Scattering properties of the leveled-wave model of random morphologies

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In the leveled-wave model, random porous morphologies are simulated by discrete maps of continuous stochastic standing waves generated by adding sinusoids over suitable distributions of wave vectors and phase constants. The mathematical structures obtained this way appear to be useful models for certain kinds of random geometries, such as the bicontinuous morphologies that occur in microemulsions and porous glasses. Properties of the leveled-wave method are developed, and the scattering behavior is derived. Scattering from an arbitrary three-phase system can be analyzed by adding different instances of scattering from a generalized film morphology in which the scattering phase is confined to the interior of the interspatial region bounded by two leveled-wave interfaces that are separated everywhere. General results for this interspace scattering are derived. Fractal surfaces, dimension $2 \leq D \leq 3$, are incorporated into the leveled-wave scheme by using a wave-number distribution having an appropriate long-tailed asymptotic behavior. Scenarios are discussed for the scattering as $D \to 2$ from above and for $D \to 3$ from below. For $D = 3$ the asymptotic scattering falls faster than algebraically as the scattering wave vector $Q \to \infty$.

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I. INTRODUCTION

This paper follows the author's first discussion of the leveled-wave model for random morphologies, hereafter referred to as I [1]. The leveled-wave approach to such structures was conceived by Cahn [2] originally as a simulation technique for systems, such as phase-separated porous glasses, whose morphology was thought to be influenced by spinodal decomposition. Some time passed, however, before Cahn's method received its first mathematical treatment by Hopper [3]. The subsequent analysis in I was independent of Hopper's treatment and turned out to be substantially different in methods and motivation, although not in its conclusions, where comparison is easy. An important facet of I was the notion that the mathematical principles implicit in Cahn's idea were significant beyond the original context of spinodal decomposition and might usefully apply to a wider range of applications in which morphology was constrained by a well-defined length scale, whatever its physical origins might be. In particular, the leveled-wave model was advanced as a means for rationalizing the striking contrast variation of small-angle-scattering experiments [4, 5] on microemulsions believed to exhibit random bicontinuous microstructure, in line with Scriven's [6] much earlier suggestion of linking Cahn's model to microemulsions. This also entailed generalizing the method to provide a means of describing structures such as surfactant films—or surface layers in other contexts—having finite volume fraction. The ultimate role this approach may play in the understanding of microemulsion morphology remains to be seen, but the idea continues to receive attention [7–9]. Chen, Chang, and Strey [8, 9], in particular, have critically examined the leveled-wave method in the analysis of small-angle scattering from microemulsions. The model has also been applied recently to small-angle scattering from high-porosity controlled-pore glasses [10] and from porous silica glasses, both dry and imbibed with surfactant-bearing solution [11].

The present discussion will focus, in Sec. III, on the asymptotic behavior of the small-angle scattering from leveled-wave morphologies, which was left implicit in I. In particular we describe how fractal interfaces can be incorporated into the methodology, thereby extending its application to structures without a length scale. Beyond the mathematical interest of the technique, it may be relevant to the observed deviations from Porod's law in small-angle-scattering experiments on various porous glass systems [12, 13], which usually are attributed to fractal roughness. A model that adds fractal roughness to the surfaces of a bicontinuous structure in a geometrically consistent way has been lacking up to now. The development in Sec. III, while straightforward, needs preparation, so in Sec. II and in the Appendixes the main ingredients of the leveled-wave model—as defined by the approach of I—are laid out. While that account is more or less complete in its essentials, this opportunity is used to expand on important points.

II. THE LEVELED-WAVE MODEL

A. Density functions

In the leveled-wave scheme continuous interfaces are mathematically modeled by level sets—two-dimensional contours—of a stochastic standing wave $S(\mathbf{r})$, which is defined by

$$S(\mathbf{r}) = \frac{1}{\sqrt{N\langle A^2\rangle}} \sum_{n=1}^{N} A_n \cos(\mathbf{k}_n \cdot \mathbf{r} + \phi_n) , \qquad (1)$$

where the subscripted quantities on the right are taken to be independent random variables. Thus an interface between two material phases of uniform densities ρ_1 and ρ_0 is associated in the construction with an α set of the wave in (1), i.e., the point set on which $S(\mathbf{r}) = \alpha$, the value of α being chosen to give the required volume fractions for the two partitions. For example, the 0 set of $S(\mathbf{r})$ separates two equal subvolumes in the absence of other interfaces, since $S(\mathbf{r})$ is equally likely to be positive or negative everywhere. A corresponding density function $\rho(\mathbf{r})$ can be defined pointwise by $\rho(\mathbf{r}) = \rho_0$ or $\rho(\mathbf{r}) = \rho_1$, say, according to whether $S(\mathbf{r}) > \alpha$ or $S(\mathbf{r}) < \alpha$, respectively. One way of writing this function is

$$\rho(\mathbf{r}) = (\rho_0 - \rho_1)\Theta(S(\mathbf{r}) - \alpha) + \rho_1 , \tag{2}$$

where $\Theta(x)$ is the Heaviside function: $\Theta(S) = 1$ for $S \geq 0$, and 0 otherwise.

In the limit $N \to \infty$ the properties of the leveled-wave morphology are independent of the distribution of A_n, as long as $\langle A^2 \rangle$ exists [1]. Thus in subsequent references to Eq. (1) we may assume that $A_n = 1$ and that N is very large. The phase constants ϕ_n are chosen from a uniform distribution over $(0, 2\pi)$. This makes $S(\mathbf{r})$ spatially homogeneous; no special value of $\mathbf{r}$, including the origin, is singled out. For an isotropic morphology, the directions of the random wave vectors $\mathbf{k}_n$ are uniformly distributed over solid angle 4π. All other properties of the construction are then determined by the distribution $P(k)$ of the magnitudes of $\mathbf{k}_n$, which is normalized by

$$\int_0^\infty P(k)\, dk = 1 . \tag{3}$$

There are no *a priori* requirements on $P(k)$ other than (3), although applications of the scheme are likely to entail k distributions having fairly well-defined peaks or similar structure. Indeed it is remarkable that the prototype leveled-wave morphology, which emerges from the restriction

$$P(k) = \delta(k - k_0) , \tag{4}$$

is geometrically interesting and evocative of realistic random bicontinuous structures, giving evidence of the topological relevance of well-defined length scales, as fixed here by k_0, in such morphologies. Nevertheless, recent applications of the scheme to scattering measurements have pointed out the necessity of considering k distributions showing significant dispersion [8–11]. Later in this discussion we will see how the behavior of $P(k)$ as $k \to \infty$ provides a mechanism for incorporating certain kinds of interfacial roughness into the model.

