted to a study of the equilibrium properties of random copolymer melts or semidilute solvents with a stochastic chargelike short-range monomer interaction (1). A numerical investigation of a dense system of such stochastic copolymers and a check of the predicted scaling behavior are possible by extending the well-known fluctuating bond model.[13,14]

In this lattice Monte Carlo simulation each effective monomer occupies all eight corners of a unit cell of the simple cubic lattice and the bond lengths (all length are measured in units of the lattice spacing) between consecutive monomers vary between 2 and $\sqrt{10}$ [determined by the lattice structure; $\sqrt{8}$ is excluded, in order to maintain entanglement restrictions (see Ref. 14)]. The allowed bond lengths guarantee that the self-avoiding condition (each lattice site can be taken at most once and therefore each monomer has a hard core behavior) is satisfied and that chains cannot intersect in the course of their random motion (modeled by stochastic jumps of effective monomers to neighboring unit cells). As has been discussed elsewhere,[13–15] each effective bond of the model is thought to represent a group of successive chemical bonds along the backbone of a real polymer chain, and hence it is physically reasonable for flexible polymers that the length of such effective bonds may fluctuate.

The systems considered have a volume $V=L^3$, $L=48$, filled with n_{ch} chains of stochastic charge sequences $\{q_i\}$ (chain length $N=50$, random distribution of charges

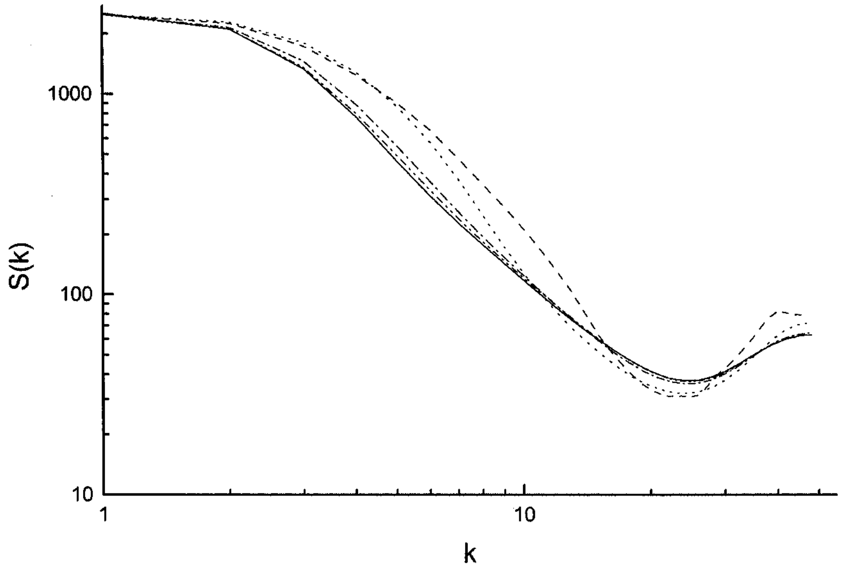

FIG. 1. Static structure factor $S(k)$ of a diluted polymer system ($\rho = 0.018$) for different temperatures [$T = 1000$ (solid line), $T = 50$ (dash-double-dotted line), $T = 25$ (dash-dotted line), $T = 5$ (dotted line), $T = 1$ (dashed line)].

$\langle q_i q_j \rangle = \Delta_0 \delta_{ij}$, the total system is neutral; note that ten configurations run in parallel to improve the statistics.) The repulsive potential U_{ij}, which acts between each monomer pair, is defined by the step function

$$U_{ij} = [w_0 + v_0 q_i q_j] \Theta(R - |r_i - r_j|), \tag{2}$$

with $\Theta(x) = 0$ for $x \leqslant 0$ and $\Theta(x) = 1$ for $x > 0$. The cutoff radius R (5 unit lengths) corresponds to the first neighbor peak in the static structure factor $S(k)$. This energy is considered in the transition probability $W = \exp(-\Delta U/kT)$, ΔU being the energy change associated with an elementary motion, as standard in the framework of the Metropolis algorithm.[16,17] Because the excluded volume interaction is controlled by the hard core character of the monomers, we can neglect the contribution w_0. Furthermore, the athermal hard core interaction allows the introduction of a reduced temperature $\tau \sim T/(v_0 \Delta_0)$. In other words, there exists only one relevant energy parameter, e.g., the temperature T, scaled by the interaction strength $v_0 \Delta_0$.

