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Distribution function of the radius of gyration for Gaussian molecules

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An analysis of the distribution function of the radius of gyration for Gaussian molecules of arbitrary complexity imbedded in d-dimensional space is carried out. An explicit expression for the coefficients in an exact series expansion of the distribution function is obtained as finite sums of terms involving only the eigenvalues of the connectivity matrix. For odd d, a recursive method is developed for evaluating the distribution function as a sum of simple integrals. For even d, a finite-sum representation exists. The leading term of the asymptotic series for the distribution function for large values of the argument is analytically evaluated for any Gaussian molecule of arbitrary length. Moments of the distribution are given in terms of either the eigenvalue spectrum or the coefficients of the eigenpolynomial of the connectivity matrix. For a special class of molecules, the limiting distribution functions are found to share a generic form which can be represented by a double series. The length dependence of the 3D distribution function for linear chains and the dimensionality effect on the breadth of the limiting distributions for both circular and linear chains are investigated numerically. It is found that higher dimensionality tends to broaden the distribution.

INTRODUCTION

The study of the configuration statistics of macromolecules has played an important role in many areas of polymer theory.[1–3] No single model that mimics the general behavior of chain molecules in either the unperturbed or perturbed state has displaced all other contenders. The Gaussian model, however, has been widely used in the polymer field[4] due to the simplicity of the effective potential energy. The analytical description of the statistics of the model system, namely, Gaussian molecules of arbitrary complexity, has been well developed in the works of Fixman,[5] Šolc and Stockmayer,[6] and Eichinger.[7,8] This paper is based on these earlier works and is focused on the solution of the problem of the size distribution for Gaussian molecules with the use of some newly developed analytical techniques.

DISTRIBUTION FUNCTION

The distribution function of the radius of gyration for Gaussian molecules imbedded in arbitrary d-dimensional space is given by[8]

$$P(s^2)ds^2=\left(\frac{ds^2}{2\pi}\right)\int_{-\infty}^{\infty}\exp(i\beta s^2)\prod_{j=1}^{n-1}\left(1+\frac{i\beta}{\gamma n\lambda_j}\right)^{-d/2}d\beta, \tag{1}$$

where $\gamma=d/2\langle l^2\rangle_0$ with $\langle l^2\rangle_0$ the mean-square unperturbed length of a segment, n the number of beads in the molecule, and the λ_j are the nonzero eigenvalues of the Kirchhoff matrix $\mathbf{K}$ that depends only upon the architecture of the molecule. For long linear chains in 3D space, Eq. (1) reduces to the well-known Fixman formulation.[5]

We now introduce the reduced variable $\tilde{s}^2=\gamma s^2/n^\alpha$, where α is a constant so chosen such that the second moment $\langle\tilde{s}^2\rangle$ has a nonzero finite large-n limit for a given type of molecule. For linear and circular chains, for example, α takes the value 1. To take into account any degeneracy that the $n-1$ nonzero eigenvalues may have, it is convenient to order those distinct eigenvalues, numbered from 1, the smallest, to p, the largest, where $p\leqslant n-1$, each with the degeneracy ω_j with $j=1,2,\ldots,p$, and denote the product of the jth distinct eigenvalues with $n^{\alpha+1}$ by κ_j. Then, in terms of $\tilde{s}^2$ and the κ_j and ω_j, Eq. (1) becomes

$$P(\tilde{s}^2)d\tilde{s}^2=\left(\frac{d\tilde{s}^2}{2\pi}\right)|\kappa|^{d/2}\int_{-\infty}^{\infty}\exp(i\beta\tilde{s}^2)\times\prod_{j=1}^{p}(\kappa_j+i\beta)^{-d\omega_j/2}d\beta, \tag{2}$$

where $|\kappa|=\Pi_{j=1}^{p}\kappa_j^{\omega_j}$. Equation (2) represents a class of nontrivial Fourier transforms of characteristic functions having higher-multiplicity poles and/or branch points. For even $d\omega_j$ for all j, Eq. (2) can be solved with the use of the residue theorem. The resulting finite-sum representation for $P(\tilde{s}^2)d\tilde{s}^2$ is