Now it proves useful to rewrite Eq. (2) in a slightly more generalized form as

$$\rho(\mathbf{r}) = \lim_{\beta \to \infty} \rho^{(2)}_{\alpha,\beta}(\mathbf{r}) , \tag{5}$$

where

$$\rho^{(2)}_{\alpha,\beta}(\mathbf{r}) = (\rho_0 - \rho_1)\Theta_{\alpha,\beta}(S(\mathbf{r})) + \rho_1 , \tag{6}$$

and $\Theta_{\alpha,\beta}(S)$ is the indicator function for the (half-closed) $\alpha\beta$ interval

$$\Theta_{\alpha,\beta}(x) = \begin{cases} 1 \text{ if } S \in [\alpha, \beta), \ \alpha < \beta \\ 0 \text{ otherwise.} \end{cases} \tag{7}$$

The density function $\rho^{(2)}_{\alpha,\beta}(\mathbf{r})$ actually represents a special case of two-phase morphology in which the density has the value ρ_0 at all points in the interspace "sandwiched" between the α-set and β-set interfaces of $S(\mathbf{r})$ and the value ρ_1 elsewhere. The volume fraction c_0 occupied by the $\alpha\beta$ interspace is given by

$$\begin{aligned} c_0 &= \langle \Theta_{\alpha,\beta}(S(\mathbf{r})) \rangle \\ &= \frac{1}{2}[\mathrm{erf}(\beta) - \mathrm{erf}(\alpha)] , \end{aligned} \tag{8}$$

where $\mathrm{erf}(x)$ is the error function. This formula may be inferred from the fact that as $N \to \infty$, the unleveled wave $S(\mathbf{r})$, being a sum of independent random variables, tends to a Gaussian random variable at each $\mathbf{r}$; alternatively, it can be derived explicitly from a later stage of the formalism (Appendix A). For ease of discussion, we may call the volume complementary to the interspace the "bulk," which comprises two nonintersecting subvolumes separated everywhere by the interspace. This terminology reflects the original motivation [1] in which the $\alpha\beta$ interspace is advanced as a model for a film or layer in a random bicontinuous medium. The volume fractions of the two bulk partitions can be obtained as special cases of Eq. (8): ignoring sets of measure 0, the bulk partition in contact with the α interface is equivalent to an $\alpha'\beta'$ interspace with $\alpha' \to -\infty$, $\beta' = \alpha$, while that in contact with the β interface is equivalent to an instance having $\alpha' = \beta$, $\beta' \to \infty$. Thus we have

$$\begin{aligned} c_1 &= \frac{1}{2}[1 + \mathrm{erf}(\alpha)] , \\ c_2 &= \frac{1}{2}[1 - \mathrm{erf}(\beta)] , \end{aligned} \tag{9}$$

respectively, for these two partitions, and we note that $c_0 + c_1 + c_2 = 1$.

If now we take $\beta \to \infty$ in Eq. (5), the $\alpha\beta$ interspace expands to fill up the portion of the bulk in contact with the β interface, and we retrieve the morphology for the case of a single α interface. More generally, still, the bulk partitions in contact with the α and β interfaces can be assigned different material densities ρ_1 and ρ_2, respectively, while the interspace continues to have a third, distinct density ρ_0. The density function for this three-phase system can be represented by

$$\rho^{(3)}_{\alpha,\beta}(\mathbf{r}) = \rho_{01}\Theta_{\alpha,\beta}(S(\mathbf{r})) + \rho_{21}\Theta_{\beta,\infty}(S(\mathbf{r})) + \rho_1 , \tag{10}$$

where $\rho_{ij} = \rho_i - \rho_j$.

The autocorrelation function of the three-phase density is defined by

$$\Gamma^{(3)}_{\alpha,\beta}(\mathbf{r}) = \langle \rho^{(3)}_{\alpha,\beta}(\mathbf{r})\rho^{(3)}_{\alpha,\beta}(\mathbf{0}) \rangle , \tag{11}$$

where the angular brackets denote an average over the random-parameter space. The explicit representation of (11) will turn out to involve the autocorrelation function for an arbitrary $\alpha\beta$ interspace,

$$\Gamma_{\alpha,\beta}(\mathbf{r}) = \langle \Theta_{\alpha,\beta}(S(\mathbf{r}))\Theta_{\alpha,\beta}(S(\mathbf{0}))\rangle\,, \tag{12}$$

where

$$\Gamma_{\alpha,\beta}(\mathbf{0}) = \langle \Theta_{\alpha,\beta}(S(\mathbf{0}))\rangle = c_0\,. \tag{13}$$

Equivalently, this is the autocorrelation of $\rho^{(3)}_{\alpha,\beta}(\mathbf{r})$ with the particular densities $\rho_0 = 1$ and $\rho_1 = \rho_2 = 0$, or of $\rho^{(2)}_{\alpha,\beta}(\mathbf{r})$ with $\rho_0 = 1$ and $\rho_1 = 0$. Substituting (10) into (11), and using (12) one can obtain

$$\begin{aligned}\Gamma^{(3)}(\mathbf{r};\alpha,\beta) = {} & \rho_{01}\rho_{02}\Gamma_{\alpha,\beta}(\mathbf{r}) \\ & + \rho_{01}\rho_{21}\Gamma_{\alpha,\infty}(\mathbf{r}) + \rho_{20}\rho_{21}\Gamma_{\beta,\infty}(\mathbf{r}) \\ & + (2c_0\rho_{01} + 2c_2\rho_{21} + \rho_1)\rho_1\,. \end{aligned}\tag{14}$$

Actually, to get (14) we also need the step

$$\begin{aligned}&\langle \Theta_{\alpha,\beta}(S(\mathbf{r}))\Theta_{\beta,\infty}(S(\mathbf{0}))\rangle + \langle \Theta_{\alpha,\beta}(S(\mathbf{0}))\Theta_{\beta,\infty}(S(\mathbf{r}))\rangle \\ &\qquad = \Gamma_{\alpha,\infty}(\mathbf{r}) - \Gamma_{\beta,\infty}(\mathbf{r}) - \Gamma_{\alpha,\beta}(\mathbf{r})\,, \end{aligned}\tag{15}$$

which follows easily from the union property

$$\Theta_{\alpha,\infty}(S) = \Theta_{\alpha,\beta}(S) + \Theta_{\beta,\infty}(S)\,. \tag{16}$$

If we reinterpret the density factors ρ_i as scattering densities for the appropriate kind of radiation, then the small-angle scattering from the three-phase leveled-wave structure is obtained from the Fourier transform of (14),

$$I^{(3)}(Q) = 4\pi V\int_0^\infty \Gamma^{(3)}_{\alpha,\beta}(r) j_0(Qr) r^2\,dr\,, \tag{17}$$

for isotropic $\Gamma^{(3)}_{\alpha,\beta}(\mathbf{r})$. In (17), V is the volume of the scattering system. We see from (14) that in the leveled-wave model, the autocorrelation of the three-phase density—and thus the scattering—is determined by the autocorrelation function of the generalized interspace, i.e., the function $\Gamma_{\alpha',\beta'}(\mathbf{r})$ in the three instances: $\alpha',\beta' = \alpha,\beta$; $\alpha',\beta' = \alpha,\infty$; and $\alpha',\beta' = \beta,\infty$. Because of (15) and similar identities, cross correlations among the volume partitions are expressible as linear combinations of this family of autocorrelations. The analysis of $\Gamma_{\alpha,\beta}(\mathbf{r})$ for arbitrary α and β thus provides a formally complete description of the scattering properties of the leveled-wave model.

