

## Simple Cell Model with Collapse Instability

F. H. Stillinger<sup>1</sup>

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A cell model is introduced in which pairs of particles interact only within the same cell, and then only with a constant coupling  $\phi_0$ . For positive  $\phi_0$  the statistical thermodynamics is normal, but as  $\phi_0$  changes sign, the system manifests a collapse phenomenon with all particles tending to aggregate in the same cell. This collapse instability causes high-temperature series to diverge, but known asymptotic properties of Stirling numbers of the second kind allow one to establish Borel summability. The present model is equivalent to continuum models with bounded pair potentials when in the latter the space dimension  $D$  is permitted to go to  $0+$ .

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**KEY WORDS:** High-temperature expansions; divergent series; Borel summability; cell model; collapse instability; Stirling numbers; communal entropy.

### 1. INTRODUCTION

Within the realm of classical statistical mechanics the study of high-temperature series has been an effective tool for understanding equilibrium properties. This technique has been applied both to discrete lattice systems of the Ising type,<sup>(1)</sup> and to continuous systems comprising molecules with hard cores.<sup>(2)</sup> For these types of systems it appears that the high-temperature expansions of the usual thermodynamic properties are series with non-vanishing radii of convergence, provided that intermolecular forces have short range.

In order to simplify the otherwise formidable job of evaluating high-order series coefficients for continuous systems, it is advantageous to have pair potentials consisting of finite sums of Gaussians. The simplest of these involves just one Gaussian component, and yields the "Gaussian core model" that has already been extensively studied.<sup>(3-5)</sup> However, any finite sum of Gaussians is a bounded potential, and evidence has been presented indicating that in such a circumstance the high-temperature expansions of thermo-

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<sup>1</sup> Bell Laboratories, Murray Hill, New Jersey.

dynamic properties have vanishing radii of convergence.<sup>(6)</sup> This is connected with the fact that a formal change in sign of the bounded pair potential leads to a collapse instability of the system, as a result of which all particles in the system become virtually coincident. In the complex plane of the variable  $\beta = 1/k_B T$ , all thermodynamic properties then apparently possess essential singularities at  $\beta = 0$ .

Hence it is desirable to characterize the mathematical implications of the collapse instability in sufficient detail that asymptotic high-temperature series can be properly interpreted. That is the motivation behind the present study. For the sake of economy a conceptually simple cell model has been constructed and analyzed, in the belief that collapse instability and its effects are largely insensitive to specific details of the bounded interactions that cause them. We conclude that the naturally occurring high-temperature series for the model is Borel summable.

Section 2 defines the cell model. Section 3 concerns construction and numerical investigation of its grand partition function, from which thermodynamic properties follow. Section 4 examines the collapse instability and how it influences the asymptotic magnitude of high-order series coefficients; these are the results which assure Borel summability. Finally, Section 5 presents discussion of this and related problems.

## 2. CELL MODEL

Let the volume  $V$  be divided into a number  $\Omega$  of equivalent cells, each with volume

$$\omega = V/\Omega \quad (2.1)$$

When  $N$  point particles inhabit  $V$  at specific positions  $\mathbf{r}_1, \dots, \mathbf{r}_N$  there will be a potential energy  $\Phi(\mathbf{r}_1, \dots, \mathbf{r}_N)$  describing interactions in the system. We suppose that  $\Phi$  has a particularly simple form, namely that it consists only of pair interactions which vanish unless both particles involved reside in the same cell; and furthermore the intracell pair interaction is just a constant:

$$\phi_0 > 0 \quad (2.2)$$

If  $\phi_0$  is much larger than the thermal energy  $k_B T = 1/\beta$ , then  $\Phi$  acts so as to inhibit multiple occupancy in any of the cells. But if  $\phi_0$  and  $1/\beta$  are comparable, multiple occupancy becomes energetically feasible. When  $\phi_0$  is changed from positive to negative, crowding of many particles without bound into the same cell is encouraged, and it is this instability which makes the cell model worth examining.

