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Analysis of spectral densities using modified moments*

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Modified moments provide coefficients in orthogonal polynomial expansions for spectral densities. Using these expansions as a starting point, we develop a number of methods which can be used in the analysis of density functions. Expansions for averages over densities are described. These expansions, when combined with nonlinear convergence acceleration methods based on the Padé approximant, give apparently exponentially convergent results. By exploiting connections between the orthogonal polynomial expansion, Fourier series, and power series, we show how to obtain an accurate picture of the density itself. A rational approximation is described which gives very accurate results near the ends of the interval. Procedures are given for determining the number, types, and locations of singularities in a density from its modified moments. By an analysis of the asymptotic form of the modified moments, we show how this information about singularities can be incorporated into an expansion for the density using the "moment-singularity" method. We illustrate these methods by applications to harmonic solid models. We obtain extremely accurate results for averages and we obtain accurate representations for spectral densities which faithfully reproduce the Van Hove singularities.

I. INTRODUCTION

Many problems in theoretical chemistry and physics can be formulated in terms of density functions. Examples include the harmonic solid (normal-mode spectral density), band theory (density of states), and the Ising model (density of zeroes of the grand-partition function). It is often the case in such problems that the density itself is unknown, but that several power moments of it can be obtained. Modified moments, a generalization of power moments, can provide a powerful tool for the analysis of these cases.

In previous papers we have used modified moments in a number of ways in the analysis of the spectral density of harmonic solids. We have shown, by modification of a method of Isenberg,[1] how modified moments can be computed from the dynamical matrix.[2] We have used modified moments to obtain Gaussian quadrature formulas which provide rigorous bounds to averages over the density and have shown that they are superior to power moments for this purpose.[3,4] We have also shown that appropriately chosen modified moments are coefficients in an orthogonal polynomial expansion of the density which, in a transformed variable, becomes a Fourier expansion.[5]

In this paper we describe a number of methods for the analysis of density functions which take the orthogonal polynomial expansion as a starting point. These methods contain, either implicitly or explicitly, assumptions about the properties of the density in addition to the modified moments. When the density conforms to these assumptions, these methods can provide results which are strikingly better than those that can be obtained rigorously from the moments alone.

Using model harmonic solids as examples, we show how to obtain very accurate results for certain averages over density functions and how to obtain an accurate picture of the density itself. Previous methods for obtaining representations of density functions from moments have suffered from the inability to accurately reproduce singularities in the density without recourse to very detailed information about the singularities obtained by other means. Included in the methods we describe is a procedure by which, using only modified moments and the (implicit) assumption of a limited number of singularities in the density, a density function with singularities can be accurately represented.

In Sec. II we introduce notational conventions and definitions which will be used and review the Chebyshev orthogonal polynomial expansion and its connection with the Fourier expansion. In Sec. III we examine the Chebyshev polynomial expansion for a model harmonic solid and compare the results with those from two other methods, the Legendre polynomial expansion[6] and the continued-fraction method.[7]

In some problems the result of interest is not the density itself, but rather some average over the density. In Sec. IV we show how convergent expansions for some of these averages can be obtained by a formal integration of the orthogonal polynomial expansion. The convergence of these expansions, when applied to the spectral density of a model solid, is shown to be greatly accelerated by a nonlinear method based on the Padé approximant. This method is particularly useful for the determination of "singular" averages, such as the zero-point energy of the harmonic solid, for which Gaussian quadrature formulas do not provide satisfactory bounds.

In Sec. V we give a method for obtaining coefficients in a power-series expansion of the density function near the origin. In this case formally divergent expansions for the coefficients are obtained by a rearrangement of the orthogonal polynomial expansion. However, when the nonlinear method introduced in Sec. IV is applied to the expansions for coefficients of density functions known to possess a power-series expansion near the origin, good estimates for the first few coefficients are obtained.

A nonlinear method may also be used to improve the convergence of the expansion for the density itself. In Sec. VI we use the Laurent-Padé approximant[8,9] to obtain rational approximations for the spectral densities of two model harmonic solids. This method gives improved convergence near Van Hove singularities and gives a very marked improvement in the regions near the ends of the interval.

In Sec. VII we consider the asymptotic behavior of the modified moments. This behavior is determined by singularities in the density and leads very naturally to the introduction of a modification of the "moment-singularity method."[10] We show how this modification can provide a very accurate representation of a density with singularities when information about the singularities is known.

The modification of the moment-singularity method introduced here has the additional advantage that it suggests methods for obtaining information about singularities in a density directly from the modified moments. In Sec. VIII some procedures are described which may be used for this purpose. We show how a new method of series analysis proposed by Joyce and Guttman[11] can be used to determine the number and locations of singularities and (less precisely) their types. Linear and nonlinear least-squares methods are used to estimate the coefficients of the singularities and, in some cases, to obtain improved estimates for their locations and types. We apply these procedures to the spectral densities of two model solids which contain a variety of singularities (including a discontinuity) and obtain very good results.

II. MODIFIED MOMENTS

In this section we give definitions and review some basic results which will be used in the rest of the paper.

Let $G(x)$ be a non-negative density function defined on the interval $[0, 1]$. The power moments, μ_k, of $G(x)$ are defined by

$$\mu_k \equiv \int_0^1 x^k G(x)\, dx \equiv \langle x^k \rangle . \tag{2.1}$$

Consider the kth-degree monic polynomials $p_k(x)$ orthogonal with respect to some weight function $H(x)$:

$$\int_0^1 p_k(x) p_l(x) H(x)\, dx = \delta_{kl} N_k , \tag{2.2}$$

where N_k is a normalization constant. From the theory of orthogonal polynomials we know that the p_k satisfy a three-term recursion relation

$$p_{k+1}(x) = (x - a_k) p_k(x) - b_k p_{k-1}(x)$$
$$(p_0 \equiv 1,\ p_{-1} \equiv 0), \tag{2.3}$$

where a_k and b_k are constants. Further,

$$N_k = b_0 b_1 \cdots b_k \quad \left[b_0 = \int_0^1 H(x)\, dx \right] . \tag{2.4}$$

We define modified moments, ν_k, in terms of the p_k by

$$\nu_k \equiv \langle p_k(x) \rangle . \tag{2.5}$$

It is also useful to define normalized modified moments,

$$\nu_k^* \equiv \langle p_k^*(x) \rangle , \tag{2.6}$$

where the p_k^* are orthonormal polynomials with respect to $H(x)$.

The density function can formally be represented by an expansion in these orthonormal polynomials with coefficients which are just the normalized modified moments

$$G(x) \sim H(x) \sum_{k=0}^{\infty} \nu_k^* p_k^*(x) . \tag{2.7}$$

The convergence of such an expansion depends on the properties of $G(x)$ and the choice of $H(x)$. It is desirable that $H(x)$ be nonzero on the same interval as $G(x)$ and be as similar to $G(x)$ as possible, particularly at the ends of the interval. It may also be convenient to choose $H(x)$ to be a weight function of the classical (Chebyshev, Legendre, etc.) orthogonal polynomials since the properties of these polynomials are well known.

Many of the results in this paper will be derived using shifted Chebyshev polynomials of the second kind. For these polynomials

$$H(x) = (8/\pi)[x(1-x)]^{1/2} , \tag{2.8}$$

and the recursion relation is given by

$$p_{k+1}(x)=(x-\tfrac{1}{2})p_k(x)-\tfrac{1}{16}p_{k-1}(x)$$
$$(p_0\equiv 1,\ p_{-1}\equiv 0) \tag{2.9}$$

The normalized polynomials

$$p_k^*(x)=(-4)^k p_k(x) \tag{2.10}$$

can be reexpressed in the form

$$p_k^*(x)=\frac{\sin(k+1)\theta}{\sin\theta}\quad (x=\sin^2\tfrac{1}{2}\theta). \tag{2.11}$$

Under this transformation the orthogonal polynomial expansion (2.7) becomes

$$G(\sin^2\tfrac{1}{2}\theta)\sim\frac{4}{\pi}\sum_{k=0}^{\infty}\nu_k^*\sin(k+1)\theta\quad(0\leqslant\theta\leqslant\pi), \tag{2.12}$$

placing at our disposal, in addition to the theory of orthogonal polynomials, the methods of Fourier analysis. The series (2.12) can be shown to converge to the density for a very broad class of functions[12,13] and convergence can be established for all of the densities which we will consider. More importantly, the asymptotic behavior as $k\to\infty$ of the coefficients ν_k^* in this series (and therefore its *rate* of convergence) can be determined,[12] a point to which we shall return in Sec. VII.

III. EXPANSIONS FOR DENSITIES

In physical problems we may know or believe that the density function of interest is, in some sense, "well behaved." In such cases we may wish to represent the density by some smooth continuous function. A natural choice for such a representation is a partial sum to the orthogonal polynomial expansion Eq. (2.7). Another moment method, recently proposed by Gordon, which has been used for this purpose is the extrapolation of continued fraction expansions.[7,14] We examine the behavior of such representations for the spectral density of a three-dimensional harmonic solid below.

The spectral density of a harmonic solid, $\rho(\omega)$, is defined on the interval $[0, \omega_{max}]$. Since we usually know only the even moments of $\rho(\omega)$, it is convenient to define a squared-frequency spectral density, $G(x)$, with

$$x=\omega^2/\omega_{max}^2,\quad G(x)\,dx=\rho(\omega)\,d\omega. \tag{3.1}$$

As an example, we consider a cubic-close-packed (ccp) model with nearest-neighbor central forces for which large number of modified moments have been computed.[2] In Fig. 1 we show the 20th and 40th partial sums to a Chebyshev polynomial expansion for the squared-frequency density of this model.[15] In Fig. 2 we show the continued fraction extrapolations for the density obtained from 20 and 40 modified moments. A modification of Gordon's technique which was used to obtain Fig. 2 is described in Appendix A.

The two methods give essentially equivalent representations. The qualitative shape of the density is correctly represented but, as would be expected, the Van Hove singularities are only faintly suggested, even with 40 moments. An advantage of the Chebyshev expansion is, as we shall see, the ease with which information about these Van Hove singularities can be incorporated.

A more commonly used orthogonal polynomial expansion for the squared-frequency density is the expansion in Legendre polynomials.[6] This expan-

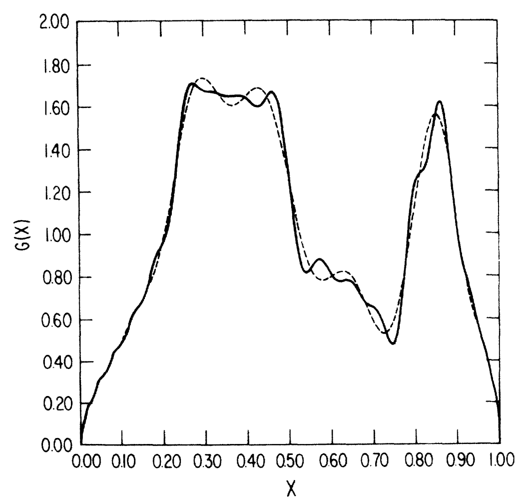

FIG. 1. Partial sums to a Chebyshev polynomial expansion for the spectral density of the ccp model. Solid line is 40th partial sum; dashed line is the 20th partial sum.