It is well known from a series of publications that the fluctuating bond model works well also for dense systems at low temperatures. For example, numerical simulations of the glass transition[20,21] of (uncharged) polymers show no serious ergodicity problems or other unusual behavior.

Therefore, the complete thermodynamic system is controlled by two relevant parameters, the temperature T and the monomer density ρ (a third parameter is the chain length N, whose influence is not analyzed in the present investigations). We expect a strong temperature dependence of the equilibrium structure for dilute polymer solvents (swelling regime in the high-temperature limit and collapse regime in the low-temperature regime) determined by the strength $v_0 \Delta_0 / T$. On the other hand one can observe a strong screening effect[22] in dense systems; i.e., the difference between the equilibrium structures for different temperatures should decrease with increasing density.

III. NUMERICAL RESULTS

A. Static structure factor

After reaching the equilibrium distribution of the polymer system, the values discussed below are measured as a time average over approximately 10^2 configurations (time separated by approximately 10^5 elementary steps per monomer) in ten ensembles (i.e., the total number of configurations used was $\sim 10^3$). Note that the time interval between two successive configurations is sufficiently long to scan a reasonable part of the phase space also in dense systems. The mean-square displacement of the center of mass of the chains during the whole simulation is approximately 1/3 of the radius of gyration in the most unfavorable case. Thus, the accuracy of the numerical simulations is determined by a numerical error of approximately 3%.

The static structure factor of the whole system is defined by

$$S(k) = \sum_{i,j} \langle \exp\{\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)\} \rangle, \tag{3}$$

whereas the static structure factor of a polymer chain is given by

$$S(k) = \frac{1}{n_{\mathrm{ch}}} \sum_{\mathrm{chains}} \sum_{i,j \in \mathrm{chain}} \langle \exp\{\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)\} \rangle \tag{4}$$

(n_{ch} is the total number of chains of the system). Note that the computation of the static structure factor using (3) leads to unwanted Bragg reflections of the underlying lattice structure. Therefore, we use for the discussion the static structure factor of the polymer chains (4), in which such effects vanish as result of the incoherent averaging procedure.

The simulation shows significant differences of the static structure factors for different temperatures in the case of strongly diluted systems; see Fig. 1. Note that the wave number k is scaled in units of $2\pi/L$ ($L = 48$). Here, the plot $\ln S(k)$ versus $\ln k$ for the high-temperature solution shows a characteristic slope $\nu \approx 1.2$ for $3 \leqslant k \leqslant 8$, which is character-