For any set of particle positions  $\mathbf{r}_1, \dots, \mathbf{r}_N$  there will be a corresponding set of cell occupation numbers  $n_1, \dots, n_\Omega$ , where an arbitrary numbering scheme

has been assumed for the cells. Obviously

$$N = \sum_{i=1}^{\Omega} n_i \quad (2.3)$$

Furthermore

$$\Phi = \frac{1}{2} \phi_0 \sum_{i=1}^{\Omega} n_i (n_i - 1) \quad (2.4)$$

As we shall see, collapse instability in this and related models is intimately connected to the quadratic nature of  $\Phi$  expressed in local occupation numbers, for this implies an ability for properties that are normally extensive to behave in an abnormal, nonextensive manner.

It has been brought to the author's attention that this cell model is structurally identical to one that Hertel and Thirring examined to study gravitational condensation.<sup>(7)</sup> However, these authors impose important constraints, namely that  $N$  be fixed, and that the intracell coupling  $\phi_0$  be real and negative and inversely proportional to  $N$ . None of these constraints apply to the present work, and the respective results are essentially unrelated.

### 3. GRAND PARTITION FUNCTION

The most natural way to examine the statistical mechanics of the cell model is by using the grand ensemble. The grand partition function is

$$Z_G(\beta, y) = \sum_{N=0}^{\infty} (y^N / N!) \int d\mathbf{r}_1 \cdots \int d\mathbf{r}_N \exp(-\beta\Phi) \quad (3.1)$$

where  $y$  is the absolute activity, and where the integrations span the system volume  $V$ . Connection to the thermodynamic pressure  $p$  is established by the formula

$$\beta p V = \ln Z_G \quad (3.2)$$

while the mean number of particles in the system and the mean potential energy are given by

$$\langle N \rangle = (\partial \ln Z_G / \partial \ln y)_{\beta} \quad (3.3)$$

$$\langle \Phi \rangle = -(\partial \ln Z_G / \partial \beta)_y \quad (3.4)$$

Due to the simple form chosen for  $\Phi$ , the grand partition function reduces to a product of identical factors, one for each cell:

$$Z_G(\beta, y) = [f(u, v)]^{\Omega} \quad (3.5a)$$

$$f(u, v) = \sum_{n=0}^{\infty} (1/n!) \exp(-un - vn^2) \quad (3.5b)$$

$$u = -\ln(\omega y) - \frac{1}{2}\beta\phi_0 \quad (3.5c)$$

$$v = \frac{1}{2}\beta\phi_0 \quad (3.5d)$$

In terms of the function  $f$  the reduced thermodynamic properties of the model are determined by the following expressions:

$$\beta p \omega = \ln f \quad (3.6)$$

$$\langle N \rangle / \Omega = \langle n \rangle = -(\partial \ln f / \partial u)_v \quad (3.7)$$

$$2\langle \Phi \rangle / \phi_0 \Omega + \langle N \rangle / \Omega = -(\partial \ln f / \partial v)_u \quad (3.8)$$

These can be supplemented by an expression for  $S$ , the entropy per cell, relative to its value  $S_0$  for noninteracting particles at the same density:

$$\frac{S - S_0}{k_B \Omega} = \ln f - \left( \frac{\partial \ln f}{\partial \ln u} \right)_v - \left( \frac{\partial \ln f}{\partial \ln v} \right)_u + \langle n \rangle \ln \langle n \rangle - \langle n \rangle \quad (3.9)$$

It is clear from Eq. (3.5b) that the sum defining  $f$  fails to converge when  $v < 0$ . For  $v = 0$  the sum is elementary, yielding

$$f(u, 0) = \exp[\exp(-u)] \quad (3.10)$$

The case  $v > 0$  does not produce elementary functions, but the sums for  $f$  and its derivatives are rapidly converging, so that numerical study is straightforward and convenient.