B. Scattering properties

A usable development of Eq. (12) can begin with the Fourier representation of the indicator function

$$\Theta_{\alpha,\beta}(S) = \frac{1}{2\pi i}\int_{-\infty}^{\infty} dx \frac{1}{x}(e^{i\beta x} - e^{i\alpha x})e^{-iSx}\,. \tag{18}$$

Substitution of this into (12) at once brings in the quantity

$$\langle\{-i[xS(\mathbf{r}) + yS(\mathbf{0})]\}\rangle\,,$$

where x and y are dummy integration variables introduced by (18). The indicated average can be promoted to the exponent as a cumulant expansion by using the rule [14]

$$\langle e^{\zeta}\rangle = \exp\left(\sum_{n=0}^{\infty}\langle\zeta^n\rangle_c \Big/ n!\right)\,,$$

where $\langle\zeta^n\rangle_c$ represents the nth cumulant average of the random variable ζ: $\langle\zeta\rangle_c = \langle\zeta\rangle$, $\langle\zeta^2\rangle_c = \langle\zeta^2\rangle - \langle\zeta\rangle^2$, and so on. The average over phase constant ϕ in (1) makes $\langle S(\mathbf{r})\rangle = 0$, so only even cumulants contribute to the sum. Furthermore [1], in the limit $N \to \infty$, the second cumulant average (the first nontrivial one) is dominant, assuming that $P(k)$ is normalizable as required in (3). Thus only averages of the type $\langle S(\mathbf{r})S(\mathbf{r}')\rangle$, for $\mathbf{r}' = \mathbf{r}$ and $\mathbf{0}$, will contribute. Therefore, after some rearranging, we eventually derive [1]

$$\begin{aligned}\Gamma_{\alpha,\beta}(r) = \frac{1}{\pi^2}\iint_{-\infty}^{\infty} dx\,dy \frac{1}{xy}\sin(\delta x)\sin(\delta y) \\ \times e^{2i\epsilon(x+y)}e^{-[x^2+y^2+2xy\tau(r)]}\,, \end{aligned}\tag{19}$$

where $\delta = \beta - \alpha$ and $\epsilon = (\beta + \alpha)/2$, and where

$$\begin{aligned}\tau(r) &= 2\langle\cos(\mathbf{k}\cdot\mathbf{r} + \phi)\cos(\phi)\rangle \\ &= \int_0^\infty P(k) j_0(kr)\,dk \end{aligned}\tag{20}$$

is the autocorrelation of the unleveled wave, having the boundary value $\tau(0) = 1$. Because of the normalization of (1), $\Gamma_{\alpha,\beta}(r)$ is independent of the A distribution in (1), as already mentioned. The model also has the property of asymptotic independence [1], i.e.,

$$\lim_{r\to\infty}\Gamma_{\alpha,\beta}(r) = c_0^2\,, \tag{21}$$

corresponding in (12) to the average of the product going over to the product of averages. While unsurprising, the result is not trivial, since it depends on the assertion

$$\lim_{r\to\infty}\tau(r) = 0. \tag{22}$$

This turns out also to be true as consequence of Eq. (3). These limits are proven in Appendix A.

With the discussion following Eq. (14) in mind, we consider now the case of scattering from a generalized interspace, which is equivalent to having a contrast with $\rho_1 = \rho_2$. Thus, ignoring the forward scattering, we rewrite (17) in standardized form as

$$I(Q) = 4\pi V\rho^2 c_0(1 - c_0)\int_0^\infty r^2\gamma(r) j_0(Qr)\,dr\,, \tag{23}$$

where $\gamma(r)$ is the Porod-Debye correlation function, which is defined by the relation

$$\Gamma(r) = c_0^2 + c_0(1 - c_0)\gamma(r)\,. \tag{24}$$

Here, simplifying the earlier notation, $\Gamma(r)$ stands for $\Gamma_{\alpha,\beta}(r)$, as given by the right-hand side of (19), and c_0 is the function given by (8). Note that

$$\gamma(0) = 1\,,\quad \lim_{r\to\infty}\gamma(r) = 0\,. \tag{25}$$

Also in (23), we have set $\rho = \rho_0 - \rho_1$; for our purpose we could also set $\rho = 1$, but one is accustomed to seeing a contrast variable in such expressions.

The representation in Eq. (19) does not seem to reduce, in general, to an elementary formula for the functional $\Gamma[\tau(r)]$, although there is an important exception (rederived below) that by now is more or less well known. As a result, substitution of (19) into Eq. (23) does not lead to an easily manageable expression for the scattering. However, as shown in I, a formally exact and useful representation can be obtained by first replacing $\exp[-2xy\tau(r)]$ in (19) with its Taylor-series expansion and then integrating term by term, which is straightforward since the resulting integrands are separable in the double integration variables x and y and are easily put into a standard form. Taking account of (24), this procedure leads to

$$c_0(1-c_0)\gamma(r) = \sum_{n=1}^{\infty} C_n(\alpha,\beta)\tau^n(r)\,, \tag{26}$$

where

$$C_n(\alpha,\beta) = \frac{1}{\pi n! 2^n}[e^{-\alpha^2}H_{n-1}(\alpha) - e^{-\beta^2}H_{n-1}(\beta)]^2\,, \tag{27}$$

and the $H_n(x)$ are Hermite polynomials. The expansion of $\gamma(r)$ given by (26) can be substituted in Eq. (23), and the scattering function then has the series representation

$$I(Q) = 4\pi V\rho^2 \sum_{n=1}^{\infty} C_n(\alpha,\beta)\int_0^{\infty} r^2\tau^n(r)j_0(Qr)\,dr\,. \tag{28}$$

As discussed in I, the first few terms of the series dominate the scattering at sufficiently small Q. In particular, the first integral produces a peak, proportional to $P(Q)/Q^2$, while, if $P(k)$ is sharp, the second tends to Q^{-1} as $Q \to 0$. However, since

$$C_1(\alpha,\beta) = \frac{1}{2\pi}(e^{-\alpha^2} - e^{-\beta^2})^2\,,$$
$$C_2(\alpha,\beta) = \frac{1}{2\pi}(\alpha e^{-\alpha^2} - \beta e^{-\beta^2})^2\,, \tag{29}$$

these contributions tend to be mutually exclusive in $I(Q)$. For ease of discussion, let us give names to two special morphologies within the model. The case of $\alpha = 0$ and $\beta \to \infty$ corresponds to a single interface (the 0 set) separating two equivolume bulk partitions $c_1 = c_2 = \frac{1}{2}$; call this the standard interface model. With $\alpha = -\beta$, an interspace having finite volume separates two equivolume bulk partitions; designate this as the standard film morphology, although the "film" need not be thin. The standard film has the 0 set in its interior. Then we can note that in the case of the standard interface $C_1(0,\infty) = (2\pi)^{-1}$, $C_2(0,\infty) = 0$; while for the standard film, $C_1(-\beta,\beta) = 0$, $C_2(-\beta,\beta) = 2\pi^{-1}\beta^2\exp(-2\beta^2)$. More generally, one sees from (27) that $C_n(0,\beta) = 0$ for all even n, while $C_n(-\beta,\beta) = 0$ for all odd n. Implications of this "competition" in interpreting contrast variation experiments were analyzed in I. The integrals appearing in Eq. (28) can be done analytically for the prototype family of morphologies $P(k) = \delta(k - k_0)$. These results are given in Appendix B.