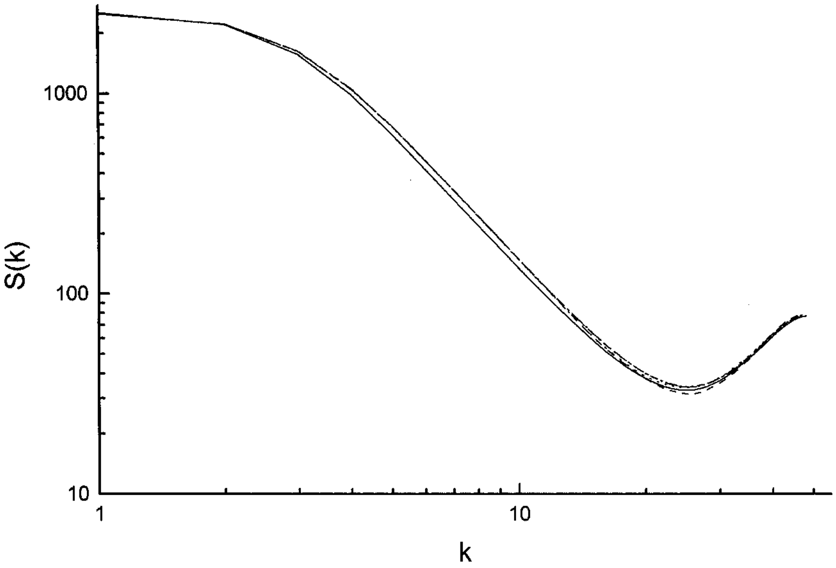

FIG. 2. Static structure factor $S(k)$ of a dense polymer system ($\rho=0.375$) for different temperatures [$T=1000$ (solid line), $T=50$ (dash-double-dotted line), $T=25$ (dash-dotted line), $T=5$, (dotted line), $T=1$ (dashed line)].

istic for a swelling regime. On the other hand, the structure factor for low temperature behaves like the one of a system of more compact objects.

However, in the case of high density, no significant differences between the static structure factors for different temperatures can be observed; see Fig. 2. This behavior indicates an increasing screening effect with increasing density.

B. End-to-end distances

A second possibility for the numerical characterization of the equilibrium state is the determination of the mean square end-to-end distance $\langle R^2\rangle$ of the polymer chains. Figure 3 shows $\langle R^2\rangle$ for different temperatures and densities. As expected, the dilute systems shows a strong temperature dependence (swelling for the high-temperature limit, collapse for the low-temperature regime). This difference decreases with increasing densities and vanishes almost for the highest possible density.

IV. SCALING HYPOTHESIS

Now we will predict and check some scaling arguments for the quantitative description of the equilibrium structure of polymer melts and solutions of stochastic copolymers. Scaling theory or scaling arguments can be defined in physical or geometrical terms,[18] but also simply postulated mathematically or empirically.[19] The latter approach is used in the following discussion. Experience with various critical phenomena (including the theory of equilibrium properties of polymer chains) has indicated that most functions $g(x,y)$ of two variables approach the scaling hypothesis

$$g(x,y)\simeq x^{\lambda}G\left(\frac{y}{x^{\mu}}\right) \tag{5}$$

if x and y approach zero or infinity (G, scaling function; y/x^{μ}, scaling ratio; λ, μ, critical exponents). Note that this equation is only a reasonable assumption, not a mathematical or physical theorem. But equations of this structure are used in many cases as a first approach to complex systems.

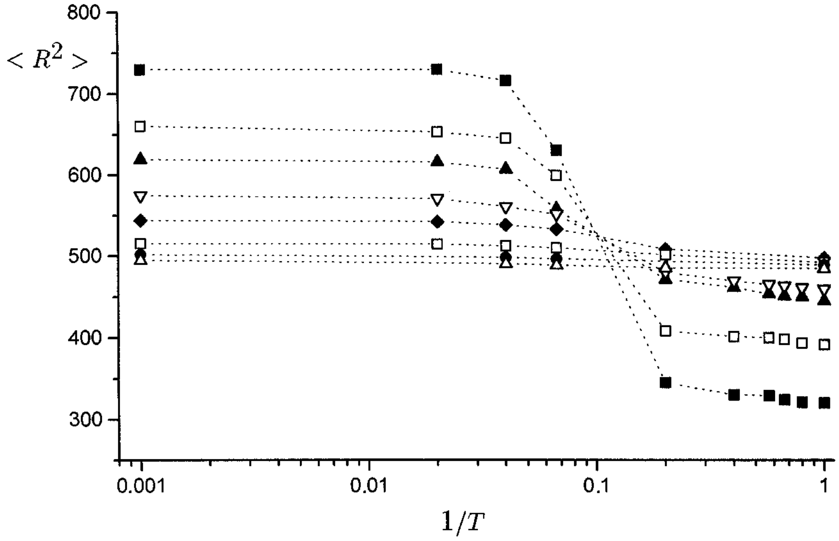

FIG. 3. Mean-square end-to-end distance of the stochastic copolymer chains for different densities [$\rho=0.018$ (solid square), $\rho=0.069$ (open square), $\rho=0.119$ (solid triangle), $\rho=0.170$ (open inverted triangle), $\rho=0.221$ (solid diamond), $\rho=0.271$ (open square), $\rho=0.322$ (solid circle), $\rho=0.375$ (open triangle)] as a function of the inverse temperature.