Table I presents a selection of thermodynamic properties calculated for  $\langle n \rangle = 1$ . The excess entropy (3.9) at this density has been plotted against the mean interaction energy in Fig. 1. The end points of this curve refer respectively to strict single occupancy ( $2\langle \Phi \rangle / \phi_0 \Omega = 0$ ) and to independent occupancy ( $2\langle \Phi \rangle / \phi_0 \Omega = 1$ ), and the corresponding entropy change has often been called "communal entropy."<sup>(8)</sup>

**Table I. Cell Model Properties for  $\langle n \rangle = 1$**

$u$	$v$	$\ln f$	$2\langle \Phi \rangle / \phi_0 \Omega$	$(S - S_0) / k_B \Omega$
0	0	1	1	1
-0.1	0.0340438	1.0330000	0.9380128	0.9989773
-0.5	0.1822228	1.1607909	0.7528607	0.9802021
-1.0	0.3880839	1.3171483	0.5979509	0.9372873
-1.5	0.6104552	1.4748564	0.4871796	0.8827129
-2.0	0.8442916	1.6374886	0.4022383	0.8213866
-3.0	1.3309898	1.9876593	0.2763570	0.6864774
-5.0	2.3269321	2.8023841	0.1213836	0.4117677
-7.0	3.3267241	3.7228141	0.0483328	0.2103281
-10.0	4.8267133	5.1845548	0.0112049	0.0653508
-15.0	7.3267131	7.6742166	0.0009293	0.0077385

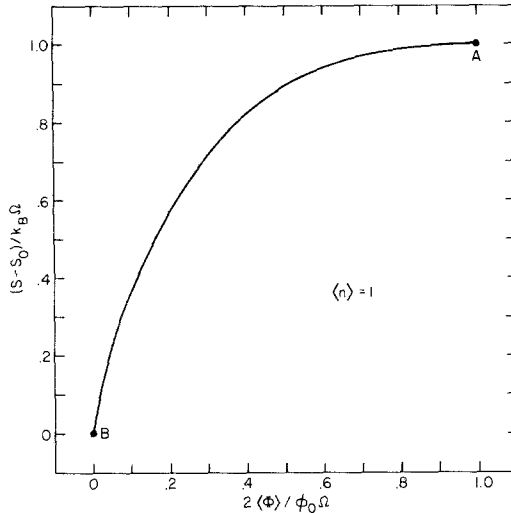


Fig. 1. Excess entropy per cell at  $\langle n \rangle = 1$  plotted vs. reduced interaction strength. Point A is the position of incipient collapse, while point B corresponds to single occupancy. Thermodynamic states with real, positive temperatures lie on the curve between A ( $T = \infty$ ) and B ( $T = 0$ ).

#### 4. COLLAPSE INSTABILITY

In order to examine the collapse instability it will be advantageous to generate the formal power series for  $f(u, v)$  in its second variable  $v$ , which measures the interaction strength in the system. From (3.5b) we have

$$\begin{aligned}
 f(u, v) &\sim \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \frac{(-v)^m n^{2m}}{m! n!} \exp(-un) \\
 &= \sum_{m=0}^{\infty} \frac{(-v)^m}{m!} \left(-\frac{d}{du}\right)^{2m} \exp[\exp(-u)]
 \end{aligned}
 \tag{4.1}$$

The question of whether this last series converges depends clearly on the behavior of the indicated derivatives as  $m \rightarrow \infty$ .