$$P(\tilde{s}^2)d\tilde{s}^2=d\tilde{s}^2\sum_{j=1}^{p}\tilde{s}^{2(d\omega_j/2-1)}\exp(-\tilde{s}^2\kappa_j)\times\sum_{k=0}^{d\omega_j/2-1}B_{jk}/[\tilde{s}^{2k}\Gamma(d\omega_j/2-k)], \tag{3}$$

where the coefficients B_{jk} are defined as

$$B_{jk}=\frac{1}{k!}\lim_{x\to 0}\frac{d^k}{dx^k}[B_j(x)]^{d/2} \tag{4}$$

with $B_j(x)$ given by

$$B_j(x)=\kappa_j^{\omega_j}\Big/\prod_{l\neq j}^{p}[1+(x-\kappa_j)/\kappa_l]^{\omega_l}. \tag{5}$$

The B_{jk} defined in Eq. (4) may be identified as the kth coefficient in the power-series expansion of $[B_j(x)]^{d/2}$ around $x=0$. For circular and linear chains in two dimensions and circular chains with an odd number of beads in one- and three-dimensional spaces, Eq. (3) yields the results obtained by Šolc.[9,10] It is seen that for large $\bar{s}^2$, the first term in Eq. (3) involving the smallest eigenvalue only gives the leading term in an asymptotic expansion of the distribution function.

When d is odd and the $d\omega_j$ are not even for all j, complications occur as a result of the existence of branch points in the integrand in Eq. (2) if contour integration is employed. The analytic solution to the distribution function has, therefore, been obtained only for linear chains in one-dimensional space.[11] Nevertheless, a method has been developed to evaluate $P(\bar{s}^2)$ for this case. To illustrate the method, a recursive relation is first derived for evaluating $P(\bar{s}^2)d\bar{s}^2$ for linear chains for which $\omega_j = 1$ for $j = 1,2,...,p$, with $p = n - 1$. In this case, with $P(\bar{s}^2)$ denoted by $P_d(\bar{s}^2)$, one can write from Eq. (2)

$$P_{d+2}(\bar{s}^2)d\bar{s}^2 = \left(\frac{d\bar{s}^2}{2\pi}\right)|\kappa|^{d/2}\sum_{j=1}^{n-1} B_j \int_{-\infty}^{\infty} \exp(i\beta\bar{s}^2)(\kappa_j + i\beta)^{-1}\prod_{l=1}^{n-1}(\kappa_l + i\beta)^{-d/2}\, d\beta, \tag{6}$$

where $B_j = B_j(0)$. We now employ the identity

$$\frac{d}{d\bar{s}^2}\int_{-\infty}^{\infty}\frac{\exp[\bar{s}^2(\kappa_j + ix)]}{\kappa_j + ix} f(x)dx = \int_{-\infty}^{\infty}\exp[\bar{s}^2(\kappa_j + ix)]\, f(x)dx, \tag{7}$$

where $f(x) = \Pi_{j=1}^{n-1}(\kappa_j + ix)^{-d/2}$. Equation (6) then becomes

$$P_{d+2}(\bar{s}^2)d\bar{s}^2 = d\bar{s}^2\sum_{j=1}^{n-1} B_j \int_0^{\bar{s}^2}\exp[(\beta - \bar{s}^2)\kappa_j]P_d(\beta)d\beta \tag{8}$$

or, in an equivalent form,

$$P_{d+2}(\bar{s}^2)d\bar{s}^2 = d\bar{s}^2\bar{s}^2\int_0^1 d\beta\, P_d[\bar{s}^2(1-\beta)]\sum_{j=1}^{n-1} B_j \exp(-\bar{s}^2\kappa_j\beta). \tag{9}$$

From the above relation, one can easily obtain $P_3(\bar{s}^2)d\bar{s}^2$ for linear chains with an arbitrary number of beads from the known solution for $P_1(\bar{s}^2)d\bar{s}^2$, which, for odd n, is given by Coriell and Jackson.[11] A plot of $P_3(\bar{s}^2)$ vs $\bar{s}^2$ for $n = 11$ calculated according to Eq. (9) is shown in Fig. 1. In general, when ω_j is not equal to 1 for all j, the above procedure may be readily modified to evaluate first $P_1(\bar{s}^2)$ for any Gaussian molecule, from which $P_d(\bar{s}^2)$ for $d \geqslant 3$ may then be obtained analogously.