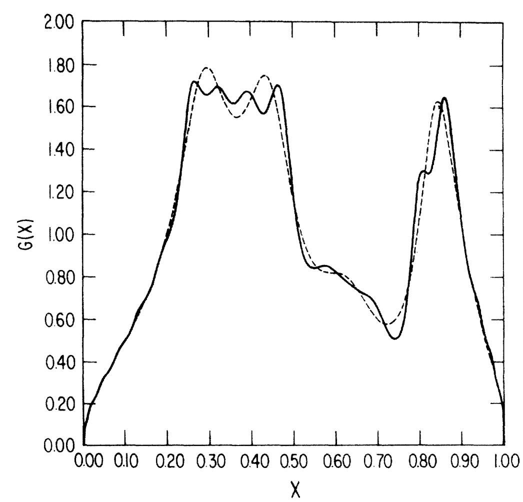

FIG. 2. Continued fraction extrapolations for the spectral density of the ccp model. Solid line is extrapolation from 40 modified moments; dashed line is extrapolation from 20 modified moments.

sion also gives the same qualitative representation of the density but does not do as well near the ends of the interval. This is because the density for a three-dimensional harmonic solid tends to zero as a square root at the ends of the interval, a feature which is built into the Chebyshev expansion and the continued fraction extrapolation employed here. The partial sums to the Legendre expansion usually tend to nonzero constants at the ends of the interval. Moreover, these constants are often negative for the model considered here. The Legendre expansion is more appropriate for densities which are nonzero at the ends of the interval (e.g., the squared-frequency density of a two-dimensional harmonic solid).

It should be noted that the problem of obtaining any of these representations from *power* moments is very ill conditioned. For example, a 40-moment continued fraction expansion cannot be obtained directly from power moments using 25 significant-figure arithmetic since no significant figures remain after the 35th moment is used. If a set of modified moments is known (either from an exact transformation of *exact* power moments[3] or by direct computation[2]), then the problem of determining coefficients in other orthogonal polynomial expansions or obtaining continued fraction expansions is usually very well conditioned. Methods for determining other expansion coefficients from a set of modified moments are described in Ref. 3. Some remarks on obtaining continued fraction expansions from modified moments are contained in Appendix A.

IV. AVERAGES

In many cases the result of interest is not the density itself but some average over the density,

$$\langle F(\{\tau\}, x)\rangle = \int_0^1 F(\{\tau\}, x)G(x)\,dx, \tag{4.1}$$

where $\{\tau\}$ is some set of appropriate independent parameters (time, temperature, etc.). For example, thermodynamic properties of the harmonic solid are obtained when the $F(\{\tau\}, x)$ are the Einstein functions for the harmonic oscillator.

Gaussian quadratures can provide very accurate bounds for many averages of the type shown in Eq. (4.1). However, if the function $F(\{\tau\}, x)$ is singular at one end of the interval, then the bounds obtained from moments alone may converge slowly or there may be no bound on $\langle F(\{\tau\}, x)\rangle$ in one direction. Methods, based on coefficients of a power-series expansion for $G(x)$ [or $G(x)$ times some known function] near $x=0$, have been given for obtaining improved results for such averages from quadratures.[3,16] However, unless the expansion coefficients are already known, their computation presents an added difficulty which limits the utility of these methods.

Another approach to the evaluation of averages is based on series expansions. Expansions in terms of power moments or "shifted" power moments, $v_k=\langle(1-x)^k\rangle$, can be given for thermodynamic properties of harmonic solids[17] and for time-autocorrelation functions.[18] However, these expansions are often characterized by rather slow convergence. Significantly more rapidly convergent expansions for some averages can be obtained in terms of modified moments by means of a term-by-term integration over the orthogonal polynomial expansion Eq. (2.7):

$$\langle F(\{\tau\}, x)\rangle = \sum_{k=0}^{\infty} \nu_k^* F_k(\{\tau\}),$$

$$F_k(\{\tau\}) = \int_0^1 F(\{\tau\}, x)p_k^*(x)H(x)\,dx. \tag{4.2}$$

When expansions in shifted Chebyshev polynomials of the second kind are used, the coefficients $F_k(\{\tau\})$ become Fourier sine components of $F(\{\tau\}, \sin^2\frac{1}{2}\theta)$:

$$F_k(\{\tau\}) = (4/\pi)\int_0^{\pi} F(\{\tau\}, \sin^2\tfrac{1}{2}\theta)\sin(k+1)\theta\,d\theta$$

$$[\theta = \sin^{-1}(x^{1/2})]. \tag{4.3}$$

In Table I we give expansions derived from the Chebyshev expansion for the "singular" averages

$$\mu_{-1}=\langle x^{-1}\rangle,\ \mu_{-1/2}=\langle x^{-1/2}\rangle,$$

$$\mu_{1/2}=\langle x^{1/2}\rangle,\ \lambda=\langle \ln(x^{1/2})\rangle. \tag{4.4}$$

For harmonic solids, the moment $\mu_{1/2}$ is just twice the zero-point energy in appropriate units, while the average λ contributes to the free energy and entropy at finite temperatures. The moments μ_{-1} and $\mu_{-1/2}$ are required for the normalization of time-autocorrelation functions in harmonic

TABLE I. Expansions for "singular" averages.

$F(x)$	$\langle F(x)\rangle$
x^{-1}	$\mu_{-1}=4\sum_{k=0}^{\infty}\nu_k^*$
$x^{-1/2}$	$\mu_{-1/2}=\frac{4}{\pi}\sum_{k=0}^{\infty}\nu_k^*\left(\frac{k+1}{(k+\frac{1}{2})(k+\frac{3}{2})}\right)$
$x^{1/2}$	$\mu_{1/2}=-\frac{2}{\pi}\sum_{k=0}^{\infty}\nu_k^*\left(\frac{k+1}{(k-\frac{1}{2})(k+\frac{1}{2})(k+\frac{3}{2})(k+\frac{5}{2})}\right)$
$\ln(x^{1/2})$	$\lambda=\nu_0^*(\frac{1}{4}-\ln 2)-\sum_{k=1}^{\infty}\nu_k^*\left(\frac{1}{k(k+1)}\right)$
x^s	$\mu_s=\frac{\Gamma(2s+3)}{2^{2s}(s+1)}\sum_{k=0}^{\infty}\nu_k^*\left(\frac{k+1}{\Gamma(s+k+3)\,\Gamma(s-k+1)}\right)$

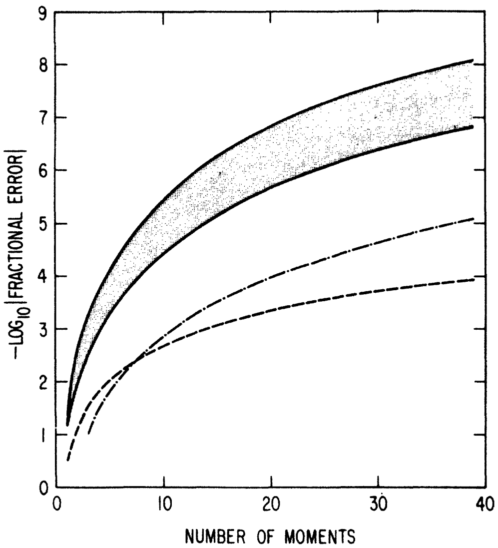

FIG. 3. Accuracy of methods for computing averages over the density. The negative of $\log_{10}(|\text{fractional error}|)$ for $\mu_{1/2}$ in the ccp model is plotted against the number of moments used. Envelope bounded by solid lines encloses results from modified moment expansion; dashed line shows results from shifted moment expansion; dotted line shows differences between bounds obtained from Gaussian quadratures.

solids; μ_{-1} has also been used for the computation of other averages.[19] Also included in Table I is an expansion for the general power moment, $\mu_s = \langle x^s \rangle$, from which the averages in (4.4) can be obtained (μ_{-1} and λ are limiting cases).

In Fig. 3 we examine results for $\mu_{1/2}$ of the nearest-neighbor central-forces ccp solid. We compare the relative accuracy obtained from the expansion in Table I with the accuracy obtained from Gaussian quadratures and from a shifted moment expansion,[20]

$$\mu_{1/2} = \sum_{k=0}^{\infty} \frac{\Gamma(k-\frac{1}{2})}{\Gamma(-\frac{1}{2})k!} v_k . \tag{4.5}$$

The convergence of the expansion in Table I is oscillatory and its accuracy is represented by an envelope. The upper and lower edges of the envelope indicate the best and poorest results, respectively. We see from the figure that the expansion in Table I is significantly more accurate than the other methods for any number of moments.

The results obtained from the expansions in Table I can be considerably improved by the use of nonlinear convergence acceleration methods. If we formally regard the terms in an expansion as coefficients of a power series in z, we can form Padé approximants[21] to this series and then evaluate them at $z=1$. Results for the ccp model when this nonlinear method is applied to the expansions in Table I with 10, 20, 30, and 40 modified moments are given in Table II. Also given in Table II are results for $\mu_{1/2}$ obtained from 10 and 18 modified moments for a nearest-neighbor central-forces hexagonal-close-packed model. In Fig. 4 we compare the relative accuracy of results for the ccp $\mu_{1/2}$ when the nonlinear method is applied to the modified moment expansion in Table I and to the shifted moment expansion (4.5). As in Fig. 3, the accuracy of the results for the expansion in Table I is indicated by an envelope.

We see from Fig. 4 and Table II that the nonlinear method appears to give exponential convergence when applied to the modified moment expansion and that the convergence acceleration is much less dramatic when the method is applied to the shifted moment expansion. A possible explanation for this difference may be found in the behavior of the two expansions, when considered as power series in z, at the point $z=1$. An analysis of the asymptotic behavior of the shifted moments shows that $k^{3/2}v_k$ tends to a positive constant as $k \to \infty$. It follows that the expansion (4.5) must have a branch-point singularity at $z=1$. The asymptotic behavior of the modified moments (to be considered in Sec. VII) shows that the modified moment expansion is analytic at $z=1$. [This is a consequence of our choice of a weight function $H(x)$ which has the same behavior as $G(x)$ near the origin.] Experience with Padé approximants suggests that convergence will be good in regions

TABLE II. Results of nonlinear extrapolation of expansions for "singular" averages.

k \ $\langle F(x) \rangle$	μ_{-1} [a]	$\mu_{-1/2}$ [a]	λ [a]	$\mu_{1/2}$ [a]	$\mu_{1/2}$ [b]
10	$3.3\underline{61}$	$1.63\underline{88}$	$-0.428\,\underline{98}$	$0.681\,\underline{88}$	$0.681\,\underline{79}$
20	$3.358\,\underline{73}$	$1.635\,9\underline{11}$	$-0.428\,\underline{870}$	$0.681\,774\,\underline{39}$	$0.681\,840\,\underline{50}$ [c]
30	$3.358\,826\,\underline{55}$	$1.635\,907\,\underline{91}$	$-0.428\,869\,56\underline{7\,3}$	$0.681\,774\,440\,58\underline{0\,8}$	
40	$3.358\,826\,15\underline{4\,0}$	$1.635\,907\,890\,\underline{61}$	$-0.428\,869\,566\,19\underline{3\,3}$	$0.681\,774\,440\,583\,\underline{65}$	

[a] Averages for nearest-neighbor ccp model. Underlined digits do not agree with correct answer.
[b] Averages for nearest-neighbor hcp model. Underlined digits do not agree with results for $k-1$ and $k-2$ moments.
[c] From 18 modified moments.