We can write

$$\left(-\frac{d}{du}\right)^{2m} \exp[\exp(-u)] = S_{2m}[\exp(-u)] \exp[\exp(-u)]
 \tag{4.2}$$

where  $S_{2m}$  is a polynomial with degree  $2m$ . The lowest order cases are found to be

$$S_0(x) = 1, \quad S_2(x) = x + x^2, \quad S_4(x) = x + 7x^2 + 6x^3 + x^4
 \tag{4.3}$$

If the general case is written thus (for  $p > 0$ )

$$S_p(x) = \sum_{q=1}^p S_p^{(q)} x^q
 \tag{4.4}$$

it follows from Eq. (4.2) directly that the coefficients obey the following recursion relation:

$$S_{p+1}^{(q)} = qS_p^{(q)} + S_p^{(q-1)} \quad (4.5)$$

Consequently the  $S_p^{(q)}$  can be identified as Stirling numbers of the second kind,<sup>(9)</sup> for which extensive tables exist. All nonvanishing  $S_p^{(q)}$  are positive.

We thus have

$$f(u, v) \sim \exp[\exp(-u)] \sum_{m=0}^{\infty} \frac{(-v)^m}{m!} S_{2m}[\exp(-u)] \quad (4.6)$$

The convergence of this power series in  $v$  is controlled by the large- $m$  behavior of

$$R_m(u) = S_{2m}[\exp(-u)]/mS_{2m-2}[\exp(-u)] \quad (4.7)$$

i.e., the ratios of successive series coefficients. These ratios have been studied numerically to moderately high order. Figure 2 presents some typical results for  $m \leq 30$  and  $u = -1, 0, 1$ . In all cases that have been examined the  $R_m$  show an unmistakable trend toward infinity with increasing  $m$ . The ratio test for series<sup>(10)</sup> then would imply that series (4.6) has a vanishing radius of convergence.

The properties of Stirling numbers of the second kind have been presented in the mathematical literature with considerable detail.<sup>(11)</sup> These properties allow one to demonstrate that indeed the  $R_m$  diverge with  $m$ :

$$R_m \sim \frac{4m}{(\ln m)^2} \left[ 1 + O\left(\frac{1}{\ln m}\right) \right] \quad (4.8)$$

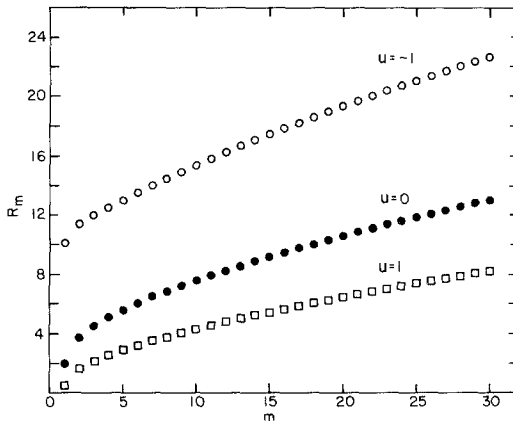


Fig. 2. Coefficient ratios for asymptotic expansion (4.6). The  $R_m$  are defined by Eq. (4.7).

The details are contained in the Appendix.

Fortunately the divergent series (4.6) is Borel summable.<sup>(1,2)</sup> If we write

$$f(u, v) = \int_0^\infty \exp(-t) g(u, vt) dt \quad (4.9)$$

then  $g$  is the Borel transform of  $f$  with respect to its second variable  $v$ . Corresponding to the divergent series (4.6) for  $f$  we have a convergent series for  $g$ :

$$g(u, s) = \exp[\exp(-u)] \sum_{m=0}^{\infty} \frac{(-s)^m}{(m!)^2} S_{2m}[\exp(-u)] \quad (4.10)$$

The coefficient ratios for this transform are  $R_m/m$ , and since these converge to zero with increasing  $m$ , we can be certain that sum (4.10) converges throughout the complex  $s$  plane.

The Borel transform evidently has banished the essential singularity at the origin of the  $v$  plane (due to the collapse instability) to the point at infinity in the  $s$  plane. It is for this reason that the Borel transform is a vital tool for interpreting high-temperature series, for without it the mathematical effects of collapse instability can completely obscure information about physical properties (such as phase transitions in some models) that occur away from  $\beta = 0$ .