The series representation in (28) is less convenient for analyzing scattering at large Q, although computational applications of it certainly are feasible [10]. The technical problem is that the rate of convergence of the series for $\gamma(r)$ in Eq. (26) decreases rapidly as $r \to 0$, where $\tau(r) \to 1$, and it is the small-r region that determines the large-Q behavior of scattering. By contrast, the small-Q scattering depends mainly on the large-r domain, where $\tau(r) \to 0$ and (26) converges rapidly. Asymptotic analysis is the subject of Sec. III, but it is appropriate to say here that the normal linear behavior of $\gamma(r)$ at the origin—which leads to the Porod scattering law—cannot be realized by any finite sequence of terms in (26) since $\tau(r)$ normally is quadratic in r at the origin. This means that numerical computation of large-Q scattering using (26) leads unavoidably to exponential decay at large enough values of Q, although reasonably good Porod law behavior is achievable over a useful Q range when care is taken to reduce rounding errors [10].

An alternative representation of $\gamma(r)$ can be derived from Eq. (19), which simplifies the small-r analysis by avoiding the series expansion and brings out other mathematical aspects of the model. The first step is to eliminate the difficult factor $(xy)^{-1}$ in the integrand of (19). Begin by letting the symbol λ represent the function $\tau(r)$, so that the functional $\Gamma[\tau(r)]$ can be treated as the ordinary function $\Gamma(\lambda)$. Then, with $\Delta(\lambda) = \partial_\lambda\Gamma(\lambda)$, we have

$$\Delta(\lambda) = -\frac{2}{\pi^2}\iint_{-\infty}^{\infty} dx\,dy\sin(\delta x)\sin(\delta y) \times \exp[2i\epsilon(x+y) - (x^2+y^2+2xy\lambda)]\,. \tag{30}$$

This can be expanded to

$$\Delta(\lambda) = \frac{1}{\pi^2}\iint_{-\infty}^{\infty} e^{-(x^2+y^2+2xy\lambda)} \times (e^{2i\beta(x+y)} + e^{2i\alpha(x+y)} - 2e^{2i(\alpha x+\beta y)})\,dx\,dy\,.$$

Next, the rigid rotation transformation

$$x = \frac{1}{\sqrt{2}}(u+v)\,, \quad y = \frac{1}{\sqrt{2}}(-u+v)$$

produces separation in u and v. The integrations that remain are elementary, and the result is

$$\Delta(\lambda)=\frac{1}{2\pi\sqrt{1-\lambda^2}}\left[\exp\left(\frac{-2\alpha^2}{1+\lambda}\right)+\exp\left(\frac{-2\beta^2}{1+\lambda}\right)-2\exp\left(\frac{-(\alpha-\beta)^2}{2(1-\lambda)}\right)\times\exp\left(\frac{-(\alpha+\beta)^2}{2(1+\lambda)}\right)\right]. \tag{31}$$

To retrieve $\Gamma(r)$ we simply recall the definition of $\Delta(\lambda)$, so that we can write

$$\Gamma(r)=c_0^2+\int_0^{\tau(r)}\Delta(\lambda)\,d\lambda\,, \tag{32}$$

where the integration constant c_0^2 gives the correct result for $\tau(r)=0$. After combining (32), (31), and (24), we have the sought after representation

$$c_0(1-c_0)\gamma(r)=\frac{1}{2\pi}\int_0^{(\pi/2)\gamma_0(r)}\left[\exp\left(\frac{-2\alpha^2}{1+\sin\theta}\right)+\exp\left(\frac{-2\beta^2}{1+\sin\theta}\right)-2\exp\left(\frac{-\alpha^2-\beta^2+2\alpha\beta\sin\theta}{\cos^2\theta}\right)\right]d\theta\,. \tag{33}$$

In (33)

$$\gamma_0(r)=\frac{2}{\pi}\arcsin[\tau(r)] \tag{34}$$

is the autocorrelation function for the standard interface morphology, which can be seen by substituting $\alpha=0$ and $\beta=\infty$ into (33) (note that $c_0=\frac{1}{2}$ in this case). Equation (34) was found independently by Hopper [3] and by Berk [1], but an equivalent formula evidently was first produced by Van Vleck and Middleton [15] in a theoretical analysis of clipped noise. It can also be derived from the series representation in Eq. (26). For the standard interface Eq. (27) gives

$$C_{2n+1}(0,\infty)=\frac{H_{2n}^2(0)}{\pi(2n+1)!\,2^{2n+1}},\quad C_{2n}(0,\infty)=0$$

for $n=0,1,\ldots$, where $H_{2n}(0)=(-1)^n2^{2n}(2n-1)!!$. Then using

$$n!!(n-1)!!=n!,\quad (2n)!!=2^n(n!)\,,$$

one gets

$$C_{2n+1}(0,\infty)=\frac{1}{2\pi}\frac{(2n)!^2}{2^{2n}(n!)^2(2n+1)}\,.$$

Substitution of this result into Eq. (26) gives the Taylor series for $(2\pi)^{-1}\arcsin(\lambda)$, and we again have the formula in (34).

In the subsequent discussion we will need the leading behavior of $\gamma(r)$ near $r=0$. Using the results in hand, this may be inferred from

$$c_0(1-c_0)\frac{d\gamma(r)}{dr}=\Delta[\tau(r)]\frac{d\tau(r)}{dr}\,. \tag{35}$$

III. ASYMPTOTIC BEHAVIOR

The asymptotic (large-Q) behavior of scattering from the leveled-wave morphology is determined by the chain of functional relationships described in the preceding section. Thus the large-Q behavior of $I(Q)$ is determined by the small-r behavior of $\gamma(r)$, which depends on the small-r dependence of $\tau(r)$; and this in turn is determined by the large-k behavior of $P(k)$. The asymptotic behavior of $P(k)$, therefore, ultimately fixes the asymptotic law of the scattering. At the links defined by Fourier transformation we may appeal directly to the Riemann-Lebesque lemma, as generalized by Lighthill [16].

The first link in the chain is established by Eq. (20), which we restate here for convenience:

$$\tau(r)=\int_0^\infty P(k)j_0(kr)\,dk\,, \tag{36}$$

where the normalization of $P(k)$ gives $\tau(0)=1$. Assuming also that $P(k)$ falls off faster than a power law as $k\to\infty$ (i.e., that all its moments exist), we may expand the Bessel function under the integral to obtain

$$\tau(r)=1-\frac{1}{6}\langle k^2\rangle r^2+\cdots\,. \tag{37}$$

Then using (37) in Eq. (35), one finds straightforwardly, for $\alpha\neq\beta$, that

$$\gamma(r)\sim 1-\frac{r}{\xi}\,,\quad r\to 0\,, \tag{38}$$

with

$$\xi=\frac{c_0(1-c_0)2\pi\sqrt{3}}{k_{\rm rms}(e^{-\alpha^2}+e^{-\beta^2})}\,, \tag{39}$$

and $k_{\rm rms}=\sqrt{\langle k^2\rangle}$. By identifying ξ in (38) with the Porod correlation length [17], we can combine (39) with Porod's formula $\xi=4c_0(1-c_0)V/\Sigma$, where Σ is the total interfacial area, to obtain

$$\Sigma/V=2(\pi\sqrt{3})^{-1}(e^{-\beta^2}+e^{-\alpha^2})k_{\rm rms}\,. \tag{40}$$

The well-known scattering consequence of the linearity in Eq. (38) is Porod's law, viz. $I(Q)\propto\Sigma/Q^4$, as $Q\to\infty$. Such linearity is characteristic of an interface that is both sharp—in the sense of distinguishing between the two phases in contact with it—and smooth—i.e., differentiable in the "lateral" directions. We see from Eq. (39) that within the leveled-wave scheme this association is tied to a finite value of $k_{\rm rms}$. Since the sharpness of the interface is a matter construction, only its smoothness can be at issue in this model. Heuristically, an infinite value

of $k_{\rm rms}$ in Eq. (40) implies an infinite area-to-volume ratio, suggestive of a fractal surface [18]. In this case Eq. (37) must be modified, as we discuss next.