With two cases, one for even d and the other for odd d, treated separately above, we now seek a series-expansion solution for $P(\bar{s}^2)$ as an unified descriptive representation other than the parent Fourier transform. This approach was first employed by Eichinger with the first four coefficients worked out by a recursive method.[8] To provide an explicit epxression for the coefficients in the expansion, we first note that with a change of variable Eq. (2) can be put into the form

$$P(\bar{s}^2)ds^2 = \left(\frac{d\bar{s}^2}{2\pi}\right)|\kappa|^{d/2}\bar{s}^{2[d(n-1)/2-1]}\int_{-\infty}^{\infty}\exp(i\beta) \times \prod_{j=1}^{p}(\bar{s}^2\kappa_j + i\beta)^{-d\omega_j/2}\, d\beta. \tag{10}$$

Upon expanding the product in Eq. (10) as a power series in $\bar{s}^2$ and carrying out the integration for each coefficient of the resulting series, we find

$$P(\bar{s}^2)d\bar{s}^2 = \left(\frac{d\bar{s}^2}{C}\right)\bar{s}^{2[d(n-1)/2-1]}\left[1 + \sum_{m=1}^{\infty}(-1)^m a_m \bar{s}^{2m}\right], \tag{11}$$

where $C = |\kappa|^{-d/2}\Gamma[d(n-1)/2]$ and the coefficients a_j are given by

$$a_j = \sum_{l_1=0}^{l_0}\sum_{l_2=0}^{l_1}\cdots\sum_{l_m=0}^{l_{m-1}}\prod_{k=1}^{p}(l_{k-1} - l_k + 1)_{d\omega_k/2-1}\frac{\kappa_k^{l_{k-1}-l_k}}{\Gamma(d\omega_k/2)}, \tag{12}$$

where $m = p - 1$, $l_0 = j$, $l_p = 0$, $(a)_n = \Gamma(a+n)/\Gamma(a)$ and $\Gamma(x)$ is the Gamma function. Note that the first term is in exact agreement with Eichinger's result.[8]

To obtain an asymptotic expansion for $P(\bar{s}^2)$, we make a different change of variable in Eq. (2) and obtain

$$P(\bar{s}^2)ds^2 = \left(\frac{d\bar{s}^2}{2\pi}\right)|\kappa|^{d/2}\bar{s}^{2(d\omega_1/2-1)}\exp(-\bar{s}^2\kappa_1) \times \int_{-\infty-iR}^{\infty-iR}(i\beta)^{-d\omega_1/2}\exp(i\beta)\prod_{j=2}^{p}\left(\kappa_j - \kappa_1 + \frac{i\beta}{\bar{s}^2}\right)^{-d\omega_j/2} d\beta, \tag{13}$$

where $R = \bar{s}^2\kappa_1$. For large $\bar{s}^2$, the integral in Eq. (13) depends only upon the inverse of $\bar{s}^2$ and may therefore be expanded in $1/\bar{s}^2$, with the result

$$P(\bar{s}^2)ds^2 = \left(\frac{d\bar{s}^2}{2\pi}\right)|\kappa|^{d/2}\bar{s}^{2(d\omega_1/2-1)}\exp(-\bar{s}^2\kappa_1)\sum_{m=0}^{\infty}\frac{b_m}{\bar{s}^{2m}} \tag{14}$$

where b_m is defined through its generating function $G(x)$ given by

$$G(x) = \lim_{R\to\infty} \int_{-\infty-iR}^{\infty-iR} (i\beta)^{-d\omega_1/2} \exp(i\beta) \times \prod_{j=1}^{p} (\kappa_j - \kappa_1 + i\beta x)^{-d\omega_j/2}\, d\beta, \tag{15}$$

which is comparable to the multidimensional integral representation for the coefficients given by Eichinger.[8] Though no simple analytical expression for b_m exists other than for special cases, such as dumbbell molecules, Eq. (15) does permit the leading term to be explicitly expressed in terms of the eigenvalues of the Kirchhoff matrix, i.e.,

$$b_0 = 2\pi(B_1/|\kappa|)^{d/2}/\Gamma(d\omega_1/2) \tag{16}$$

which represents a generalization to the earlier large-n-limit results.[5,8,11-13]