## 5. DISCUSSION

While the variables  $u$  and  $v$  are the natural choice for discussing the properties of the present cell model, it is also valuable to consider alternatives. In particular one can inquire about the high-temperature series at fixed density. One approach would employ Eq. (3.7) to eliminate  $u$  from  $f$ ; however, this involves a cumbersome series reversion. Alternatively, one can start with the *canonical* partition function for a fixed number  $N$  of particles [cf. Eq. (3.1)]:

$$Z_N(\beta) = \frac{1}{\lambda_T^{DN} N!} \int d\mathbf{r}_1 \cdots \int d\mathbf{r}_N \exp(-\beta\Phi) \quad (5.1)$$

where  $\lambda_T$  is the mean thermal de Broglie wavelength and  $D$  is the space dimension. Then established procedures can be employed to develop  $\ln Z_N$ , the Helmholtz free energy times  $-\beta$ , in a  $\beta$  series.

We will not dwell upon the details of this last expansion technique, but only note that it closely parallels the analogous procedure that has been already carried out for the Gaussian core model.<sup>(6)</sup> Not only are the irreducible cluster graphs encountered the same topologically, but interest-

ingly the values to be assigned to those graphs for the present case correspond exactly to setting  $D = 0$  in the Gaussian core model; this reduction demands that density  $\rho$  for the present cell model be taken as  $\langle N \rangle / \Omega$  and that  $k_B T$  be measured in units  $\phi_0$ . The excess Helmholtz free energy  $F_N^{(ex)}$  per particle in the conventional infinite-system limit is thus found to be

$$\lim_{N \rightarrow \infty} \frac{\beta F_N^{(ex)}(\beta)}{N} = b_{12}(0)\rho\beta\phi_0 - \sum_{n=2}^{\infty} \left[ \sum_{j=2}^n b_{nj}(0)\rho^{j-1} \right] (-\beta\phi_0)^n \quad (5.2)$$

Table I of Ref. 6 lists all  $b_{nj}(D)$  for  $n \leq 8$ .

Just as the ratios  $R_m$ , Eq. (4.7), were useful in analyzing convergence properties of  $f$ , so too are the corresponding ratios

$$r_n(\rho) = \left[ \sum_{j=2}^n b_{nj}(0)\rho^{j-1} \right] / \left[ \sum_{j=2}^{n-1} b_{n-1,j}(0)\rho^{j-1} \right] \quad (5.3)$$

useful for analyzing the convergence properties of the series (5.2). Figure 3 shows a plot of these ratios for  $n \leq 8$  and  $\rho = e^{-1}, 1, e$ . Although these calculations unfortunately cannot be carried to the same high order as shown for the previous case in Fig. 2, it may be significant that the beginnings of the same qualitative trends seem to be present. This gives rise to the speculation that the same type of essential singularity may be present in the complex  $\beta\phi_0$  plane for the constant- $\rho$  expansion of Helmholtz free energy as we encountered earlier with constant- $u$  expansion of the grand partition function. This in turn implies that the Borel transform is also appropriate for interpretation of constant- $\rho$  series.

The argument can be carried a bit further, though with less certainty. Our

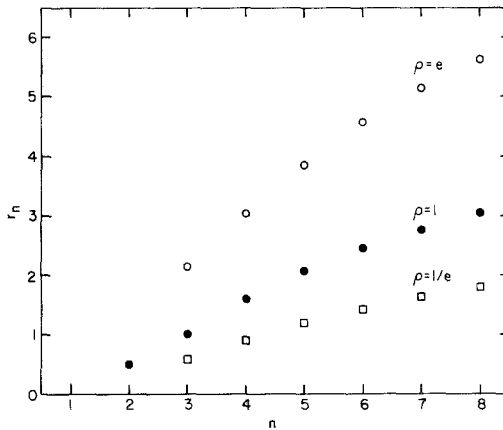


Fig. 3. Coefficient ratios [Eq. (5.3)] for the constant-density expansion of the excess Helmholtz free energy. The point  $r_2 = \frac{1}{2}$  is common to all densities.