Let us consider a class of "long-tailed" $P(k)$ that behave asymptotically as

$$P(k) \sim a(\varepsilon) k^{-2\varepsilon-1}, \quad k \to \infty, \tag{41}$$

with $0 < \varepsilon < 1$ (this ε has no connection with the usage in Sec. II). The coefficient $a(\varepsilon)$ is left unspecified for now but must have dimension $L^{-2\varepsilon}$ to make $P(k)$ have the required dimension of L^{-1}. The restriction on ε in (41) allows $P(k)$ to be normalizable over this domain, but guarantees that it has no second or higher moment. In particular, term-by-term integration of the Bessel function expansion in (36) is undefined, so Eq. (37) no longer holds. In this circumstance, the easiest route to the required modification of (37) is to use the inverse of Eq. (36), which is given by

$$P(k) = (2\pi)^3 k \int_0^\infty r\tau(r) \sin(kr)\, dr. \tag{42}$$

With substitutions $r = x$ and $k = 2\pi y$, Eq. (42) assumes a standard form as

$$g(y) = \int_{-\infty}^{\infty} f(x) e^{-i2\pi xy}\, dx, \tag{43}$$

where

$$f(x) = x\tau(x), \quad g(y) = \frac{P(2\pi y)}{8\pi^3 iy}, \tag{44}$$

and where $\tau(x)$ has been extended to the negative-x domain as an even function, which is consistent with its definition in (36).

The Riemann-Lebesque-Lighthill theorem relates the behavior of $g(y)$ as $|y| \to \infty$ to the occurrence of certain removable singularities in $f(x)$ and its derivatives. If $f(x)$ has no singularities, then $g(y)$ decays faster than a power law in $|y|$, i.e., exponentially. Otherwise, the leading singularity of $f(x)$ determines the weakest decay of $g(y)$. To help automate this type of analysis, Lighthill [16] has derived a table that matches asymptotic expressions of $g(y)$ with specific singular forms of $f(x)$. Thus, given the proposition in Eq. (41), we may infer from the theorem that

$$f(x) \sim \mathrm{sgn}(x)[|x| - b(\varepsilon)|x|^{2\varepsilon+1}], \quad x \to 0 \tag{45}$$

where $b(\varepsilon)$ in (45) and $a(\varepsilon)$ in are related by

$$a(\varepsilon) = 8\pi^3 b(\varepsilon) \sin(\pi\varepsilon)(2\varepsilon)!. \tag{46}$$

Then from (45) we have

$$\tau(r) \sim 1 - b(\varepsilon) r^{2\varepsilon}, \quad r \to 0. \tag{47}$$

Using this in Eq. (35) then gives

$$\gamma(r) \sim 1 - B(\varepsilon) r^{\varepsilon}, \quad r \to 0 \tag{48}$$

with

$$B(\varepsilon) = \frac{\sqrt{b(\varepsilon)}\,\chi(\alpha,\beta)}{\sqrt{2}\,\pi}, \tag{49}$$

and where

$$\chi(\alpha,\beta) = \frac{e^{-\alpha^2} + e^{-\beta^2}}{c_0(1-c_0)} \tag{50}$$

is used for a recurring factor. The last link, from $\gamma(r)$ to $I(Q)$, once again is a Fourier transform. To simplify notation, first define $J(Q)$ by way of

$$I(Q) = 4\pi V \rho^2 c_0 (1-c_0) J(Q), \tag{51}$$

where $I(Q)$ is given in Eq. (23), and then define

$$f(x) = x\gamma(x), \quad g(y) = -4\pi i y J(2\pi y). \tag{52}$$

These new definitions of $f(x)$ and $g(y)$ also stand in the relationship given by (43), with

$$f(x) \sim \mathrm{sgn}(x)[|x| - B(\varepsilon)|x|^{\varepsilon+1}], \quad x \to 0 \tag{53}$$

so that using Lighthill's table we arrive at

$$J(Q) \sim A(\varepsilon) Q^{-\varepsilon-3} \quad Q \to \infty \tag{54}$$

where $A(\varepsilon)$ and $B(\varepsilon)$ are related by

$$A(\varepsilon) = B(\varepsilon) \sin\left(\frac{\pi\varepsilon}{2}\right) (\varepsilon+1)!. \tag{55}$$

By comparing (54) with the Bale and Schmidt [19] $6-D$ asymptotic law for scattering from surfaces having fractal dimension D, we may identify our parameter ε with the codimension of a fractal interface,

$$\varepsilon = 3 - D, \quad 2 < D < 3. \tag{56}$$

To complete the chain of aysmptotic dependences that start with (41), we combine (55), (49), and (46), so that in Eq. (54) we have

$$A(\varepsilon) = \frac{\chi(\alpha,\beta)(\varepsilon+1)!}{2\pi^{5/2}\sqrt{(2\varepsilon)!}} \sqrt{a(\varepsilon) \tan\left(\frac{\pi\varepsilon}{2}\right)}. \tag{57}$$

The behavior of the square root in (57) at the Euclidean limits $D \to 2$ and $D \to 3$ requires discussion. First we note that the tangent in (57) diverges as $\varepsilon \to 1$. This follows from the relation between $a(\varepsilon)$ and $b(\varepsilon)$ in (46); if $a(1) \neq 0$, then $b(\varepsilon) \propto (1-\varepsilon)^{-1}$ as $\varepsilon \to 1$ ($D \to 2$). The divergence expresses a discontinuity in the moment expansion of long-tailed functions. The second moment $\langle k^2 \rangle$ does not exist for functions acting as (41) when $\varepsilon < 1$, but $\langle k^2 \rangle$ is finite if $\varepsilon > 1$ is allowed. The way in which leveled-wave fractal surfaces approach $D = 2$ thus depends on the class of functions in which the asymptotic behavior of $P(k)$ is embedded. For ease of discussion, we consider here that $P(k)$ is contained in a set of convex linear combinations represented by

$$P(k) = [1 - \mu(\epsilon)] P_{\rm ST}(k) + \mu(\epsilon) P_{\rm LT}(k), \tag{58}$$

where $\mu(\varepsilon) \in [0,1]$. We treat $P_{\rm ST}(k)$ as a short-tailed function possessing *all* moments, and $P_{\rm LT}(k)$ as a long-tailed function which behaves asymptotically as in (41). We have in mind that $P_{\rm ST}(k)$ contains the wave numbers which, if acting alone ($\mu \equiv 0$), lead to a leveled-wave porous structure with smooth surfaces. The long tail of