We assume that such an empirical scaling hypothesis leads to a first simple representation of the equilibrium structure of random copolymer systems. The basic idea, which we will use in the present investigation, is the blob picture, introduced by de Gennes.[22] We give a brief discussion of this concept and generalize the scaling arguments used to the case of stochastic copolymers.

A. Geometrical scaling function

We start our investigation from a polymer system without a stochastic monomer interaction (which corresponds to the high-temperature limit of our model). A blob is a volume, which is mainly occupied by monomers of one chain. If the chain length is N, each chain consists of m blobs of n monomers, i.e., $N=mn$. Therefore, the monomers of each blob behave approximately as a free chain of length n and diameter ξ, which are connected by the relation

$$\xi^2 \sim n^{2\nu} \tag{6}$$

with the swelling exponent ν. In addition, we have the condition that all blobs of the system occupy the whole volume of the system, i.e., $\rho = n\xi^{-d}$ (d, spatial dimension; ρ, averaged monomer density). Thus, one obtains $n \sim \rho^{\beta}$ and $\xi \sim \rho^{\nu\beta}$ [with $\beta=(1-\nu d)^{-1}$]. Both relations determine the diameter and monomer number, respectively, of a blob as a function of the monomer density ρ. Note that these relations are valid only in the case that a chain (diameter R, $R \approx \sqrt{\langle R^2 \rangle}$) occupies a volume large as compared to N/ρ. Otherwise, we have no effective overlap between different chains and therefore $\xi=R$ and $n=N$. Thus, we can introduce a characteristic density

$$\rho_c = \frac{N}{R^d}, \tag{7}$$

with

$$n \sim \rho^{\beta} \text{ and } \xi \sim \rho^{\beta\nu} \text{ for } \rho \gg \rho_c,$$
$$n = N \text{ and } \xi = R \text{ for } \rho \ll \rho_c. \tag{8}$$

Furthermore, R can be determined as the averaged end-to-end distance of a random walk of m blobs with a diameter ξ,

$$R^2 \sim m\xi^2 \sim \frac{N}{n}\xi^2. \tag{9}$$

A unification of both limiting cases (8) by using the scaling function Ψ leads to

$$n = N\Psi\left(\frac{\rho}{\rho_c}\right), \tag{10}$$

with

$$\Psi(x) = \begin{cases} 1 & \text{for } x \to 0, \\ x^{2/2-d} & \text{for } x \to \infty. \end{cases} \tag{11}$$

Note that $\rho/\rho_c \to 0$ yield the second limiting case of (8), whereas the first relation follows by using (9) and (11) in the limit $\rho/\rho_c \to \infty$.

The scaling relation (10) and Eqs. (9) and (6) form a complete set of equations for the determination of R, ξ, and n as functions of the chain length N and the monomer density ρ. It should be remarked that (10) and the function Ψ, respectively, depend only on the spatial dimension, not on the swelling exponent ν. Therefore, we can assume that the scaling function Ψ is a purely geometrical (or topological) scaling function, which is not related to a possible swelling or collapse regime. Furthermore, the scaling ratio ρ/ρ_c is a measure only for the mutual overlap of the polymer chains at the density ρ.

B. Temperature scaling

The influence of the swelling or collapse regime, respectively, on R is determined by the structure of the blobs and therefore defined by (6) only (in the case of a swelling regime). Thus, we need an extension of this high-temperature relation (swelling regime) to include also the low-temperature regime.