cell model is in fact the zero-dimensional limit for *any* classical system of particles with bounded pair interactions. One can show from the available theory of arbitrary- $D$  spaces<sup>(13)</sup> that the limit  $D \rightarrow 0+$  concentrates “volume” strongly at the origin so that interacting particles only experience the pair potential at zero separation. To the extent that the present work justifies use of Borel transforms as the appropriate tool for handling the collapse instability at least when  $D = 0$ , it appears then reasonable that the same technique might be appropriate for this general family of models when  $D > 0$ .

With respect to collapse instability and its series implications, it appears that system dimension  $D$  plays far less important a role than does the pairwise additivity of the interaction. Series divergence in Section 3 can readily be traced to the fact that the number of simultaneously interacting particles in a cell  $n(n-1)/2$  grows quadratically with occupation number  $n$ . When  $\phi_0 < 0$  the quadratic plunge of cell potential to  $-\infty$  then cannot be inhibited by entropy effects (which are only linear in  $n$ ). Nevertheless, we have seen that the resulting collapse singularity remains mathematically tractable.

However, suppose that the potential energy within a cell increases in magnitude with  $n$  more rapidly than quadratically. In particular, replacement of Eq. (2.4) by the following cubic alternative (corresponding roughly to three-body interactions):

$$\Phi = \frac{1}{2}\phi_0 \sum_{i=1}^{\Omega} n_i(n_i^2 - 1) \quad (5.4)$$

causes simple but profound changes in the foregoing analysis. Equation (3.5b) defining  $f(u, v)$  must then be replaced by

$$f_3(u, v) = \sum_{n=0}^{\infty} (1/n!) \exp(-un - vn^3) \quad (5.5)$$

and the asymptotic  $v$  expansion (4.6) becomes

$$f_3(u, v) \sim \exp[\exp(-u)] \sum_{m=0}^{\infty} \frac{(-v)^m}{m!} S_{3m}[\exp(-u)] \quad (5.6)$$

From this it follows that

$$R_m(u) \sim \frac{27m^2}{(\ln m)^3} \left[ 1 + O\left(\frac{1}{\ln m}\right) \right] \quad (5.7)$$

and because these grow faster than linearly with  $m$ , a single Borel transform operation will not convert the divergent series to a convergent one.

In connection with this last point it would be interesting to construct and study a classical continuum model with specific three-body interactions. If these were chosen to be Gaussian functions of the triplet relative configuration

variables, the irreducible clusters required for high-temperature series (at constant  $\rho$ ) could again be obtained in closed form. It would then be possible to check if leading-order coefficients seemed to obey the qualitative pattern indicated by Eq. (5.7).

## APPENDIX

Moser and Wyman<sup>(11)</sup> have derived the asymptotic behavior of the numbers  $S_p^{(q)}$  for  $p$  and  $q$  both large. In leading order their results state

$$S_p^{(q)} \sim \frac{p! (e^R - 1)^q}{2R^p q! (\pi q R H)^{1/2}} [1 + O(q^{-1})] \quad (\text{A.1})$$

where

$$R/(1 - e^{-R}) = p/q \quad (\text{A.2})$$

and

$$H = e^R(e^R - 1 - R)/2(e^R - 1)^2 \quad (\text{A.3})$$

Regarding  $R$  and  $H$  as functions of the ratio

$$\theta = q/p \quad (\text{A.4})$$

we have for small  $\theta$

$$R(\theta) = (1/\theta)[1 + O(e^{-1/\theta})], \quad H(\theta) = \frac{1}{2}\{1 + O[(1/\theta)e^{-1/\theta}]\} \quad (\text{A.5})$$

while for  $\theta$  near 1

$$R(\theta) = 2(1 - \theta) + O[(1 - \theta)^2], \quad H(\theta) = \frac{1}{4} + O(1 - \theta) \quad (\text{A.6})$$

Both  $R$  and  $H$  are monotonically decreasing functions of  $\theta$  in the relevant interval  $0 \leq \theta \leq 1$ .