Finally, we note that general behavior of the distribution functions near their maxima may be shown to be Gaussian[14]:

$$P(\tilde{s}^2)d\tilde{s}^2 = (d\tilde{s}^2/\sqrt{2\pi\sigma^2})\exp[-(\tilde{s}^2 - \langle\tilde{s}^2\rangle)^2/2\sigma^2], \tag{17}$$

where $\sigma^2 = \langle\tilde{s}^4\rangle - \langle\tilde{s}^2\rangle^2$. In particular, $P(\langle\tilde{s}^2\rangle) = (2\pi\sigma^2)^{-1/2}$, which turns out to be a very good approximation to the exact value of the distribution function evaluated at that argument.

MOMENTS OF THE DISTRIBUTION

It is well known that moments are important parameters that characterize a distribution. In fact, certain systems of distributions are uniquely determined if their moments are known up to an arbitrary positive order.[15,16] In polymer theory, knowledge of the second and fourth moments of the distribution of the radius of gyration plays an important role in characterizing the viscoelastic properties of polymers.[17]

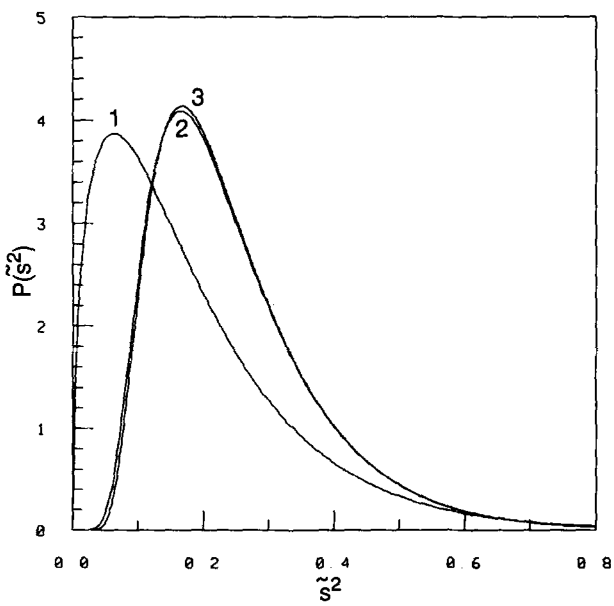

FIG. 1. The distribution functions for linear chains in three dimensional space with $n = 2$ and 11 corresponding to curves 1 and 2, respectively. Curve 3 is for the limiting distribution function.

For the distribution under consideration, the moment of order m may be written down as the mth derivative of the characteristic function evaluated at zero argument[5,11,4]:

$$\langle\tilde{s}^{2m}\rangle = \lim_{\beta\to 0} \frac{d^m}{d\beta^m} D(i\beta), \tag{18}$$

where $D(\beta)$ is given by

$$D(\beta) = \prod_{j=1}^{p} \left(1 + \frac{i\beta}{\kappa_j}\right)^{-d\omega_j/2} \tag{19}$$

or, in terms of the eigenpolynomial of the Kirchhoff matrix $\mathbf{K}$,[18,19] by

$$D(\beta) = \left[\alpha_{n-1} \Big/ \sum_{j=1}^{n-1} \alpha_j \left(\frac{-i\beta}{n^2}\right)^{n-j-1}\right]^{d/2}, \tag{20}$$

where the α_j's are the coefficients of the polynomial. To carry out the higher derivative, use is made of the Leibnitz's rule[20] when $D(\beta)$ is given by Eq. (19), yielding