The averaged diameter R of a single stochastic copolymer can be described in the course of a mean-field approximation by the relation[11]

$$\frac{R^2}{N} - 1 = \frac{\overline{w}N^2}{R^d} - \frac{R^d}{2}\left[\ln\left(1 + \frac{2\overline{\Delta}N}{R^d}\right) - \frac{2\overline{\Delta}N}{R^d + 2\overline{\Delta}N}\right] \tag{12}$$

(R is given in units of the effective monomer length l_0, and $\overline{w}$ and $\overline{\Delta}$ are temperature-dependent parameters, which describe the effective interaction between the monomers: $\overline{w}$ is a measure for the excluded volume effect, and $\overline{\Delta}$ is controlled by the ratio $\nu_0\Delta_0/T$). It is simple to see that the chain shows a swelling behavior for vanishing $\overline{\Delta}$, whereas for sufficiently large $\overline{\Delta}$ a collapse can be observed. However, the swelling or collapse regime depends also on the chain length. For fixed parameters $\overline{w}$ and $\overline{\Delta}$ a collapse regime follows for long chains, whereas short chains behave like self-avoiding walks. To determine the characteristic length $N_c(T)$ [with $N<N_c(T)$, swelling regime, and $N>N_c(T)$, collapse regime], we define the crossover between both regimes by $R_c=\sqrt{N_c}$. Using (12), we obtain

$$\frac{2\overline{w}}{\overline{\Delta}^2} = \frac{f(x)}{x^2}, \tag{13}$$

with $x=N^{1-d/2}\overline{\Delta}$ and $f(x)=\ln(1+2x)-2x/(1+2x)$. The expression $g(x)=f(x)/x^2$ decreases monotonically with increasing x. Additionally, we have $g(0)=2$ and $g(\infty)=0$. Thus, for $\overline{\Delta}<\sqrt{\overline{w}}$ only the swelling regime exists, whereas for $\overline{\Delta}>\sqrt{\overline{w}}$ a characteristic length $N_c(T)$ follows as solution of (13). It should be expected that for real stochastic copolymer chains the mean-field solution must be modified, but this does not change the existence of the characteristic length $N_c(T)$ and its general temperature behavior ($N_c \to 0$ for $\nu_0\Delta_0/T \to \infty$ and $N_c \to \infty$ for $\nu_0\Delta_0/T \to 0$ and $\nu_0\Delta_0/T \leqslant \nu_0\Delta_0/T_{\max}$, respectively).

However, the existence of a characteristic length leads now to a scaling hypothesis for the temperature dependence

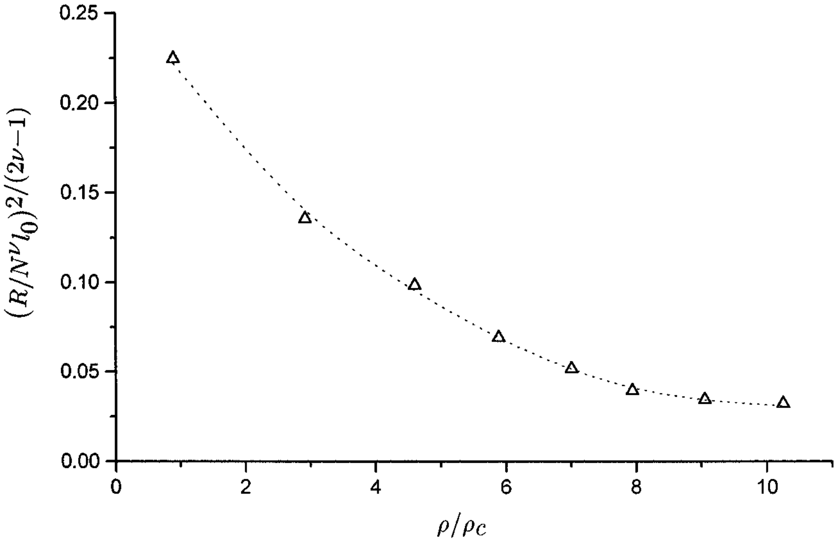

FIG. 4. Scaling function Ψ: plot of $(R/N^{\nu}l_0)^{2/(2\nu-1)}$ versus the ratio ρ/ρ_c ($\nu=0.6$).