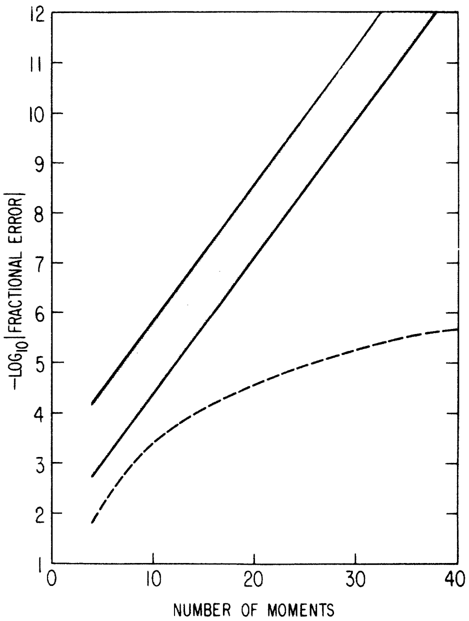

FIG. 4. Accuracy of nonlinear extrapolation of expansions for averages over the density. The negative of $\log_{10}(|\text{fractional error}|)$ for $\mu_{1/2}$ in the ccp model is plotted against the number of moments used. Envelope bounded by solid lines encloses results from modified moment expansion; dashed line shows results from shifted moment expansion.

where an expansion is analytic and will be poorer in regions where there is a branch-point singularity.[22]

Another difference between the two expansions which is of practical interest is a difference in their *apparent* rates of convergence. If we attempt to estimate the degree of convergence of partial sums by examining the last three partial sums in a sequence, then such a procedure will always lead to a conservative estimate for the convergence of the modified moment expansion but will cause an overestimation by 1 to 2 orders of magnitude for the shifted moment expansion. That is, the last partial sum to the shifted moment expansion will appear to be more accurate than it is. This difference is also observed in the apparent rate of convergence of successive Padé approximants.

Other averages for which the integration (4.3) can be performed to provide rapidly convergent series are time-autocorrelation functions (TAFs). In harmonic solids, quadrature methods provide good bounds for TAFs when the reduced time, τ, is less than twice n, the number of moments available.[4,23] However, series expansions can extend the time out to which these TAFs can be determined to $\tau\sim 3n$ and provide results which are significantly more accurate than the quadratures.[5] An expansion for the classical momentum TAF for a harmonic solid, $C_p^{\text{cl}}(t)$, has been given elsewhere.[5] In Table III we reproduce the result for $C_p^{\text{cl}}(t)$ and give two additional expansions for harmonic-solid TAFs: the quantum-mechanical momentum TAF, $C_p^{\text{qm}}(t)$, and the classical-displacement TAF, $C_q^{\text{cl}}(t)$. The TAFs which are double sums appear formidable, but the inner sums over Bessel functions can be performed quickly and accurately because of the rapid decay of $J_n(\tau)$ with increasing n. The second expression for $C_p^{\text{qm}}(t)$ is of interest because it leads to exponentially convergent results at short times if the inner sums over moments are extrapolated by the nonlinear method above. The bounds from quadratures converge more slowly.

The classical-momentum TAF is of special interest since this integral is found in a number of other problems. The method of obtaining this function by integration of orthogonal polynomial expansions of densities is not restricted to Chebyshev polynomial expansions and can give rapidly

TABLE III. Expansions for time-autocorrelation functions in terms of Bessel functions.

$F(\tau,x)$ [a]	$\langle F(\tau,x)\rangle$
$\cos(x^{1/2}\tau)$	$C_p^{\text{cl}}(t)=\sum_{k=0}^{\infty}\nu_k^*[J_{2k}(\tau)-J_{2k+4}(\tau)]$
$\dfrac{x^{1/2}\cos(x^{1/2}\tau)}{\mu_{1/2}}$	$C_p^{\text{qm}}(t)=J_0(\tau)-\dfrac{2}{\pi\mu_{1/2}}\sum_{k=0}^{\infty}\nu_k^*\left(\sum_{l=1}^{\infty}(\alpha_{k+l}+\alpha_{k-l})J_{2l}(\tau)\right)$
	$=J_0(\tau)-\dfrac{2}{\pi\mu_{1/2}}\sum_{l=0}^{\infty}J_{2l}(\tau)\left(\sum_{k=0}^{\infty}\nu_k^*(\alpha_{k+l}+\alpha_{k-l})\right)$
	where $\alpha_m=(m+1)/[(m-\frac{1}{2})(m+\frac{1}{2})(m+\frac{3}{2})(m+\frac{5}{2})]$
$\dfrac{x^{-1}\cos(x^{1/2}\tau)}{\mu_{-1}}$	$C_q^{\text{cl}}(t)=1-\dfrac{4}{\mu_{-1}}\sum_{k=0}^{\infty}\nu_k^*\left[\sum_{l=0}^{\infty}(l+1)\left(J_{2(l+k+1)}(\tau)-J_{2(l+k+1)+4}(\tau)\right)\right]$

[a] $\tau=\omega_{\max}t$ where t is time.

convergent results even for very irregular densities. A very general convergence proof for this method is given in Appendix B.

Partial averages over the density can also be obtained by integration of the Chebyshev expansion. Two examples which have recently been used in the theory of metals[24] are the cumulative density function

$$\int_0^x G(x')\,dx' = \frac{1}{\pi}\sum_{k=0}^{\infty} \nu_k^* \left(\frac{\sin k\theta}{k} - \frac{\sin(k+2)\theta}{k+2}\right), \tag{4.6}$$

and the integral

$$\int_0^x (x'-\tfrac{1}{2})G(x')\,dx' = \frac{1}{4\pi}\sum_{k=0}^{\infty} \nu_k^* \left(\frac{\sin(k+3)\theta}{(k+3)} - \frac{\sin(k-1)\theta}{(k-1)}\right), \tag{4.7}$$

where $x=\sin^2\frac{1}{2}\theta$ and where $(\sin j\theta)/j$ is defined to be equal to θ for $j=0$. These expansions are also rapidly convergent. For example, a comparison of the partial sums obtained with 40 and 60 moments suggests that the cumulative density from 40 moments is in error by less than 10^{-3} over the entire interval.

V. POWER-SERIES EXPANSIONS

We remarked in Sec. IV that good results for singular averages over $G(x)$ can be obtained from Gaussian quadratures by using coefficients of a power-series expansion for a known function times $G(x)$ near $x=0$. The fact that good results for singular averages are obtained from modified moments without the use of such coefficients suggests that it may be possible to obtain the series coefficients directly from the modified moments.

If $H(x)$ can be chosen so that the ratio $G(x)/H(x)$ possesses a convergent power-series expansion near the origin

$$G(x)/H(x) \sim \sum_{k=0}^{\infty} d_k x^k , \tag{5.1}$$

then a rearrangement of the orthogonal polynomial expansion (2.7) will lead to a formal expansion for the coefficients d_k in terms of modified moments. In the case of shifted Chebyshev polynomials of the second kind [cf. Eqs. (2.8)–(2.12)], this expansion has the form

$$d_k \sim (-4)^k \sum_{l=k}^{\infty} \binom{l+1+k}{l-k} \nu_l^* . \tag{5.2}$$

When $G(x)/H(x)$ contains singularities inside the interval (0, 1) the formal series (5.2) for the coefficients will generally be divergent even though the series in (5.1) has a nonzero radius of convergence. (Depending upon the nature of the singularities, the first few d_k's may be convergent. See Sec. VII.) Nonetheless, good estimates for the first few of these coefficients may be obtained by formally treating the expansions as power series in z, forming Padé approximants, and evaluating at $z=1$ (i.e., the nonlinear method of Sec. IV).

For harmonic solids in three dimensions the squared-frequency density is usually expanded near the origin as

$$G(x) \sim \tfrac{1}{2} x^{1/2} \sum_{k=0}^{\infty} c_k x^k . \tag{5.4}$$

The c_k are easily obtained as linear combinations of the d_k when $H(x)=(8/\pi)[x(1-x)]^{1/2}$.

In Table IV we examine the accuracy of results obtained for the first three coefficients c_k in the nearest-neighbor ccp model using the nonlinear method. The most accurate values for these coefficients have been obtained by Isenberg.[25] We see from the table that, although increasingly large numbers of moments are required to obtain accurate results for the higher coefficients, the nonlinear method appears to give exponential convergence to Isenberg's results.

An expansion of the form (5.1) in powers of $(1-x)$ which is convergent near $x=1$ can also be given. The expansion for the d_k in this case is obtained by replacing ν_l^* by $(-1)^l\,\nu_l^*$ in Eq. (5.2).

Methods for extracting power-series coefficients from power moments and shifted moments have been given,[17] but the convergence of such methods is extremely slow. Direct application of nonlinear extrapolation to these methods does not dramatically improve their convergence. It is possible, by a combination of linear and nonlinear extrapolation techniques, to obtain good estimates for the first few coefficients from shifted moments,[26] but this rather complicated procedure does not improve upon the results in Table IV.

TABLE IV. Accuracy of nonlinear extrapolation method for power-series expansion coefficients to $G(x)$. The negative of $\log_{10}$(|fractional error|) in the coefficients of expansion (5.4) for the ccp model is tabulated.

k \ c_k	c_0	c_1	c_2
10	1.5	...	...
20	3.4	1.0	...
30	6.4	4.4	1.5
40	9.3	6.5	3.8

VI. RATIONAL APPROXIMATIONS FOR DENSITIES

The success of nonlinear methods based on rational approximations in computing averages and power-series coefficients leads us to seek a rational approximation to the density itself. For this purpose it is useful to define

$$f(z)=\frac{4}{\pi}\sum_{k=0}^{\infty}\nu_k^* z^k . \tag{6.1}$$

When the ν_k^* are the modified moments obtained from shifted Chebyshev polynomials of the second kind, $f(z)$ is analytic inside the unit circle. It follows from the Fourier expansion (2.12) that

$$G(\sin^2\tfrac{1}{2}\theta)=\lim_{r\to 1^-}\operatorname{Im}[re^{i\theta}f(re^{i\theta})]. \tag{6.2}$$

Using N modified moments we form the $[n/m]$ Padé approximant to $f(z)$

$$f(z)\sim f_{n,m}(z)\equiv\frac{4}{\pi}\frac{\sum_{k=0}^{n}q_k z^k}{\sum_{k=0}^{m}r_k z^k}$$

$$(r_0\equiv 1;\ m+n+1=N;\ n\equiv m). \tag{6.3}$$

In principle we could now obtain an estimate for $G(\sin^2\frac{1}{2}\theta)$ following Eq. (6.2). In practice, an alternate (entirely equivalent) procedure[8, 9] is more convenient. We observe that

$$G(\sin^2\tfrac{1}{2}\theta)=\frac{e^{i\theta}f(e^{i\theta})-e^{-i\theta}f(e^{-i\theta})}{2i}. \tag{6.4}$$