$P_{LT}(k)$, weighted by the mixing parameter $\mu(\varepsilon)$, provides the mechanism for fractal roughness, as we have been discussing. We require that both $P_{ST}(k)$ and $P_{LT}(k)$ are normalized so that $P(k)$ is normalized for all functions in this set. For a nonsingular transition to $D = 2$, we require that $\mu(\varepsilon) \to 0$ at least as fast as $1-\varepsilon$ as $\varepsilon \to 1$ and that $\mu(\varepsilon) \equiv 0$ for $\varepsilon \geq 1$. With this restriction, the formula in (57) is modified by replacing $a(\varepsilon)$ with $a(\varepsilon)\mu(\varepsilon)$, which, as $\varepsilon \to 1$, gives

$$A(1) = \frac{\chi(\alpha,\beta)}{2^{3/2}\pi^3}\sqrt{a(1)|\mu'(1)|}\,, \tag{59}$$

where μ' is the derivative of μ. It is not required, however, that $A(1) = A_p$, where

$$A_p = \frac{2}{\xi} = \frac{\chi(\alpha,\beta)k_{\rm rms}}{\pi\sqrt{3}} \tag{60}$$

is the corresponding expression for a normal surface, i.e., the Porod constant for this representation. This approach to $D = 2$ is consistent with the Bale and Schmidt scenario, in which the weight of Q^{-6+D} is finite at $D = 2$. A similar effect could have been achieved, of course, by requiring that $a(\varepsilon) \to 0$ as $\varepsilon \to 1$, but shortly we will absorb $a(\varepsilon)$ into the normalization of $P_{LT}(k)$.

In (57) we also have $A(0) = 0$ if $a(0)$ does not diverge; in other words, the leading asymptotic scattering from the fractal surface has no weight as $D \to 3$. The nature of the asymptotic scattering as $D \to 3$ has been a source of interesting debate [20, 21]. To paraphrase that discussion, let us take the form of (48) as a starting point, which Bale and Schmidt [19] derive using measure-theoretic methods based on the geometric interpretation of the autocorrelation function. The Fourier transform produces (54), as we have seen, with $A(\varepsilon)$ and $B(\varepsilon)$ related by (55). Thus at $D = 3$ the leading power law is Q^{-3}, which coincides with the scattering power law for a $D = 3$ volume fractal. However, its weight is proportional to $3 - D$, since in the Bale and Schmidt derivation $B(\varepsilon)$ is independent of ε. In a comment on Bale and Schmidt [19], Wong and Bray [20] assert that a $D = 3$ fractal surface should be indistinguishable from a $D = 3$ fractal volume in three dimensions. They see the vanishing weight of this power law as problematic, therefore, and provide an alternative measure-theoretic derivation of (48) which, in our terminology, leads to a coefficient $B(\varepsilon)$ that goes as ε^{-1} as $\varepsilon \to 0$. This keeps $A(0)$ finite. In a tandem reply, Pfeifer and Schmidt [21] show with a general argument that the way in which one takes a family of fractal surfaces to $D = 3$ depends on assumptions regarding the treatment of certain Euclidean scales of length, including the size of the scattering system. The path to $D = 3$ implicitly followed by Wong and Bray corresponds [21] to allowing the system to expand to keep pace with the evolution of an increasingly rough surface as $D \to 3$, so that the surface is unable to cover the sample volume. The path taken by Bale and Schmidt corresponds to a fixed volume [21]. Therefore, speaking loosely, as $D \to 3$ a surface tends to fill the bulk, causing the sharp contrast normally provided by the surface to be washed out with a concomitant loss of surface scattering.

Looking at (57), we see that to obtain a result consistent with Wong and Bray requires us to take $a(\varepsilon) \sim \varepsilon^{-1}$ as $\varepsilon \to 0$. However, from (41) this means that $P(k)$ is dominated by an unnormalizable tail as $D \to 3$. If the normalizability of $P(k)$ is discarded, the leveled-wave model breaks down in several places, including at the cumulant expansion that underlies Eq. (19). This pathway to $D = 3$, therefore, seems to be outside the range of useful application of the leveled-wave method. We will see this point made again, below.

In the leveled-wave result for $A(\varepsilon)$, Eq. (57), the behavior as $D \to 3$ is determined by $\sqrt{\varepsilon a(\varepsilon)}$ as $\varepsilon \to 0$. If $a(\varepsilon) \propto \varepsilon$, the weight of the leading fractal scattering falls linearly with $3 - D$, as in the Bale and Schmidt scenario; if $a(\varepsilon)$ tends to a finite constant, the weight falls, ostensibly, as $\sqrt{3 - D}$. In fact, it turns out that a well-behaved approach to $D = 3$ in the leveled-wave scheme requires that $a(\varepsilon) \to 0$ as $\varepsilon \to 0$. To see why, we must investigate the higher-order terms in the asymptotic expansion of $\gamma(r)$.

In order to go beyond the expansion in (48) in the leveled-wave model we must start with the asymptotic behavior of $\tau(r)$. Thus using (58) in (36) we have the corresponding composition for $\tau(r)$,

$$\tau(r) = [1 - \mu(\varepsilon)]\tau_{ST}(r) + \mu(\varepsilon)\tau_{LT}(r)\,. \tag{61}$$

Referring to (37) and (47), we then find the the asymptotic result

$$\tau(r) \sim 1 - \mu b(\varepsilon)r^{2\varepsilon} - \frac{1-\mu}{6}\langle k^2\rangle_{ST} r^2\,, \quad r \to 0\,. \tag{62}$$

This can be used in Eq. (35), say, to derive the asymptotic behavior of $\gamma(r)$, but the expansion is quite involved for general α and β. For the didactic purpose at hand, we can consider the special case of the standard interface, given by Eq. (34). It follows from (33) that near the origin $\gamma(r)$ is continuous in the level parameters, so nothing essential is given up in examining the simpler function. [The last exponential in the integrand of (33) creates terms that vanish exponentially fast as $r \to 0$ and thus cannot give a power law in the asymptotic scattering; moreover, these contributions vanish identically for a single interface, for which either α or β is infinite.] To help organize the expansion, we may write (62) as

$$\tau(r) = 1 - x(r)\,,$$

where

$$x(r) = \mu(\varepsilon)b(\varepsilon)y(r)r^{2\varepsilon}$$

and

$$y(r) = 1 + \frac{[1-\mu(\varepsilon)]\langle k^2\rangle_{ST}}{\mu(\varepsilon)b(\varepsilon)}r^{2(1-\varepsilon)}\,. \tag{63}$$

Since $x(r)$ is small, we use [22]

$$\arcsin(1-x) = \frac{\pi}{2} - \sqrt{2x}\sum_0^\infty \nu_n x^n\,, \tag{64}$$

where

$$\nu_n = \frac{1\times 3\times 5\times\cdots\times(2n-1)}{2^{2n}(2n+1)n!}\,. \tag{65}$$

Then we get

$$\gamma_0(r) \sim 1 - \frac{2^{3/2}}{\pi} \sum_{n=0}^{\infty} \nu_n [\mu(\varepsilon) b(\varepsilon) y(r)]^{n+1/2} r^{(2n+1)\varepsilon} . \tag{66}$$