(thermodynamic behavior) of the single stochastic copolymer chain (R is only controlled by the chain length N and the scaling ratio of N/N_c):

$$R=N^{\nu}F\left(\frac{N}{N_c(T)}\right), \tag{14}$$

with the scaling function

$$F(x)\sim\begin{cases}1 & \text{for} \quad x\rightarrow 0,\\ x^{1/d-\nu} & \text{for} \quad x\rightarrow\infty.\end{cases} \tag{15}$$

This formula describes the behavior of a single stochastic copolymer chain with sufficient accuracy; see the molecular dynamics simulations in Ref. 11. Especially both limiting cases, i.e., collapse and swelling regimes, fulfill the scaling hypothesis (14). Only the behavior of the crossover regime is more complicated [especially the Θbehavior is violated in part: $R\sim\sqrt{N}$ is not obtained for $N=N_c(T)$ but for $N>N_c(T)$] and cannot completly be described in terms of the scaling behavior (14). Fortunately, these deviations can be neglected for sufficiently small N (for the present simulations we have always chains or blob chains with $N\leqslant 50$). However, this temperature scaling allows a reasonable description of the equilibrium behavior of a single stochastic copolymer chain.

C. General scaling

To get a general scaling hypothesis for the averaged end-to-end distance R, we need the following assumptions.

(i) The blob equation (6), which holds only for the swelling regime, must be replaced by the more general scaling relation (14), but now for the blob diameter ξ and the length n,

$$\xi=n^{\nu}F\left(\frac{n}{N_c(T)}\right).$$

(ii) The geometrical scaling hypothesis (10) depends only on the ratio ρ/ρ_c. Hence, we obtain

$$R^2=m\xi^2=\frac{N}{n}n^{2\nu}F^2\left(\frac{n}{N_c(T)}\right).$$

Finally, we get the general scaling hypothesis by using (10):

$$R=l_0N^{\nu}\Psi^{\nu-1/2}\left(\frac{\rho}{\rho_c}\right)F\left(\frac{N\Psi\left(\frac{\rho}{\rho_c}\right)}{N_c(T)}\right). \tag{16}$$

Note that the length l_0 scales R and should be chosen on the order of an effective segment length. This representation is advantageous because both functions Ψ and F become dimensionless.

V. DISCUSSION OF THE SCALING HYPOTHESIS

From the theoretical point of view the scaling law (16) is an assumption, which is supported by some evident scaling arguments. To prove this relation by the numerical results presented above, we determine in a first step the scaling function $\Psi(x)$. We expect from (16) for the high-temperature limit $T\rightarrow\infty$, i.e., $N_c\rightarrow\infty$, the behavior

$$\frac{R}{l_0}\simeq N^{\nu}\Psi^{\nu-1/2}\left(\frac{\rho}{\rho_c}\right)F(0)=N^{\nu}\Psi^{\nu-1/2}\left(\frac{\rho}{\rho_c}\right)$$

and therefore

$$\Psi\left(\frac{\rho}{\rho_c}\right)=\left(\frac{R}{N^{\nu}l_0}\right)^{2/(2\nu-1)}.$$

Figure 4 shows the representation of $(R/N^{\nu}l_0)^{2/(2\nu-1)}$ versus the ratio $x=\rho/\rho_c$. As expected, the function $\Psi(x)$ tends to a finite value for $x\rightarrow 0$, whereas for large x a power law with $\Psi\sim x^{-2}$ can be observed (with $l_0=3$ and the usual dimension $d=3$).