Then, using the approximant in Eq. (6.3) for f, bringing the sums over a common denominator and substituting $x=\sin^2\frac{1}{2}\theta$, we obtain

$$G(x)\sim G_{n,m}(x)=\frac{8}{\pi}[x(1-x)]^{1/2}\frac{\sum_{k=0}^{n}s_k p_k^*(x)}{\sum_{k=0}^{m}t_k p_k^*(x)},$$

$$s_k=\sum_{l=k}^{n}(q_l r_{l-k}-q_{l-k}r_{l+2}), \tag{6.5}$$

$$t_k=\sum_{l=k}^{m}r_{l-k}(r_l-r_{l+2})\quad(r_j\equiv 0\text{ for }j>m),$$

where the p_k^* are again the shifted Chebyshev polynomials of the second kind. This orthogonal polynomial analog of the Padé approximant has the property that its first N modified moments are identical to those for $G(x)$.[27]

In Fig. 5 we show the results from this method for the nearest-neighbor ccp harmonic solid obtained with 20 and 40 modified moments. We again, as with the methods of Sec. III, obtain a good picture of the qualitative shape of the density. With 40 moments the approximant also exhibits behavior which is suggestive of the Van Hove singularities. The most striking feature of the method is the accuracy with which it represents the density near the origin. A comparison of these results with those obtained from the expansion (5.4) using the 30 coefficients of Ref. 25 indicates that this method approximates the density on the interval $[0, 0.1]$ with a maximum absolute error of a few parts in 10^4 with 20 moments, and only a few parts in 10^9 with 40 moments. The relative error on this interval is also small; it has a maximum value of about 3×10^{-4} with 20 moments and 6×10^{-9} with 40 moments. Comparison of successive approximants suggests that the convergence is exponentially rapid near $x=1$ as well.

A possible application for this method is in the evaluation of averages which depend most heavily on the behavior of the density near the origin. (An example of such an average is the low-temperature heat capacity for harmonic solids. There appears to be no convenient expansion of the type given in Sec. IV for this average.) The average can be broken in two parts,

$$\langle F(\{\tau\},x)\rangle=\int_0^{x_0}F(\{\tau\},x)G(x)\,dx+\int_{x_0}^{1}F(\{\tau\},x)G(x)\,dx, \tag{6.6}$$

where x_0 is some point, say 0.1, near the origin. The rational approximation can be used in place of $G(x)$ in the first integral and the second integral can be evaluated using Gaussian quadratures. Experience with a similar method[3, 16] suggests that the result should be significantly better than that obtained by using Gaussian quadratures alone.

Following a procedure similar to the one we have described, one can also obtain an analog to

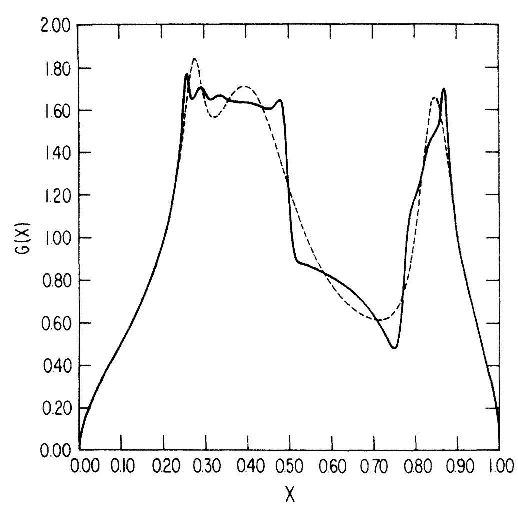

FIG. 5. Rational approximations for the spectral density of the ccp model. Results are from the orthogonal polynomial analog of the Padé approximant using shifted Chebyshev polynomials of the second kind. Solid line is approximation using 40 modified moments; dashed line is approximation using 20 modified moments.

the Padé approximant for shifted Chebyshev polynomials of the *first* kind. These polynomials are orthogonal with respect to the weight function

$$H(x) = (1/\pi)[x(1-x)]^{-1/2}\,, \qquad (6.7)$$

and, under the transformation $x = \sin^2\frac{1}{2}\theta$, they satisfy the identity

$$p_k^*(x) = \cos k\theta\,. \qquad (6.8)$$

The shifted Chebyshev polynomials of the first kind are particularly appropriate for approximating densities with square-root divergences at the ends of the interval such as the squared-frequency density of a one-dimensional harmonic chain.

We have applied this approximation technique to the spectrum of a diatomic chain ($ABAB$) with $m_A/m_B = 2$. The density $G(x)$ for this model is symmetric about $x=\frac{1}{2}$. It has square-root divergences at $x=\frac{1}{3}$ and $x=\frac{2}{3}$ as well as at the ends of the interval, and has a gap where the density is zero between $\frac{1}{3}$ and $\frac{2}{3}$. In Fig. 6 we show the exact spectral density for this model together with the 14-moment orthogonal polynomial expansion in Chebyshev polynomials of the first kind. To facilitate comparison with previous treatments[17] of this model, we have displayed $\omega_{max}\rho(\omega)$ rather than $G(x)$. In this form, the density does not diverge at the origin. The Chebyshev expansion provides results which compare favorably with the usual Legendre expansion.

In Fig. 7 we show the result for this model obtained using the rational approximation with 14

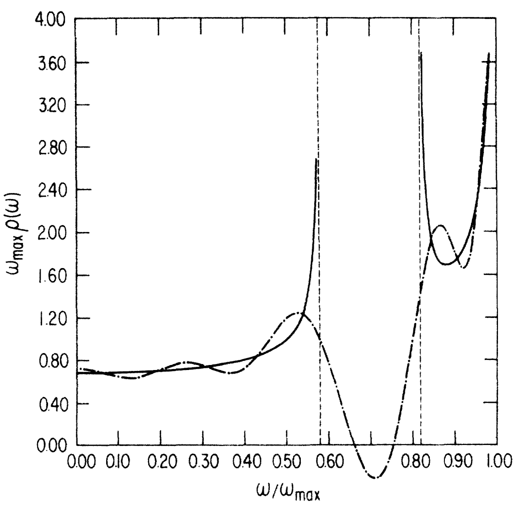

FIG. 6. Spectral density of the diatomic chain. Solid line is exact result; dotted line is result from expansion in shifted Chebyshev polynomials of the first kind using 14 modified moments. Dashed line indicates the edges of the gap in the exact result. Exact result diverges at the edges of the gap; both results diverge at 1.0.

moments. This approximation mimics the behavior of the exact result near the edges of the gap much more accurately than the polynomial expansion. It gives a good quantitative representation of the density over most of the region outside the gap: for ω/ω_{max} less than 0.55 or greater than 0.84 ($x <\sim 0.3$ or $>\sim 0.7$) it is correct to better than 1%. Moreover, it is quite accurate at the ends of the interval: for ω/ω_{max} less than 0.3 or greater than 0.95 ($x<\sim 0.1$ or $>\sim 0.9$) it is correct to one part in 10^4. When more moments are used, the accuracy at the ends of the interval increases (apparently exponentially), the representation remains accurate closer to the edges of the gap, and the approximant oscillates more closely about zero inside the gap. {B. G. Nickel [J. Phys. C 7, 1719 (1974)] has obtained a rational approximation to spectral densities essentially identical to Eq. (6.3) from a quite different point of view in connection with the randomly dilute ferromagnet.}

VII. ASYMPTOTIC FORM OF THE MODIFIED MOMENTS AND THE MOMENT-SINGULARITY METHOD

If the density function $G(\sin^2\frac{1}{2}\theta)$ is not an analytic function of θ everywhere on $[0, \pi]$, then the asymptotic behavior as $k\to\infty$ of the modified moments in the Fourier expansion Eq. (2.12) will be determined by the singularities in the density. When the nature of these singularities is known,

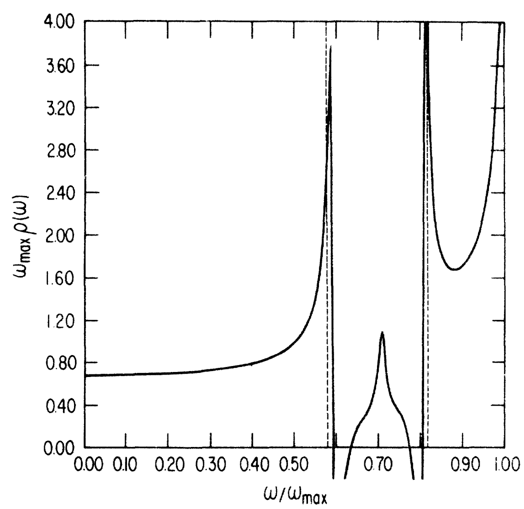

FIG. 7. Rational approximation for the spectral density of the diatomic chain. Result shown by solid line is from the orthogonal polynomial analog of the Padé approximant using shifted Chebyshev polynomials of the first kind and 14 modified moments. Dashed line indicates the edges of the gap in the exact result. Approximate result has minima off scale at ~0.596 and ~0.802 where it attains values of ~−1.79 and ~−2.40, respectively. Result diverges at 1.0 and has a maximum off scale at ~0.810 where it attains a value of ~5.25.

an expression for the asymptotic form of the ν_k^* can be obtained.[12] If the density has m singular points, $\theta_1, \ldots, \theta_m$, then each singularity will make a contribution, $(\nu_k^*)_i^s$, to the asymptotic behavior of ν_k^*:

$$\nu_k^* \sim \sum_{i=1}^{m} (\nu_k^*)_i^s \, . \tag{7.1}$$

In Table V we give expressions for the contribution of common Van Hove singularities to the asymptotic form of the modified moments.

The analysis of the asymptotic behavior of modified moments leads very naturally to an extension of the "moment-singularity method" first proposed by Lax and Lebowitz for harmonic solids.[10] We write, for a density with m singularities,

$$G(\sin^2\tfrac{1}{2}\theta) = G^r(\sin^2\tfrac{1}{2}\theta) + \sum_{i=1}^{m} G_i^s(\sin^2\tfrac{1}{2}\theta), \tag{7.2}$$

where

$$G_i^s(\sin^2\tfrac{1}{2}\theta) = \frac{4}{\pi}\sum_{k=0}^{\infty} (\nu_k^*)_i^s \sin[(k+1)\theta] \tag{7.3}$$

is the "dominant part" of the ith singularity and

$$G^r(\sin^2\tfrac{1}{2}\theta) = \frac{4}{\pi}\sum_{k=0}^{\infty}\left[\nu_k^* - \sum_{i=1}^{m} (\nu_k^*)_i^s\right] \sin[(k+1)\theta] \tag{7.4}$$

is the more "regular" remainder.

When N modified moments and the singularities are known, $G(\sin^2\frac{1}{2}\theta)$ can be approximated using the sums (7.3) and the Nth partial sum to Eq. (7.4). The sums (7.3) are generally rather slowly convergent at the singularities. In Appendix C we show how to evaluate these sums for each of the singularities in Table V, either in closed form or as extremely rapidly convergent series.