$$\langle\tilde{s}^{2j}\rangle = j! \sum_{l_1=0}^{l_0} \sum_{l_2=0}^{l_1} \cdots \sum_{l_{m}=0}^{l_{m-1}} \prod_{k=1}^{p} \left(\frac{d\omega_k}{2}\right)_{l_{k-1}-l_k} \frac{\kappa_k^{l_{k-1}-l_k}}{(l_{k-1}-l_k)!} \tag{21}$$

where $m = p-1$, $l_0 = j$, and $l_p = 0$. For $j = 1$, Eq. (21) gives the known result: $\langle\tilde{s}^2\rangle = (d/2)\Sigma_{j=1}^{p} \omega_j\kappa_j^{-1}$.

By employing the general relation for the higher derivative of a composite function,[20] one arrives at

$$\langle\tilde{s}^{2m}\rangle = \left(\frac{m!}{n^{2m}}\right) \sum \left(\frac{d}{2}\right)_j \prod_{i=1}^{l} \left(\frac{-\alpha_{n-1-i}}{\alpha_{n-1}}\right)^{j_i} \Big/ j_i! \tag{22}$$

for $D(\beta)$ given by Eq. (20), where the symbol Σ indicates summation over all solutions in nonnegative integers of the equation $\Sigma_{i=1}^{l} j_i = j$ and $\Sigma_{i=1}^{l} ij_i = m$. Note also that $\alpha_i = 0$ for $i < 0$. For $m = 1$, Eq. (22) reduces to the result obtained by Yang and Yu.[19] Equation (22) is useful when analytic expressions for the eigenvalues of the Kirchhoff matrix are difficult to obtain.

Before leaving this section, we note that the cumulants χ_m of the distribution can be easily identified by writing $D(-ia)$ in exponential form and then expanding the logarithm in the exponent. The result is simple[4,21]:

$$\chi_m = \left(\frac{d}{2}\right)(m-1)! \sum_{j=1}^{p} \omega_j \kappa_j^{-m} \tag{23}$$

which may then be used to obtain the moments by the well-known relation between the two quantities, yielding[16]

TABLE I. Definitions of ϵ, p, and q for limiting distribution functions.

ϵ	p	q
Rings		
4	d	0
Double rings		
16	$3d/2$	$d/2$
Linear chains		
1	$d/2$	0
Star molecules		
f^2	$d/2$	$(f-1)d/2$

$$\langle \tilde{s}^{2m} \rangle = \chi_m + \cdots + [\tilde{s}^2]^m \tag{24}$$

which, unfortunately, are not as convenient to use as Eq. (21) when higher moments are desired. For $\langle \tilde{s}^m \rangle$ where m is odd, no simple solution exists. Nevertheless, they may well be calculated from the analytical solutions to $P(\tilde{s}^2)d\tilde{s}^2$.

LIMITING DISTRIBUTION FUNCTION

The distribution functions for large n deserve special attention. Historically, it was the very interest in the size characterization of polymers that initiated the study of the configuration statistics for Gaussian molecules.[5,4]

In general, to obtain the limiting distribution function, one simply takes the large-n limit on the expression for $P(\tilde{s}^2)d\tilde{s}^2$.[10,11] In what follows, we focus on a special class of molecules which share a limiting distribution function of the form

$$P(\alpha)d\alpha = \left(\frac{d\alpha}{2\pi}\right)\int_{-\infty}^{\infty} \exp(i\alpha\beta)\left(\frac{\sin Z}{Z}\right)^{-p}(\cos Z)^{-q}\,d\beta, \tag{25}$$

where $\alpha = \epsilon\tilde{s}^2$ with $\tilde{s}^2 = \gamma s/n$, and $Z^2 = -i\beta$. Included in this class are linear chains, rings, double rings, and star-shaped molecules with f arms of equal length,[4,5,19] for which the values of ϵ, p, and q are summarized in Table I.