It is apparent in (66) that expansion of $y(r)$ contributes terms which degenerate to powers of r^2 as $\varepsilon \to 0$, and these will produce asymptotic scattering that falls faster than a power law. Thus the leading asymptotic scattering in this limit can be obtained with $y(r) \equiv 1$ and is independent of $P_{\rm ST}(k)$. Recalling (46) we also see that $b(\varepsilon) \propto a(\varepsilon)/\sin(\pi\varepsilon)$; thus if $a(0) \neq 0$, the expansion in (66) will be ill behaved near $\varepsilon = 0$. Normalization of $P_{\rm LT}(k)$ prevents this, however. To be concrete, assume the simple, normalized form

$$P_{\rm LT}(k) = \frac{2 l_0 \varepsilon}{(1 + k l_0)^{(2\varepsilon+1)}} , \tag{67}$$

where l_0 is a fixed but arbitrary length. For $k l_0 \gg 1$, this goes over to (41) with

$$a(\varepsilon) = 2\varepsilon l_0^{2\varepsilon} . \tag{68}$$

Then it follows that

$$b(\varepsilon) \sim \frac{1}{4\pi^4 l_0^{2\varepsilon}} , \tag{69}$$

as $\varepsilon \to 0$. Thus the normalizability of $P(k)$ is sufficient to ensure a Bale and Schmidt scenario as $D \to 3$. The resulting asymptotic scattering function is

$$J(Q) \sim \frac{2^{3/2}}{\pi} l_0^3 \sum_{n=0}^{\infty} A_n(\varepsilon) (Q l_0)^{-3-(2n+1)\varepsilon} \tag{70}$$

for $Q l_0 \to \infty$, where

$$A_n(\varepsilon) = \nu_n \left(\frac{\mu(\varepsilon)}{4\pi^4} \right)^{n+1/2} \sin\left(\frac{\pi}{2} (2n+1)\varepsilon \right) . \tag{71}$$

A rigorous, if somewhat weak, upper limit can be derived for (70). First let

$$Q l_0 = z^{-1} .$$

Then since

$$\sin\left(\frac{\pi}{2} (2n+1)\varepsilon \right) < \frac{\pi}{2} (2n+1)\varepsilon$$

and

$$\mu(\varepsilon)/(4\pi^4) < 1 ,$$

we have

$$J(Q) < \sqrt{2}\, l_0^3 G(z) ,$$

where

$$G(z) = \varepsilon \sum_{0}^{\infty} (2n+1) \nu_n z^{3+(2n+1)\varepsilon} .$$

Now using (64), one can show

$$G(z) = \frac{z^4}{\sqrt{2}} \frac{d}{dz} \left(\frac{\pi}{2} - \arcsin(1 - z^{2\varepsilon}) \right) ,$$

which easily leads to

$$J(Q) < \frac{\sqrt{2}\, l_0^3 \varepsilon}{(Q l_0)^3 \sqrt{(Q l_0)^{2\varepsilon} + \frac{1}{2}}} \tag{72}$$

for $Q l_0 \to \infty$ and $\varepsilon \to 0$. Thus at $D = 3$ the right-hand side of (72) vanishes as $O(\varepsilon)$, and the scattering from the leveled-wave morphology falls faster than algebraically as $Q \to \infty$. Near $D = 3$ the large-Q scattering falls algebraically, but not as a well-isolated power law.

Within the Bale and Schmidt scenario, Rojanski *et al.* [23] have put forward the thesis that the asymptotic scattering from porous materials with fractal surfaces should behave as

$$J(Q) \sim A_{\rm fr}(\varepsilon) Q^{-\varepsilon-3} + A_d(\varepsilon) Q^{-4} , \tag{73}$$

where $A_{\rm fr}(\varepsilon)$ has the form of (55), and $A_d(0) \neq 0$. Thus, as $D \to 3$ the first term vanishes—the Bale and Schmidt scenario—and a Porod-like law, which had been hidden by the less-steep Bale and Schmidt law, emerges as the asymptotic scattering. Working backwards, this scattering implies

$$\gamma(r) \sim 1 - B_{\rm fr}(\varepsilon) r^{\varepsilon} - B_d(\varepsilon) r , \quad r \to 0 \tag{74}$$

where the term linear in r is responsible for the Q^{-4} scattering, as usual. The significant feature of (74) is the isolation of the r^{ε} and r^1 powers as $\varepsilon \to 0$, which leads to a corresponding separation of the large-Q scattering into distinct power-law components. In Rojanski *et al.* such behavior follows *a fortiori* from a product ansatz

$$\gamma(r) = \gamma_{\rm fr}(r) \gamma_d(r) , \tag{75}$$

with

$$\gamma_{\rm fr}(r) \sim 1 - B_{\rm fr}(\varepsilon) r^{\varepsilon} , \quad \gamma_d(r) \sim 1 - B_d(\varepsilon) r ,$$

as $r \to 0$, which is intended to separate pore-surface from pore-size effects.

The leveled-wave model does not fit this picture, as we have seen. In (66) a term linear in r can occur only for rational values of ε satisfying

$$\varepsilon(N, M) = \frac{1 - 2M}{1 - 2(M - N)} ,$$

for integers $N, M \geq 0$. If $N, M \geq 1$, then $\varepsilon(N, M) > 1$, which is outside the allowed domain of ε. Only values of ε from the set $(1 + 2N)^{-1}$, corresponding to $M = 0$ [$y(r) \equiv 1$], produce a term linear in r in the expansion of $\gamma_0(r)$ and a concomitant Q^{-4} in the scattering. These essentially accidental terms are unconnected with $P_{\rm ST}(k)$, are never the leading asymptotic contribution when $N \geq 1$, and are never isolated from nearby powers when ε is small ($N \gg 1$). Indeed, as we proved in (72), they are asymptotically insignificant near $D = 3$.

APPENDIX A

It is useful to see that the integral representation of $\Gamma_{\alpha,\beta}(r)$ in Eq. (19) has the expected limiting values, consistent with the definitions in Eqs. (12) and (8). First we consider $r = 0$. Because $\tau(0) = 1$, Eq. (19) gives the starting point

$$\Gamma_{\alpha,\beta}(0)=\frac{1}{\pi^2}\iint_{-\infty}^{\infty}dx'dy'\frac{1}{x'y'}\sin(\delta x')\sin(\delta y')\times e^{2i\epsilon(x'+y')-(x'^2+y'^2)}. \tag{A1}$$

Making the coordinate transformation

$$x=\tfrac{1}{2}(x'+y'),\quad y=\tfrac{1}{2}(-x'+y')$$

produces

$$\Gamma_{\alpha,\beta}(0)=\frac{1}{\pi^2}\int_{-\infty}^{\infty}dxe^{2i\epsilon x-x^2}\int_{-\infty}^{\infty}dy\frac{1}{x^2-y^2}\cos(\delta y), \tag{A2}$$

where the y integral evaluates to $\pi\sin(\delta x)/x$. Thus

$$\Gamma_{\alpha,\beta}(0)=\frac{1}{\pi}\int_{-\infty}^{\infty}dx\frac{1}{x}e^{2i\epsilon x-x^2}\sin(\delta x). \tag{A3}$$

This integral is easily reduced to a standard form, and leads to the result in Eq. (8). Next we consider the asymptotic limit. With the assumption $\lim_{r\to\infty}\tau(r)=0$, the representation in (19) gives

$$\Gamma_{\alpha,\beta}(\infty)=\frac{1}{\pi^2}\iint_{-\infty}^{\infty}dx\,dy\frac{1}{xy}\sin(\delta x)\sin(\delta y)\times e^{2i\epsilon(x+y)-x^2-y^2}. \tag{A4}$$

Noting the separation of x and y, and checking (A3), we see at once that

$$\Gamma_{\alpha,\beta}(\infty)=[\Gamma_{\alpha,\beta}(0)]^2. \tag{A5}$$

In fact $\Gamma_{\alpha,\beta}(r)=c_0^2$ wherever $\tau(r)=0$.