The moment-singularity method should not be understood to require the separation of $G(\sin^2\frac{1}{2}\theta)$ into an analytic part and a singular part. In general, $G^r(\sin^2\frac{1}{2}\theta)$ is *not* analytic. Rather, it contains singularities which are "weaker" than those in $G(\sin^2\frac{1}{2}\theta)$. That is, the modified moments of $G^r(\sin^2\frac{1}{2}\theta)$ tend to zero faster than those for $G(\sin^2\frac{1}{2}\theta)$ (e.g., as $n^{-5/2}$ instead of $n^{-3/2}$) and hence the sum in Eq. (7.4) converges faster than the sum in Eq. (2.12).

The method may be applied with minor modification when the nature of the singularities is known as a function of $x = \sin^2\frac{1}{2}\theta$ instead of as a function of θ. Expanding about a singular point θ_c we have

$$x - x_c = (\tfrac{1}{2}\sin\theta_c)(\theta-\theta_c) + (\tfrac{1}{4}\cos\theta_c)(\theta-\theta_c)^2 - \cdots \, . \tag{7.5}$$

Thus, except when θ_c is near 0 or π, the dominant part of the singularity as a function of x is the same as the dominant part as a function of θ and results of the kind shown in Table V can be used directly. When the singularity is near an end of the interval, it may be necessary to make corrections for the higher-order terms in Eq. (7.5) but this is usually straightforward.

As an example of the application of the moment-singularity method, we consider the simple-cubic (sc) harmonic solid with nearest-neighbor central and noncentral forces which has been extensively analyzed by Montroll.[28] The singularities in the squared-frequency density for this model can be discovered by an analysis of its dynamical matrix.[17] They are given in Table VI together with their contributions to the asymptotic form of the modified moments. In Fig. 8 we show the approximate density for this model obtained from 15 moments. This approximation has a maximum error

TABLE V. Dominant contribution to the modified moments from singularities in the density.

Type	$G(\sin^2\frac{1}{2}\theta)$	$(\nu_k^*)^s$
one dimensional		
minimum, maximum	$\lvert\theta-\theta_c\rvert^{-1/2}[1\pm\mathrm{sgn}(\theta-\theta_c)]$	$\pi^{1/2}(k+1)^{-1/2}\sin[(k+1)\theta_c \pm \frac{1}{4}\pi]$
two dimensional		
saddle	$\ln\lvert\theta-\theta_c\rvert$	$-\frac{1}{2}\pi(k+1)^{-1}\sin[(k+1)\theta_c]$
minimum, maximum (discontinuity)	$1\pm\mathrm{sgn}(\theta-\theta_c)$	$-(k+1)^{-1}\sin[(k+1)\theta_c \pm \frac{1}{2}\pi]$
three dimensional		
S_1, S_2 (saddle)	$-\lvert\theta-\theta_c\rvert^{1/2}[1\pm\mathrm{sgn}(\theta-\theta_c)]$	$\frac{1}{2}\pi^{1/2}(k+1)^{-3/2}\sin[(k+1)\theta_c \mp \frac{1}{4}\pi]$
minimum, maximum	$\lvert\theta-\theta_c\rvert^{1/2}[1\pm\mathrm{sgn}(\theta-\theta_c)]$	$\frac{1}{2}\pi^{1/2}(k+1)^{-3/2}\sin[(k+1)\theta_c \pm \frac{3}{4}\pi]$
generalized		
	$\lvert\theta-\theta_c\rvert$	$-(k+1)^{-2}\sin[(k+1)\theta_c]$
	$\lvert\theta-\theta_c\rvert\ln\lvert\theta-\theta_c\rvert[1\pm\mathrm{sgn}(\theta-\theta_c)]$	$(k+1)^{-2}\{(2/\pi)\ln(k+1)\sin[(k+1)\theta_c] \mp \cos[(k+1)\theta_c]\}$

of 6×10^{-3} which occurs at the singularities located at 0.2 and 0.8. On the scale shown, it is almost indistinguishable from the exact density. We emphasize that the approximate density in Fig. 8 has mathematically sharp singularities of precisely the type given in Table V. The slope of $G(x)$ in Fig. 8 has the correct square-root divergence at the singularities.

VIII. MODIFIED-MOMENT ANALYSIS OF SINGULARITIES

While the behavior of the modified moments is rigorously determined by singularities in the density only in the limit $k\to\infty$, the dominance of the singularities is in evidence for fairly small k. For example, with the sc solid considered in Sec. VII, the relative error, $[\nu_k^* - \sum_{i=1}^m (\nu_k^*)_i^s]/\nu_k^*$, made by approximating the moments with their asymptotic form is less than 10% for $10<k<20$ and less than 3% for $40<k<50$. Thus, it is not unreasonable to attempt to determine information about the singularities from the modified moments.

Singularities in the density $G(\sin^2\frac{1}{2}\theta)$ will appear in the function $f(z)$ [cf. Eqs. (6.1) and (6.2)] as singularities on the unit circle in the complex z plane. Because $G(\sin^2\frac{1}{2}\theta)$ is the imaginary part of $e^{i\theta}f(e^{i\theta})$, simple algebraic branch points in $f(z)$ may result in Van Hove-like singularities in $G(\sin^2\frac{1}{2}\theta)$ of the type given in Table V. For example, a singularity of the form

$$f(z)=h(z)+g(z)(z-z_c)^p\,, \qquad (8.1)$$

with h, g analytic at $z=z_c=e^{i\theta_c}$ and $p=\frac{1}{2}$, could reproduce at least the dominant part of the maximum, minimum, S_1, and S_2 singularities encountered in the squared-frequency density of a three-dimensional solid.

A. Series-analysis methods

A number of methods are available for estimating the location and nature of algebraic singularities in a function from a limited number of coefficients in a power-series expansion of the function about zero such as Eq. (6.1). Two of the most widely used methods are the ratio method[29] and Padé approximants.[30] They have been applied with considerable success in the study of critical phenomena and phase transitions.[22] Recently these techniques have also been applied, using a power moment expansion, to analyze singularities at the band edges of spectra in the Hubbard model.[31] However, these methods are not well suited for the analysis of singular functions like Eq. (8.1).

A new method of series analysis recently proposed by Joyce and Guttman[11] is well suited to the analysis of singularities like that in Eq. (8.1). This method approximates the function $f(z)$ by the solution of a second- (or higher-) order linear differential equation with polynomial coefficients which are determined by the known expansion coefficients for $f(z)$. The solution of such a differential equation can have the same form as Eq. (8.1) at a singular point with p an arbitrary noninteger constant. [When the exponent p is a nonnegative integer, an additional factor of $\ln(z-z_c)$ is present in the singular term.] The imaginary part of the solution can have the same form as any of the singularities in Table V. The one-two-, and three-dimensional singularities are obtained when $p=-\frac{1}{2}$, 0, $+\frac{1}{2}$, respectively; the first generalized singularity, when $p=1$.

We have used the method of Joyce and Guttman to analyze the singularities in the squared-frequency densities of two model solids, the sc model

TABLE VI. Singularities in the density for the sc model and their dominant contributions to the modified moments.

		$(\nu_k^*)_i^s = A_i\,(k+1)^{-3/2}\,\sin[(k+1)\theta_i+\eta_i]$		
i [a]	type	$x_i=\sin^2\frac{1}{2}\theta$	A_i	η_i
1	S_2	0.100	$10\sqrt{3}\,(2\pi)^{-3/2}=1.099\,74\cdots$	$\frac{1}{4}\pi$
2	S_1	0.200	$10(2\pi)^{-3/2}=0.634\,93\cdots$	$-\frac{1}{4}\pi$
3	S_2	0.800	$10(2\pi)^{-3/2}$	$\frac{1}{4}\pi$
4	S_1	0.900	$10\sqrt{3}\,(2\pi)^{-3/2}$	$-\frac{1}{4}\pi$

[a] sc model also has a minimum at $x=0$ and a maximum at $x=1$. However, these make no contribution to the asymptotic behavior of the modified moments.

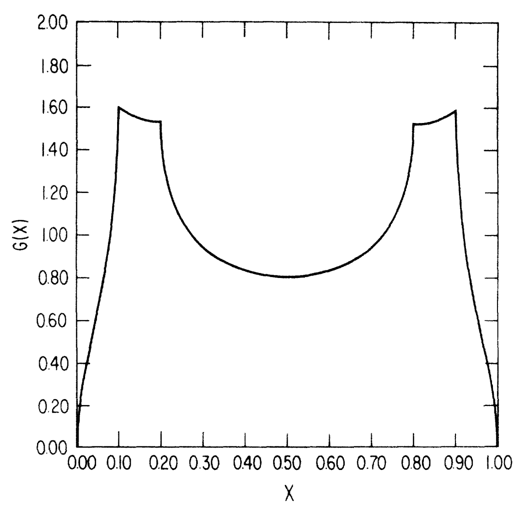

FIG. 8. Moment-singularity method for the spectral density of the sc model. Result is obtained using the functional forms for the asymptotic contributions of the singularities given in Table VI and 15 modified moments.

TABLE VII. Estimates for locations of singularities in the density for the sc model obtained using the method of Joyce and Guttman.

$[M_0, M_1, M_2]$	x_1	x_2	x_3	x_4
[9,8,8]	0.100 680	0.199 506	0.800 493	0.899 319
[11,10,10]	0.099 775	0.200 017	0.799 982	0.900 224
[13,12,12]	0.099 952	0.199 901	0.800 098	0.900 047
[15,14,14]	0.099 969	0.200 011	0.799 988	0.900 030
[17,16,16]	0.099 989	0.199 981	0.800 018	0.900 010
exact	0.100 000	0.200 000	0.800 000	0.900 000

of Sec. VII, and the nearest-neighbor central-forces ccp model. Results were obtained from the second-order equation which we write (following Ref. 11) as

$$Q_2(z)\Delta^2\psi_2(z) + Q_1(z)\Delta\psi_2(z) + Q_0(z)\psi_2(z) = 0 \quad \left(\Delta = z\frac{d}{dz}\right), \tag{8.2}$$

where the $Q_i(z)$ are polynomials and $\psi_2(z)$ is an approximant for $f(z)$. Singularities are located at the roots of $Q_2(z)$ and their exponents can be determined from ratios of the polynomials. We use the notation $[M_0, M_1, M_2]$ to refer to the approximant obtained when the degrees of Q_0, Q_1, and Q_2 are M_0, M_1, and M_2. Such an approximant is determined by $M_0 + M_1 + M_2 + 2$ expansion coefficients for $f(z)$.

The Joyce-Guttman method is very successful when applied to the analysis of singularities in the sc model. In Table VII we give results obtained for their locations from a sequence of $[2n+1, 2n, 2n]$ approximants with $n=4, \ldots, 8$. The results are given in terms of the variable $x = \sin^2\frac{1}{2}\theta$ and exact results are given for comparison. The higher approximants also contain nonphysical singularities. However, these nonphysical singularities lie well off the unit circle. Thus, an important advantage of the Joyce-Guttman method is that it provides a method for estimating *the number* of singularities in the density. Results for the exponents are not as dramatic as those for the locations. Values as good as 0.505 are obtained from the [17, 16, 16] approximant while the poorest value, 0.604, is obtained from the [9, 8, 8] approximant. Nonetheless, all of the values give a good indication of the correct square-root behavior.