Integrals of the type in Eq. (25) were first solved analytically by Fujita and Norisuye for $p = 3/2$ and $q = 0$.[12] For other values of p and q, we find that the contour devised by these authors for their specific case is still useful for the evaluation of Eq. (25). Therefore, their method may be readily applied to solve Eq. (25) to obtian

$$P(\alpha)d\alpha = \left(\frac{d\alpha}{\pi}\right)2^{p+q-1}\alpha^{-(p+2)} \times \sum_{m,n=0}^{\infty}(-1)^n\frac{(p)_m(q)_n}{m!n!} \times \mathrm{Re}\int_0^{\infty}\exp\left[\frac{-\gamma_{mn}(\xi^2+1)}{\alpha}\right] \times (1+i\xi)^{p+1}\,d\xi, \tag{26}$$

where $\gamma_{mn} = [n + m + (p+q)/4]^2$. The integral in Eq. (26) can be evaluated analytically. For $2p$ odd, the integral may be expressed in terms of the modified Bessel function of the second kind as shown by Fujita and Norisuye.[12] Here a different procedure is used, yielding

$$P(\alpha)d\alpha = \left(\frac{d\alpha}{\pi}\right)2^{p+q}\alpha^{-(p/2+1)} \times \sum_{m,n=0}^{\infty}(-1)^n\frac{(p)_m(q)_n}{m!n!}W(\gamma_{mn}/\alpha), \tag{27}$$

where $W(\alpha) = A_{p/2+1}(\alpha)\exp(-\alpha)$ and $A_\nu(x)$, with $\nu = p/2+1$, is defined as

$$A_\nu(x) = \begin{cases} \displaystyle\sum_{j=1}^{[\nu-1/2]}(-1)^j\binom{2\nu-1}{2j}\Gamma(j+1/2)x^{\nu-j-1/2}, & \text{for } 2p \text{ even} \\ \Gamma(\nu)M(1/2-\nu,1/2,x)\sin(\pi\nu) & \\ \quad -2\sqrt{x}\,\Gamma(\nu+1/2)M(1-\nu,3/2,x)\cos(\pi\nu), & \text{for } 2p \text{ odd} \end{cases}. \tag{28}$$

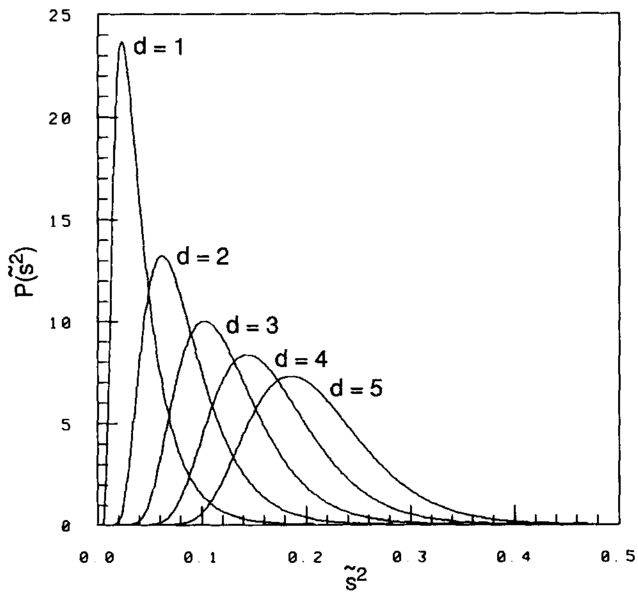

FIG. 2. Dimensionality dependence of the limiting distribution functions for circular chains. The dimensionality of space is d.

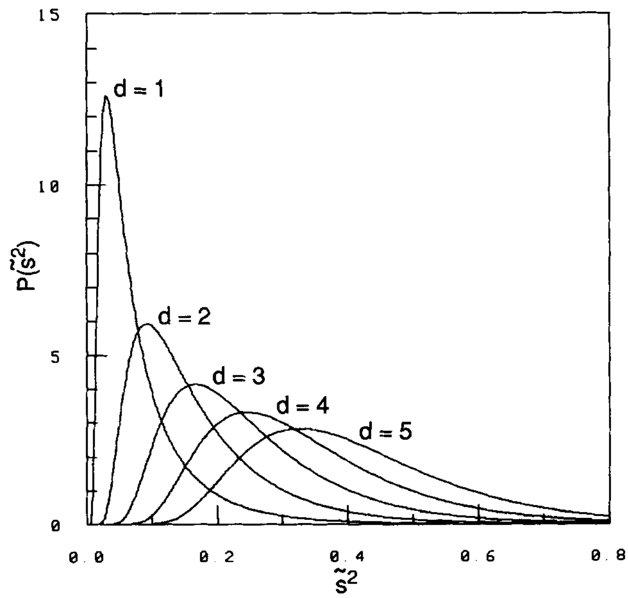

FIG. 3. Dimensionality dependence of the limiting distribution functions for linear chains. The dimensionality of the space is d.