From the scaling function Ψ we determine for each temperature T and density ρ the values

$$Y=\frac{R}{l_0N^{\nu}\Psi^{\nu-1/2}\left(\frac{\rho}{\rho_c}\right)} \quad \text{and} \quad X=N\Psi\left(\frac{\rho}{\rho_c}\right). \tag{17}$$

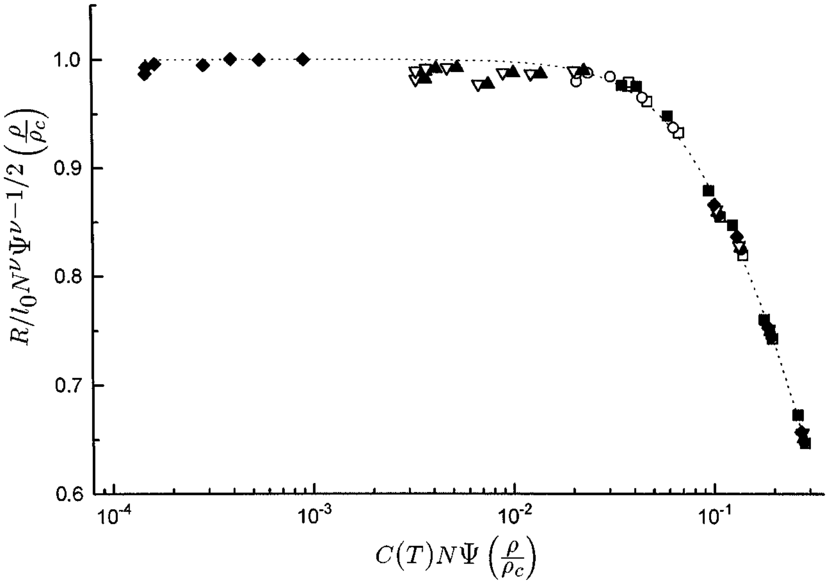

FIG. 5. Scaling function F: plot of $R/l_0N^{\nu}\Psi^{\nu-1/2}(\rho/\rho_c)$ versus $C(T)N\Psi(\rho/\rho_c)$. The function $C(T)$ corresponds to the inverse characteristic length $N_c(T)\sim 1/C(T)$. Each symbol corresponds to a fixed temperature; i.e., all points with the same symbol have the same scaling constant $C(T)$ [$T=1000$ (solid diamond), $T=50$ (open inverted triangle), $T=25$ (solid triangle), $T=15$ (open circle), $T=5$ (solid square), $T=2.5$ (solid diamond), $T=1.75$ (open inverted triangle), $T=1.5$ (solid triangle), $T=1.25$ (solid circle), $T=1$ (solid square)].

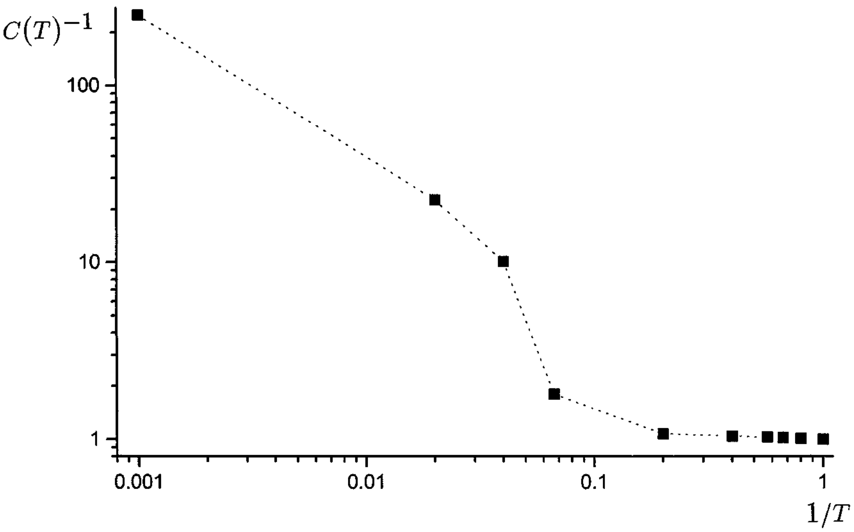

FIG. 6. Characteristic length $N_c(T)=1/C(T)$ as a function of the inverse temperature.