Results are also good when the Joyce-Guttman method is applied to the ccp model. However, some difficulty is encountered with two of the singularities. In Table VIII we give results for the locations of the singularities obtained from a sequence of $[2n, 2n, 2n]$ approximants with $n = 5, \ldots, 10$. From an examination of symmetry points in the Brillouin zone, it can be determined that the density for the ccp model has singularities at 0.25, 0.5, 0.75, and 0.78125.[32] A fifth singularity can be inferred to lie near 0.9.[32] The locations of the singularities at 0.25 and 0.5 are accurately determined (Table VIII) by the approximants and the one near 0.9 is consistently determined to be 0.877⋯. The exponents for these singularities are known to be 0.5, 0, and 0.5, respectively.[32,33] The [20, 20, 20] approximant estimates them as 0.500 01, 0.0047, and 0.4992, while the [10, 10, 10] approximant gives 0.4990, −0.072, and 0.390. Reasonable estimates for the remaining two locations, 0.75 and 0.781 25, are obtained from the [14, 14, 14] and [16, 16, 16] approximants but estimates for their exponents are inconsistent. The [18, 18, 18] and [20, 20, 20] approximants place an additional singularity on the unit circle in the region of these two singularities.

Experience with the Joyce-Guttman method is still rather limited and some remarks on the practical aspects of its application seem warranted. In our experience, the locations of singularities are determined more reliably than the exponents. The best results are obtained at singularities which are well isolated such as those in the ccp model at 0.25 and 0.5. None of the sin-

TABLE VIII. Estimates for locations of singularities in the density for the ccp model obtained using the method of Joyce and Guttman.

$[M_0, M_1, M_2]$	x_1	x_2	x_3	x_4	x_5
[10,10,10]	0.250 001 54	0.501 601	0.656 195	0.761 414	0.876 828
[12,12,12]	0.249 996 77	0.500 282	0.720 972	0.775 276	0.878 908
[14,14,14]	0.250 000 63	0.500 123	0.753 871	0.794 273	0.877 792
[16,16,16]	0.250 000 18	0.500 101	0.747 118	0.786 456	0.877 723
[18,18,18]	0.250 000 01	0.500 043	0.754 858	0.779 750	0.877 680
			0.771 343		
[20,20,20]	0.249 999 99	0.500 024	0.751 612	0.777 254	0.877 650
			0.764 846		
exact	0.250 000 00	0.500 000	0.750 000	0.781 250	?

gularities in the approximants lies precisely on the unit circle and one indication of the reliability of a result is how close it is to the unit circle. Any result which lies more than 0.01 off the circle should be regarded with suspicion and very good results usually lie within 0.0001 of the circle. Within this range, the locations are usually quite reliable and the exponents are reasonable but may still contain errors on the order of 20%.

The difficulties with the singularities at 0.75 and 0.781 25 in the ccp model require special comment. In addition to the fact that these singularities lie close to one another, the situation is complicated by the fact that the singularity at 0.75 has a rather unusual character. A preliminary analysis[33] indicates that this singularity is a confluent singularity of a kind which cannot appear in the solution of a homogeneous second-order differential equation with polynomial coefficients. Thus, inconsistent results from the Joyce-Guttman method may serve to warn the user of (perhaps unsuspected) complications in his problem.

B. Least-squares methods

Before the locations and exponents of the singularities can be used in the moment-singularity method of Sec. VII, the coefficients of the singularities must be obtained. That is, for each singularity, a constant which is equivalent to the leading term in a power-series expansion for $g(z)$ in Eq. (8.1) must be determined. This constant appears in the asymptotic form of the modified moments as a magnitude and a phase shift (e.g., the constants A_i and the phase shifts η_i in Table VI).

In some problems, physical considerations may limit the choice of the phase shifts, η_i, to a few discrete values. (This happens, for example, in the harmonic-solid models considered here.) In this case the appropriate η_i can usually be determined by inspection of the orthogonal polynomial expansion or rational approximation. The problem is then reduced to estimating the magnitudes, A_i. One way to do this is to choose them to give the best least-squares fit to the known modified moments. That is, given $n+1$ modified moments, we determine m constants A_i which minimize

$$\sum_{k=0}^{n}\left(\nu_k^* - \sum_{i=1}^{m} A_i f_i(k)\right)^2 w(k)\,, \tag{8.3}$$

where the $f_i(k)$ are the functional forms of the asymptotic contributions of the m singularities as determined by their locations, exponents, and phase shifts and where $w(k)$ is some weight function which anticipates the decreasing residual as k increases.

Using this procedure with 40 modified moments, the exact f_i, and $w(k)=k^5$, the magnitudes A_i for the sc model are determined to within a few tenths of a percent. The density which is computed using these estimates is somewhat more accurate than that shown in Fig. 8 which was computed with exact magnitudes and 15 moments.

When no *a priori* information on the phase shifts is known, they too must be estimated from the modified moments. Linear least squares can be used to estimate both A_i and η_i by writing

$$A_i \sin[(k+1)\theta_i+\eta_i] = B_i \sin[(k+1)\theta_i] + C_i \cos[(k+1)\theta_i]\,, \tag{8.4}$$

with B_i and C_i to be determined. While we have not tested this procedure in detail, limited experience suggests that the η_i can be determined to within a few percent in this manner.

The combination of the Joyce-Guttman method, linear least-squares estimates for the coefficients, and the moment-singularity method of Sec. VII constitutes a prescription for determining the density from the modified moments. However, if the Joyce-Guttman method does not unambiguously determine the locations and exponents of all of the singularities, either because an insufficient number of moments is known or as a result of more fundamental difficulties, then other techniques are required.

In some cases, it may be possible to determine the locations of the singularities by other means. For example, although the Joyce-Guttman method is not successful in the analysis of the singularities in the ccp model at 0.75 and 0.781 25, their locations are easily found by an examination of symmetry points in the Brillouin zone. In these cases we can attempt to determine the exponents by trying linear least squares with different functional forms for the asymptotic behavior of the modified moments, using the "goodness of fit" as a criterion for which form is best. The success of this procedure is dependent upon limiting the possible functional forms by physical considerations to a reasonable number of choices.

The locations of the singularities can also be treated as variables in the least-squares fitting procedure. In this case the problem becomes nonlinear. A very fast nonlinear least-squares algorithm has been given by Golub and Pereyra[34] which makes explicit use of the fact that some of the unknowns enter linearly. With this algorithm and reasonable initial estimates for the locations of the singularities (e.g., from an inspection of the rational approximation or from the Joyce-Guttman method), quite good estimates for the locations can be obtained.

Finally, the results from the least-squares fitting procedure can sometimes be improved by using a linear combination of functional forms for the asymptotic contribution of a singularity. This technique is important for some complicated singularities (see the discussion below of the singularity at 0.75 in the ccp model). It can also be used to include the second-order contribution from a singularity to the asymptotic form of the modified moments. [This is equivalent to finding the *second* coefficient in a power-series expansion for $g(z)$ in Eq. (8.1).]

We have investigated least-squares analysis methods for singularities in some detail for the ccp model. A combination of the techniques described above was used to determine the correct functional forms for the singularities at 0.5 and 0.75 and to test the nonlinear least-squares method for determining locations.

The singularities at 0.25, 0.781 25, and 0.877⋯ were taken to be S_2, S_2, and S_1, respectively. A least-squares weighting of $w(k)=k^5$ was used. The best result for the singularity at 0.5 was obtained when a discontinuity plus a second-order contribution from a discontinuity in slope (the first generalized singularity in Table V) was used. The best result for the remaining singularity, at 0.75, was obtained when a linear combination of the two generalized singularities in Table V was used.

It has been reported that the singularity at 0.5 contains a logarithmic divergence.[32] To investigate this possibility, we tried a linear combination of the two two-dimensional singularities in Table V. [This is equivalent to determining the phase, η_i, at the singularity following Eq. (8.4).] The resulting magnitude of the coefficient for the logarithm was less than 1% of that for the discontinuity—zero to within the accuracy with which magnitudes are typically determined. This is in agreement with a detailed analysis of this singularity using the dynamical matrix which indicates a discontinuity with no logarithmic contribution.[33]

A variety of functional forms were tried for the singularity at 0.75, including a three-dimensional maximum and minimum, and the two generalized singularities in Table V. A much better fit was obtained with either of the generalized singularities than with a maximum or minimum. In agreement with a preliminary dynamical matrix calculation indicating the presence of both singularities,[33] the best fit was obtained using a linear combination of the two.

As an example of the nonlinear least-squares method, in Table IX we give the results obtained using this technique for the ccp model. Forty modified moments were fitted with all of the locations of the singularities treated as variables to be determined. In Fig. 9 the corresponding density obtained using the moment-singularity method is shown. While the exact solution for this model is not known, Fig. 9 is in excellent agreement with results from root sampling methods.[33]

The nonlinear least-squares technique determines the locations of the singularities in the ccp model with an error of less than 1.0×10^{-3}. Most of the constants A_i and B_i in Table IX are not known exactly. However, A_2 has been determined to be 0.3417⋯[33] so that the least-squares result for this constant is correct to about one part per thousand. A comparison of the results in Table IX with those obtained using different numbers of moments and those from linear least squares suggests that A_1 is accurate to within a few parts per thousand, and that A_4 and A_5 are accurate to a few percent, while A_3 and B_3 are probably only reliable to about 10%. The variations in A_3 and B_3 seem to be correlated so that the representation of the singularity at 0.75 is probably more faithful than the reliability of these constants might suggest.

The least-squares methods we have described have the advantage of providing considerable flexibility. Some or all of the locations may be fixed in advance, more than one type of singularity may be assigned to a given (variable) location, and the phase may be treated as a discrete or continuous variable. On the other hand, the convergence of the least-squares results is not exceedingly rapid. While a quite good representation of $G(x)$ is ob-

TABLE IX. Singularities in the density for the ccp model and their dominant contributions to the modified moments as determined by nonlinear least squares.

			$(\nu_k^*)_i^s = A_i(k+1)^{-(p+1)}\sin[(k+1)\theta_i+\eta_i] + B_i\gamma_i(k+1)$			
i	$x_i=\sin^2\frac{1}{2}\theta_i$	p	A_i	η_i	B_i	$\gamma_i(k+1)$
1	0.250 22	$+\frac{1}{2}$	1.0046	$+\frac{1}{4}\pi$	⋯	⋯
2	0.500 04	0	0.3414	$+\frac{1}{2}\pi$	0.0890	$(k+1)^{-2}\sin[(k+1)\theta_c]$
3	0.749 05	+1	2.1639	π	−0.8330	$(k+1)^{-2}\{(2/\pi)\ln(k+1)\sin[(k+1)\theta_c]+\cos[(k+1)\theta_c]\}$
4	0.782 22	$+\frac{1}{2}$	0.6522	$+\frac{1}{4}\pi$	⋯	⋯
5	0.877 87	$+\frac{1}{2}$	1.2844	$-\frac{1}{4}\pi$	⋯	⋯

tained from 30 moments, the results from 60 moments are not dramatically better than those using 40 moments. It would, of course, be highly desirable to have a method for determining the coefficients which combined the flexibility of the least-squares methods with the rapid convergence of the series-analysis methods of Joyce and Guttman.