Upon using Stirling's approximation for the factorials it contains, the leading term in (A.1) may be put into the following form:

$$S_p^{(q)} \sim [L(p, \theta)]^p / [2\theta(\pi p R H)^{1/2}] \quad (\text{A.7})$$

where

$$L(p, \theta) = p^{1-\theta} \theta^{-\theta} e^{\theta-1} R^{-1} (e^R - 1)^\theta \quad (\text{A.8})$$

For very large values of  $p$ , the behavior of expression (A.7) is controlled primarily by the  $L^p$  factor, with the remaining factor providing a minor modulation. In fact  $L$  possesses a single maximum as a function of  $\theta$  in  $0 < \theta < 1$ , and so raising  $L$  to the high power  $p$  causes the  $S_p^{(q)}$  to exhibit a single, narrow maximum vs.  $q$  at fixed  $p$ .

We can locate the  $L$  maximum by taking logarithms in Eq. (A.8) and setting the first  $\theta$  derivative to zero:

$$0 = \ln p + \ln \theta - \ln(e^R - 1) + \left( \frac{1}{R} - \frac{\theta}{1 - e^{-R}} \right) R'(\theta) \quad (\text{A.9})$$

The large parameter in this equation is  $\ln p$ , and it follows from the properties given for  $R(\theta)$  that the root  $\theta_0$  to (A.9) must perforce be close to zero. If that is the case, the first of Eqs. (A.5) can be used to eliminate  $R(\theta)$  from Eq. (A.9). We finally conclude that the  $L$  maximum for large  $p$  must asymptotically be located at

$$\theta_0 \approx 1/\ln p \tag{A.10}$$

For  $q$  values reasonably close to  $\theta_0 p$ , the region of the  $S_p^{(q)}$  maximum, it will suffice for present purposes to introduce a Gaussian approximation to  $L^p$ , i.e., to  $L$  itself. This is accomplished by calculating first the second  $\theta$  derivative of  $\ln L$  at  $\theta = \theta_0$  to establish the proper Gaussian width. Substitution in Eq. (A.7) then yields the following result:

$$S_p^{(q)} \sim \frac{p! \exp(p/\ln p)}{2\pi p (\ln p)^{p - (1/2) - (p/\ln p)}} \exp[-\frac{1}{2} p \ln^2 p (\theta - \theta_0)^2] \tag{A.11}$$

As  $p$  increases, this Gaussian distribution of values for the  $S_p^{(q)}$  widens with respect to  $q$ .

Now we can estimate the polynomials  $S_p$ :

$$S_p(e^{-u}) = \sum_{q=1}^p S_p^{(q)} e^{-uq} \approx p \int_0^1 d\theta S_p^{(\theta p)} e^{-u\theta p} \tag{A.12}$$

The last expression is intended to be used with (A.11). In fact it is appropriate to extend the range of  $\theta$  integration to infinity in both directions. The integration then becomes elementary, yielding

$$S_p[\exp(-u)] \sim \frac{p! \exp[(1-u)p/\ln p + u^2 p/(2 \ln^2 p)]}{(2\pi p)^{1/2} (\ln p)^{p + (1/2) - (p/\ln p)}} \tag{A.13}$$

Finally we are in a position to estimate the ratios  $R_m$  appearing in Eq. (4.7) when  $m$  is large. Using Eq. (A.13), we find

$$R_m \sim \frac{4m}{(\ln m)^2} \left[ 1 + O\left(\frac{1}{\ln m}\right) \right] \tag{A.14}$$

The dependence of these ratios on  $u$  appears only in those higher order correction terms not explicitly shown.

### ACKNOWLEDGMENT

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