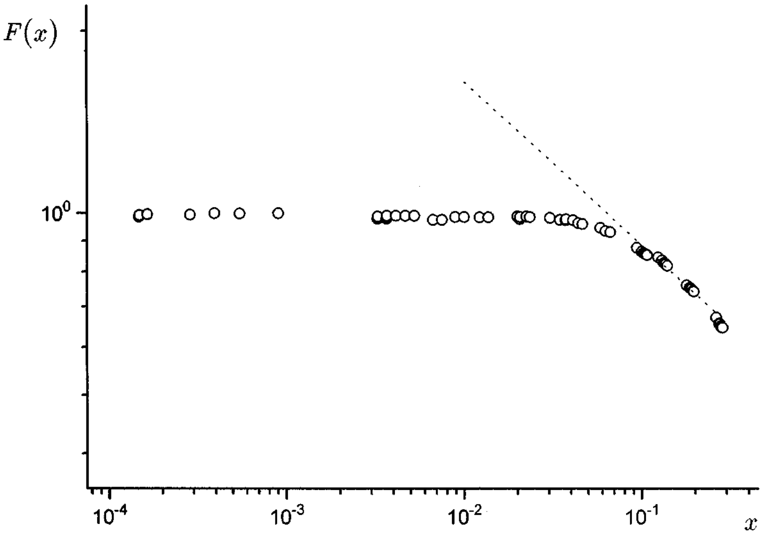

FIG. 7. Scaling function $F(x)$ in comparison with $x^{1/d-\nu}$.

After rescaling $X(T,\rho)$ with a temperature-dependent constant $C(T)$ we obtain one common curve, which connects all values $Y(T,\rho)$ with the corresponding $X(T,\rho)$,

$$Y = F(C(T)X). \tag{18}$$

The scaling function is shown in Fig. 5. The characteristic length $N_c(T)$ is defined by $N_c(T) \sim 1/C(T)$. Figure 6 shows $N_c(T)$ versus the inverse temperature. As discussed above, the characteristic length $N_c(T)$ increases with increasing temperature. The scaling function $F(x)$ has the limit $F(0)=1$ and behaves as $F(x) \sim x^{1/d-\nu}$ for sufficiently large x; see Figure 7.

In conclusion, the general scaling hypothesis discussed above leads to a first reasonable description of the equilibrium structure of dilute and dense systems of random copolymers. It should be remarked that the complete spectrum of different possible regimes (collapse, swelling, and screening regimes) is controlled by two different ratios of length scales. The temperature-dependent characteristic length $N_c(T)$ controls the thermodynamics of the system; i.e., the ratio $N/N_c(T)$ controls the thermodynamic influence of the stochastic disorder along the polymer chains in comparison to the excluded volume effect. The second length scale — the size of the effective volume per chain — is a measure for the mutual overlap of the chains. Therefore, the ratio between the end-to-end distance R and this size controls the effective geometrical overlap of different polymer chains. This ratio can be expressed by $\rho R^d/N = \rho/\rho_c$. The numerical simulations confirm the present scaling arguments for different equilibrium regimes.

A dilute systems at low temperatures shows a collapselike behavior of the macromolecules. This collapse results from the large value $\nu_0 \Delta_0/T$, which emphasizes the influence of the stochastic disorder along the polymer chains. On the other hand, the chains of a dilute system of stochastic copolymers are represented by self-avoiding walks (swelling regime) at high temperatures. High temperatures suppress the chargelike interaction of the monomers and only the athermal excluded volume effect remains. The intramolecular interaction of the chains disappears with increasing density of the system in favor of the intermolecular interaction. Therefore, one can observe an increased screening effect of the monomers. Consequently, the differences between equilibrium states at different temperatures decrease and vanish for dense systems. This can be observed in the temperature invariance of the static structure factor or of the equilibrium end-to-end distance. The structure of the chain behaves now for each temperature as a random walk, i.e., $R \sim N^{1/2}$.

However, the presented empirical scaling arguments demand a more detailed microscopical investigation, which will be determined in a next step of our investigation on stochastic copolymer systems.

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