IX. CONCLUSION

We have given a number of methods which can be used to extract information about a density function from its modified moments. By exploiting the connections between Chebyshev expansions, Fourier series, and power series, we have, we believe, made a significant advance in the practical problem of estimating a spectral density from a limited number of moments. It seems certain, however, that the possibilities offered by these connections have not been exhausted. We anticipate that both more sophisticated numerical methods and more rigorous analytical results will be discovered.

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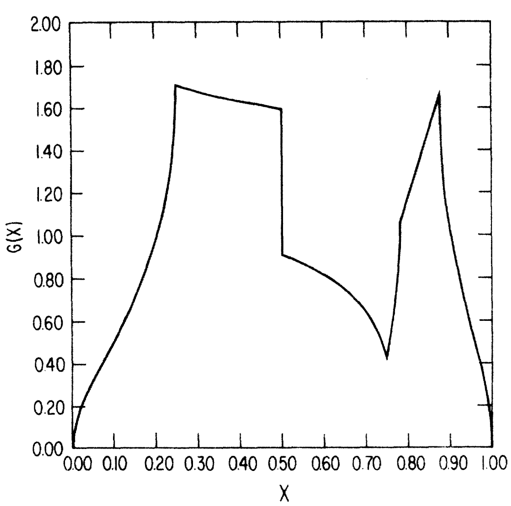

FIG. 9. Moment-singularity method for the spectral density of the ccp model. Result is obtained using the nonlinear least-squares results shown in Table IX and 40 modified moments.

APPENDIX A

For three-dimensional harmonic solids, the continued fraction extrapolation as proposed by Gordon[7] has the disadvantage that it does not have the correct frequency dependence at the low-frequency end of the interval. We give below a slightly modified version of Gordon's procedure which does not have this difficulty.

Let $\mathcal{G}(z)$ be a function of the complex variable z defined by

$$\mathcal{G}(z) \equiv \int_0^1 \frac{G(x)}{z-x}\, dx . \tag{A1}$$

Then,

$$\operatorname{Im}\{\lim_{\epsilon\to 0^+}(1/\pi)\mathcal{G}(x_0+i\epsilon)\} = G(x_0) . \tag{A2}$$

$\mathcal{G}(z)$ can be represented by the power moment expansion

$$\mathcal{G}(z) \sim \sum_{k=0}^{\infty} \mu_k z^{-(k+1)} , \tag{A3}$$

or by the associated continued fraction

$$\mathcal{G}(z) \sim \cfrac{\beta_0}{z-\alpha_0-\cfrac{\beta_1}{z-\alpha_1-\cfrac{\beta_2}{z-\alpha_2\cdots}}} . \tag{A4}$$

The coefficients α_k, β_k in the continued fraction are the recursion coefficients for the monic orthogonal polynomials defined by $G(x)$. That is, they define a set of polynomials, $\pi_k(x)$ [as in Eq. (2.3) with $a_k=\alpha_k$ and $b_k=\beta_k$] which satisfy

$$\int_0^1 \pi_k(x)\pi_l(x)G(x)\, dx = \delta_{kl}N_k . \tag{A5}$$

These recursion coefficients are determined by the moments of $G(x)$; the first n α's and n β's can be obtained from the first $2n$ moments. However, the transformation from power moments to recursion coefficients is exponentially ill conditioned.[35] (This is the ill-conditioned problem we noted in Sec. III.) On the other hand, the transformation from modified moments to recursion coefficients is usually well conditioned.[35] A stable procedure by which the latter transformation can be accomplished is given in Ref. 3.

If the continued fraction (A4) is truncated at any finite order and the limit (A2) is taken to obtain an approximate density, then the result is a sum of weighted delta functions. (This approximate density is equivalent to a Gaussian quadrature formula.[36]) Thus, unless the continued fraction can be summed to all orders, it does not provide a continuous approximation to the density. However, if the coefficients α_k, β_k approach well-defined limits as $k\to\infty$, then a continued fraction for which a limited number of coefficients are known can be extrapolated by assuming that all the unknown coefficients have attained their limiting values.

For the squared-frequency density of three-dimensional harmonic solids it is typically the case that

$$\alpha_k \to \tfrac{1}{2},\ \ \beta_k \to \tfrac{1}{16},\ \ \text{as } k\to\infty. \tag{A6}$$

In order to evaluate an extrapolated continued fraction with these limiting values for the coefficients, we define

$$\mathcal{H}(z) \equiv \int_0^1 \frac{H(x)}{z-x}\,dx, \tag{A7}$$

where

$$H(x) = (8/\pi)[x(1-x)]^{1/2} \tag{A8}$$

is the weight function for the shifted Chebyshev polynomials of the second kind. The associated continued fraction for $\mathcal{H}(z)$ is

$$\mathcal{H}(z) \sim \cfrac{1}{z-\frac{1}{2}-\cfrac{\frac{1}{16}}{z-\frac{1}{2}-\cfrac{\frac{1}{16}}{z-\frac{1}{2}\cdots}}}. \tag{A9}$$

That is, all the α_k are $\frac{1}{2}$ and all the β_k are $\frac{1}{16}$ [cf, Eq. (2.9)]. By standard methods of analysis,

$$\lim_{\epsilon\to 0^+} \tfrac{1}{16}\mathcal{H}(x_0+i\epsilon) = \{(\tfrac{1}{2}x_0-\tfrac{1}{4}) - \tfrac{1}{2}i[x(1-x)]^{1/2}\}. \tag{A10}$$

If α_n and β_n are the last-known recursion coefficients for $G(x)$ and we define

$$\mathcal{G}_n(z) \equiv \cfrac{\beta_0}{z-\alpha_0-\cfrac{\beta_1}{z-\alpha_1 \ddots -\cfrac{\beta_n}{z-\alpha_n-\frac{1}{16}\mathcal{H}(z)}}}, \tag{A11}$$

then the corresponding density, $G_n(x)$, defined by Eq. (A2) is given by

$$G_n(x) = \operatorname{Im}\left(\cfrac{\beta_0/\pi}{x-\alpha_0-\cfrac{\beta_1}{x-\alpha_1 \ddots -\cfrac{\beta_n}{\frac{1}{2}x-\alpha_n+\frac{1}{4}+\frac{1}{2}i[x(1-x)]^{1/2}}}}\right). \tag{A12}$$

The approximate density $G_n(x)$ is non-negative and has the same first $2n+2$ moments as $G(x)$.[7] Also, it is proportional to $[x(1-x)]^{1/2}$ and therefore has the correct limiting behavior for a three-dimensional harmonic solid at both $x=0$ and $x=1$.

APPENDIX B

We give here a very general proof of the convergence of expansion (4.2) for a time-autocorrelation function in terms of the modified moments of its spectral density whenever that density and the weight function $H(x)$ are restricted to finite intervals. The proof is independent of the choice of the non-negative normalized weight function $H(x)$ and makes no assumptions about $G(x)$ save that it is non-negative and zero outside of some finite interval. In particular, the intervals of definition of $G(x)$ and $H(x)$ need not be the same. Some remarks on the convergence for infinite intervals are given at the end of the appendix.

When the spectral densities $G(x)$ and $H(x)$ are restricted to finite intervals, the problem may always be rescaled so that $H(x)$ is defined on the unit interval. Let the resulting interval of definition of $G(x)$ be $[0, a^2]$. With this scaling, we wish to approximate the integral

$$C(t) = \int_0^{a^2} G(x)\cos(x^{1/2}\tau)\,dx \equiv \int_{-a}^{a} I(w)\cos(w\tau)\,dw, \tag{B1}$$

$$I(w) \equiv I(-w) \equiv wG(w^2) \quad (0 \leq w \leq a),$$

by the sum

$$C_N(t) = \sum_{k=0}^{N} \nu_k^* F_k(\tau), \tag{B2}$$

where

$$F_k(\tau) = \int_0^1 H(x)p_k^*(x)\cos(x^{1/2}\tau)\,dx,$$

$$\nu_k^* = \int_0^a G(x)p_k^*(x)\,dx, \tag{B3}$$

$$\int_0^1 H(x)p_k^*(x)p_j^*(x)\,dx = \delta_{kj}.$$

It will be convenient to define the function

$$K(w) = K(-w) = wH(w^2) \quad (0 \leq w \leq 1), \tag{B4}$$

which has the property

$$\int_{-1}^{1} K(w)p_k^*(w^2)p_j^*(w^2)\,dw = \delta_{kj}. \tag{B5}$$

Thus, the orthonormal polynomials $p_k^\dagger(w)$, belonging to $K(w)$, have the properties

$p_{2k}^\dagger(w) = p_k^*(w^2)$ (containing only even powers),

$p_{2k+1}^\dagger(w) =$ a polynomial containing only odd powers.

Therefore, we may write

$$F_k(\tau) = \int_{-1}^{1} K(w)p_{2k}^\dagger(w)\cos w\tau\,dw. \tag{B6}$$

Now $\cos(w\tau)$ is an entire function of w. Consequently, by a theorem due to Bernstein,[37] for each fixed time τ there exists a polynomial $Q_{2k-1}(w)$ of *best approximation* to $\cos(w\tau)$ which is of degree

$2k-1$ and which satisfies the identity

$$\cos(w\tau)=Q_{2n-1}(w)+\frac{\cos[\lambda(w)]\tau^{2k}}{2^{2k-1}(2k)!}\,,\qquad \lambda(w)\in[0,\pi],\quad w\in[-1,1]. \tag{B7}$$

By the orthogonality of the $p_k^\dagger(w)$ over $K(w)$ it follows that

$$F_k(\tau)=\int_{-1}^{1}K(w)p_{2k}^\dagger(w)\left(\frac{\cos[\lambda(w)]\tau^{2k}}{2^{2k-1}(2k)!}\right)dw\,. \tag{B8}$$

Taking absolute values and making use of the inequality $(2|p|\leq 1+p^2)$, we obtain the inequality

$$|F_k(\tau)|\leq\frac{2\tau^{2k}}{2^{2k}(2k)!}\leq\left(\frac{e\tau}{4k}\right)^{2k}. \tag{B9}$$

This bound is somewhat stronger than is really necessary for the proof here. A weaker but sufficient bound can be obtained from Taylor's theorem. The inequality (B9) is valuable, however, in establishing the useful range of the partial sum (B2) when estimates of the magnitudes of the modified moments are available.

It remains to be shown that regardless of the choice of $H(x)$ and the behavior of $G(x)$, the modified moments ν_k^* cannot grow rapidly enough to destroy the convergence of (B2). This is most easily accomplished by the following indirect argument. Consider the expansion of $\cos(w\tau)$ at any fixed τ in the orthogonal polynomials $p_k^\dagger(w)$

$$\cos(w\tau)=\sum_{k=0}^{\infty}f_k(\tau)p_k^\dagger(w)\,,\qquad f_k(\tau)=\int_{-1}^{1}K(w)p_k^\dagger(w)\cos(wt)\,. \tag{B10}$$

From (B3)–(B6) we have

$$f_{2k}(\tau)=F_k(\tau),\quad f_{2k+1}(\tau)=0\,. \tag{B11}$$

By a theorem due to Szego[38] this expansion converges *uniformly* inside any ellipse in the complex w plane with foci at ±1 and with the sum of its semiaxes less than

$$R\equiv\lim_{n\to\infty}\inf|f_n|^{-1/n}. \tag{B12}$$

By Eq. (B9), R is infinite. Thus, for any fixed τ, the series (B10) converges uniformly in any bounded region of the complex w plane, in particular on the interval $[-a,a]$. Consequently, integration of (B10) over $I(w)$ may be performed term by term, which, with (B11), establishes the convergence of the partial sums (B2) immediately.