TABLE II. Numerical results from distribution functions for linear chains in three dimensions.

n	$^*\bar{s}^2$	$P(^*\bar{s}^2)$	$\langle \bar{s}^2 \rangle$
2	0.063	3.87	0.188
11	0.165	4.09	0.248
∞	0.166	4.14	0.250

In Eq. (28), $M(a,c,x)$ is the confluent hypergeometric function.[20] For small α, Eq. (26) becomes

$$P(\alpha)d\alpha = d\alpha\pi^{-1/2}2^{q-1}p^{p+1}\alpha^{-(p+3/2)} \times \exp(-p^2/4\alpha)[1+O(\alpha)]. \tag{29}$$

The leading term in Eq. (29) reproduces the earlier results obtained for linear chains by various methods.[5,12,13] For an asymptotic expansion at large α, one may introduce an auxiliary function in the defining equation, Eq. (25), for $q=0$, following the procedure employed by Fujita and Norisuye[12] to obtain for arbitrary p

$$P(\alpha)d\alpha = d\alpha[(2\pi^2)^p/\Gamma(p)]\alpha^{p-1} \times \exp(-\pi^2\alpha)[1+O(1/\alpha)]. \tag{30}$$

It must be noted that the higher order terms in Eq. (30) yield simple expressions only for $d=1$ and for linear chains at $d=1,2,3$, plus all higher values of odd d.

To obtain the cumulants χ_k for the distribution, one identifies

$$D(\beta) = [\sin(Z)/Z]^{-p}(\cos Z)^{-q}$$

as the characteristic function in Eq. (25). It then follows that

$$\ln D(-i\beta) = \sum_{k=1}^{\infty} \frac{(-1)^k B_k}{k(2k)!}\left[q - 4^k\left(q+\frac{p}{2}\right)\right](-\beta)^k, \tag{31}$$

where the B_n are Bernoulli numbers.[20] From Eq. (31), we have

$$\chi_k = (-1)^k(k-1)![q-4^k(q+p/2)]B_{2k}/(2k)!. \tag{32}$$

For linear and circular chains in three dimensions, Eq. (32) yields $\langle \bar{s}^2 \rangle = 1/4$ and $1/8$, respectively.

The plots of $P(\bar{s}^2)$ vs $\bar{s}^2$, based on Eq. (27), are shown in Figs. 2 and 3. Tables II and III contain some numerical results for most probable configurations. It is seen from Fig. 1 that for linear chains in three-dimensional space the limiting distribution is rapidly approached as n increases. In the asymptotic region, linear chains with an arbitrary number of beads behave like flexible dumbbells, as expected. Figures 2 and 3 show the dimensionality effect on the breadth of the distribution curves. As is seen, the distribution is broadened as d increases. This broadening effect simply confirms the speculation that the fluctuations away from the most probable configurations tend to spread out when more spatial directions become available for the imbedded molecules.

TABLE III. Numerical results from limiting distribution functions for circular and linear chains in low dimensionality spaces.

d	$^*\bar{s}^2$	$P(^*\bar{s}^2)$	$\langle \bar{s}^2 \rangle$
Circular			
1	0.023	23.69	0.042
2	0.061	13.24	0.083
3	0.103	10.01	0.125
4	0.143	8.36	0.167
5	0.185	7.33	0.208
Linear			
1	0.030	12.60	0.083
2	0.092	5.92	0.167
3	0.166	4.14	0.250
4	0.245	3.31	0.333
5	0.326	2.83	0.417

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