The final matter is to verify the asymptotic behavior of $\tau(r)$. Following the notational convention used in Sec. III, Eq. (20) can be rewritten as

$$\tau(2\pi y)=\frac{g(y)}{4\pi y},$$

where $g(y)$ is defined as in (43) with

$$f(x)=\mathrm{sgn}(x)\frac{P(|x|)}{|x|}.$$

Thus $\lim_{r\to\infty}\tau(r)=0$ requires that $g(y)<O(y)$ as $y\to\infty$. According to the Riemann-Lebesque-Lighthill theorem, discussed in Sec. III, we must then have $P(|x|)<O(|x|^{-1})$ as $|x|\to 0$; in other words, $P(k)<O(k^{-1})$ as $k\to 0$. But this is also a requirement of the normalizability of $P(k)$, as expressed in Eq. (3), thus confirming the assertion.

APPENDIX B

The integrals in Eq. (28) can be reduced to formulas in the special case of the prototype leveled-wave morphologies, i.e., the class for which $P(k)=\delta(k-k_0)$. First let

$$J^{(n)}(Q)=4\pi\int_0^{\infty}\tau^n(r)j_0(Qr)\,r^2dr=\int e^{-i\mathbf{Q}\cdot\mathbf{r}}\tau^n(\mathbf{r})\,d^3r. \tag{B1}$$

Also introduce the isotropic function of wave vector $\mathbf{k}$,

$$\mathcal{P}(\mathbf{k})=2\pi^2\frac{P(k)}{k^2}, \tag{B2}$$

in terms of which the definition of $\tau(r)$, Eq. (20), can be expressed as

$$\tau(r)=\frac{1}{(2\pi)^3}\int e^{i\mathbf{k}\cdot\mathbf{r}}\mathcal{P}(\mathbf{k})\,d^3k, \tag{B3}$$

which maintains $\tau(0)=1$. Then we have

$$J^{(n)}(Q)=(2\pi)^3\frac{1}{(2\pi)^3}\cdots\frac{1}{(2\pi)^3}\times\int\cdots\int\delta(\mathbf{k}_1+\cdots+\mathbf{k}_n-\mathbf{Q})\times\mathcal{P}(\mathbf{k}_1)\cdots\mathcal{P}(\mathbf{k}_n)\,d^3k\cdots d^3k. \tag{B4}$$

Now $\mathcal{P}(\mathbf{k})/(2\pi)^3$ is the probability density of the random wave *vector* $\mathbf{k}$—recall that $P(k)$ is the density of wave *number* k—which is isotropic in the present context. Thus, according to (B4), $J^{(n)}(Q)/(2\pi)^3$ is the (isotropic) probability density of the random sum of wave vectors $\mathbf{k}_1+\cdots+\mathbf{k}_n$. The scattering from the leveled-wave morphology, as given by the series representation in (28), is thus interpretable as a sum of probabilities that the scattering wave vector $\mathbf{Q}$ is the resultant of exactly n randomly chosen wave vectors, with weights $C_n(\alpha,\beta)$ depending on the level sets associated with interfaces. For $n=1$, we have at once that

$$J^{(1)}(Q)=\mathcal{P}(Q), \tag{B5}$$

which corresponds to scattering from a continuous density function proportional to the unleveled wave (1); it is simply the distribution of wave vectors in the unleveled wave. The $n\geq 2$ contributions stem from the leveling operation that defines the density discontinuities (i.e., the sharp interfaces) in the model. For $n=2$, the integrations in (B4) can be manipulated to give

$$J^{(2)}(Q)=\frac{\pi^2}{Q}\iint_{-\infty}^{\infty}dk_1dk_2\frac{1}{k_1k_2}\Lambda(Q;k_1,k_2)\times P(k_1)P(k_2), \tag{B6}$$

where $\Lambda(Q;k_1,k_2)=\Lambda(Q;k_2,k_1)=1$ for $|Q-k_1|\leq k_2<Q+k_1$, and 0 otherwise. Corresponding formulas for $n\geq 3$ are cumbersome and computationally impractical. For given $P(k)$, it is easier to obtain $\tau(r)$ from (20) and compute the functions $J^{(n)}(Q)$ from (B1). Indeed, for some interesting choices of $P(k)$, $\tau(r)$ can be reduced to a formula [1, 8, 11].

For the prototype morphologies, all the $J^{(n)}(Q)$ can be worked out. Thus, with (4),

$$\tau(r)=j_0(k_0r), \tag{B7}$$

and using (B1), we arrive at [24]

$$J^{(n)}(Q) = \frac{4\pi^2}{2^n(n-2)!k_0^2 Q} \sum_{m=0}^{[(1/2)(n-Q/k_0)]} (-1)^m \binom{n}{m} (n - 2m - Q/k_0)^{n-2} \tag{B8}$$

for $n \geq 2$ and $Q \leq nk_0$. For $Q > nk_0$, $J^{(n)}(Q) = 0$. In the upper limit of the summation, the symbol $[x]$ is the largest integer in x. For $n = 1$, we use (B5)

$$J^{(1)}(Q) = \frac{2\pi^2}{k_0^2}\delta(Q - k_0)\,. \tag{B9}$$

Thus for $n \geq 2$, $J^{(n)}(Q)$ is a piecewise continuous function supported on the interval $[0, nk_0]$. This property follows easily from (B4), since when $|\mathbf{k}_n| = k_0$, the resultant Q cannot exceed nk_0. From this point of view it is again easy to see why a truncated series in (28) cannot achieve the Porod law—or any power-law behavior—as $Q \to \infty$. For $n > 2$ the prefactor Q^{-1} in (B8) is canceled by the polynomial multiplying it for $Q < k_0$, so only $J^{(2)}(Q)$ is singular at the origin. Examples may help clarify the formula in (B8):

$$J^{(2)}(Q) = \frac{\pi^2}{k_0^2} \begin{cases} Q^{-1}, & Q \leq 2k_0 \\ 0, & 2k_0 < Q \end{cases} \tag{B10}$$

$$J^{(3)}(Q) = \frac{\pi^2}{2k_0^3} \begin{cases} 2, & Q \leq k_0 \\ 3k_0 Q^{-1} - 1, & k_0 < Q \leq 3k_0 \\ 0, & 3k_0 < Q \end{cases} \tag{B11}$$

$$J^{(4)}(Q) = \frac{\pi^2}{8k_0^4} \begin{cases} 8k_0 - 3Q, & Q \leq 2k_0 \\ Q^{-1}(4k_0 - Q)^2, & 2k_0 < Q \leq 4k_0 \\ 0, & 4k_0 < Q \end{cases} \tag{B12}$$

and so on.

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