When a scaling of the interval is required to transform $H(x)$ to the interval $[0,1]$, the bound (B9) will be altered by a multiplicative factor, b^k. This does not alter the limit (B12), however, so that convergence is unaffected.

Much less is known about the convergence of expansions in general orthogonal polynomials on an infinite interval than for the finite case. However, if $H(x)$ is taken to be one of the classical weight functions [for example, those defining the Hermite or (generalized) Laguerre polynomials], and if $G(x)/H(x)$ is bounded by some constant for sufficiently large x, then convergence of the expansion for $C(t)$ is easily established.

APPENDIX C

The sums (7.3) to the "most singular" part of the density are rather slowly convergent series. We show here how they can be evaluated, for each of the singularities in Table V, either in closed form or as rapidly convergent series.

Each of the required sums can be expressed in terms of the function

$$\Phi(z,s,v)=\sum_{n=0}^{\infty}(n+v)^{-s}z^n\,, \tag{C1}$$

the properties of which are discussed in standard works on special functions.[39] The required sums are of the form

$$\sum_{k=0}^{\infty}(k+1)^{-s}\sin[(k+1)\theta_c+\eta]\sin(k+1)\theta =\tfrac{1}{2}\sum_{k=0}^{\infty}(k+1)^{-s}\{\cos[(k+1)(\theta_c-\theta)+\eta]-\cos[(k+1)(\theta_c+\theta)+\eta]\}. \tag{C2}$$

The sums of cosines may be expressed in terms of Φ through the relation

$$\sum_{k=0}^{\infty}(k+1)^{-s}\cos[(k+1)\phi+\eta]=\mathrm{Re}[e^{i\eta}z\Phi(z,s,1)]\,\big|_{z=e^{i\phi}}\,. \tag{C3}$$

For noninteger s the function Φ satisfies the identity

$$e^{i\phi}\Phi(e^{i\phi},s,1)=\Gamma(1-s)(-i\phi)^{s-1}+\sum_{r=0}^{\infty}\zeta(s-r)\frac{(i\phi)^r}{r!}\,,\qquad |\phi|<2\pi,\ s\neq1,2,3,\ldots, \tag{C4}$$

where $\zeta(z)$ is the Riemann zeta function. For $s=m$, an integer,

$$e^{i\phi}\Phi(e^{i\phi},m,1)=\frac{(i\phi)^{m-1}}{(m-1)!}\left[\left(\sum_{k=1}^{m-1}k^{-1}\right)-\ln(-i\phi)\right]+\sum_{n=0}^{\infty}{}'\zeta(m-n)\frac{(i\phi)^n}{n!}\quad(|\phi|<2\pi;\ m=2,3,4,\ldots), \tag{C5}$$

where the prime indicates that the term with $(m-n)=1$ is omitted, or, when $m=1$,

$$e^{i\phi}\Phi(e^{i\phi},1,1)=-\ln(1-e^{i\phi}) \quad (|\phi|<2\pi). \tag{C6}$$

Equations (C4)–(C6) may be put in more convenient form by making use of the identities

$$\zeta(s)=2(2\pi)\Gamma(1-s)\sin(\tfrac{1}{2}\pi s)\zeta(1-s), \tag{C7}$$

$$\Gamma(1-s)\Gamma(s)=\pi/\sin(\pi s). \tag{C8}$$

Combining these with (C5) and (C6) we obtain the sums required for the two-dimensional singularities and the first generalized singularity in Table V

$$\sum_{k=0}^{\infty}(k+1)^{-1}\cos[(k+1)\phi+\eta]=-(\cos\eta)\ln[2(1-\cos\phi)]+(\sin\eta)[\tfrac{1}{2}\pi\,\mathrm{sgn}(\phi)-\tfrac{1}{2}\phi] \quad (|\phi|<2\pi), \tag{C9}$$

$$\sum_{k=0}^{\infty}(k+1)^{-2}\cos(k+1)\phi=\tfrac{1}{6}\pi^2-\tfrac{1}{2}\pi|\phi|+\tfrac{1}{4}\phi^2 \quad (|\phi|<2\pi), \tag{C10}$$

where $\phi=(\theta_c\pm\theta)$.

Applying (C7) and (C8) to (C4), the sum (C3) for noninteger s becomes

$$\sum_{k=0}^{\infty}(k+1)^{s}\cos[(k+1)\phi+\eta]=\Gamma(1-s)\{|\phi|^{s-1}\cos[\eta-\tfrac{1}{2}(s-1)\pi\,\mathrm{sgn}(\phi)]+F_{s-1}(\phi)\cos[\eta+\tfrac{1}{2}(s-1)\pi] + F_{s-1}(-\phi)\cos[\eta-\tfrac{1}{2}(s-1)\pi]\}, \tag{C11}$$

where

$$F_p(\phi)\equiv(2\pi)^p\sum_{n=0}^{\infty}\frac{\Gamma(n-p)}{\Gamma(-p)n!}\zeta(n-p)\left(\frac{\phi}{2\pi}\right)^n \quad (|\phi|<2\pi). \tag{C12}$$

Since the sum of cosines in (C11) is periodic, ϕ may always be chosen to lie in $[-\pi,\pi]$. Because $\zeta(n-p)$ rapidly approaches unity for large n, the series always converges exponentially for $|\phi|\le\pi$. Even more rapid convergence of (C12) can be obtained by noting that $[\Gamma(n-p)/\Gamma(-p)n!]$ is just the coefficient of x^n in the binomial expansion of $(1-x)^p$ so that

$$F_p(\phi)=(2\pi-\phi)^p+(2\pi)^p\sum_{n=0}^{\infty}\frac{\Gamma(n-p)}{\Gamma(-p)n!}\times[\zeta(n-p)-1]\left(\frac{\phi}{2\pi}\right)^n. \tag{C13}$$

This procedure may be repeated l times to give

$$F_p(\phi)=\sum_{k=1}^{l}(2\pi k-\phi)^p+(2\pi)^p\sum_{n=0}^{\infty}\frac{\Gamma(n-p)}{\Gamma(-p)n!}\times\left[\zeta(n-p)-\sum_{k=1}^{l}k^{p-n}\right]\left(\frac{\phi}{2\pi}\right)^n. \tag{C14}$$

An extremely rapidly convergent series can be obtained in this way. As an example, for the three-dimensional singularities in Table V ($p=\frac{3}{2}$), when $l=7$, eight terms in the series give $F_p(\phi)$ to one part in 10^{10} on $[0,\pi]$.

The required Riemann zeta functions of noninteger argument are easily and rapidly computable by standard methods or can be found in tables.[40] For the singularities in Table V only the Riemann zeta functions of integer and half-integer arguments are required.

The second generalized singularity in Table V,

$$(\nu_k^*)^s=(k+1)^{-2}[(2/\pi)\ln(k+1)\sin(k+1)\theta_c \pm\cos(k+1)\theta_c], \tag{C15}$$

requires somewhat more attention. The contribution from the second term is obtained from (C5) with $m=2$. That from the first is obtained by differentiating (C3) and (C4) with respect to s and letting s approach 2. In the same way that (C5) is obtained as a limit of (C4), a finite limit to the derivative is obtained. The combined result may be written in the form

$$\sum_{k=0}^{\infty}(k+1)^{-2}\left[\frac{2}{\pi}\ln(k+1)\cos(k+1)\phi\pm\sin(k+1)\phi\right]=\left(\frac{2}{\pi}\sum_{k=1}^{\infty}k^{-2}\ln k\right)+|\phi|\ln|\phi|[1\pm\mathrm{sgn}(\phi)]-\phi[1\pm\mathrm{sgn}(\phi)-\gamma] -2\pi\log(2\pi)\left(\frac{\phi}{2\pi}\right)+4\pi\sum_{k=2}^{\infty}[k(k+1)]^{-1}\zeta(k)\left(\pm\frac{\phi}{2\pi}\right)^{k+1}, \tag{C16}$$

where $\gamma=0.577\,215\,6\cdots$ is Euler's constant and the sum $\sum k^{-2}\ln k$ is easily evaluated by the Euler-Maclauren formula to be $0.937\,548\,25\cdots$.

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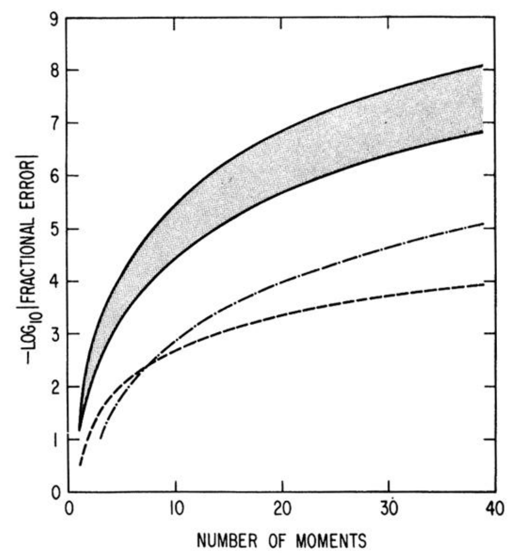

FIG. 3. Accuracy of methods for computing averages over the density. The negative of $\log_{10}(|\text{fractional error}|)$ for $\mu_{1/2}$ in the ccp model is plotted against the number of moments used. Envelope bounded by solid lines encloses results from modified moment expansion; dashed line shows results from shifted moment expansion; dotted line shows differences between bounds obtained from Gaussian quadratures.

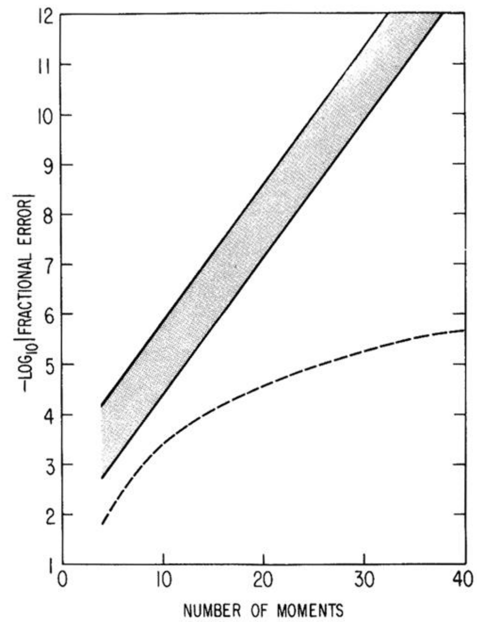

FIG. 4. Accuracy of nonlinear extrapolation of expansions for averages over the density. The negative of $\log_{10}(|\text{fractional error}|)$ for $\mu_{1/2}$ in the ccp model is plotted against the number of moments used. Envelope bounded by solid lines encloses results from modified moment expansion; dashed line shows results from shifted moment expansion.