Photographic granularity: mathematical formulation and effect of impenetrability of grains

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We develop a new and general formula for the granularity of a wide class of anisotropic media (for apertures of arbitrary shape) that exactly accounts for the microstructure. Using this formula, we compute for the first time to our knowledge, the granularity of a two-dimensional distribution of impenetrable, opaque circular disks for a wide range of densities and aperture sizes. These results are compared with the much-investigated, random-dot model. The effect of impenetrability of the grains for a fixed aperture area is to reduce the granularity relative to that of the random-dot model, especially at high densities.

1. INTRODUCTION

Considerable effort has been put forth in describing and measuring the granularity of photographic materials.[1–11] Granularity is a measure of the transmittance fluctuations in a scanning aperture. For photographic emulsions, many theoretical studies of granularity have been based on the so-called random-dot model, which neglects spatial correlations between the grains in the emulsion layer; i.e., the grains are able to overlap.[1,4–6,8,10] But in fact a real emulsion is composed of impenetrable grains. [The effect of the impenetrability of the grains in the emulsion will become stronger when the layer becomes thinner (relative to the dimension of the grain) and the density becomes higher.] Bayer[6] has shown that the random-dot model predicts a rapid increase in the granularity at high densities and noted that measurements on developed images have failed to show any effect as great as that in the random-dot model. Consequently, the random-dot model may be misleading for theoretical investigations of granularity. Attempts have been made to take into account the effect of impenetrability of the grains,[7,9,11] but these treatments either have been approximate (in that they do not directly account for the microstructure)[7,9] or have considered special, limiting cases.[11]

The problem of predicting fluctuations in transmittance is also of importance in electrophotography,[12] in which black pigmented particles distributed on paper absorb light to form the image that one sees. The effect of impenetrability of the particles on the granularity is expected to be particularly important in this application.

In this paper we present a general analysis to predict the granularity of anisotropic heterogeneous media, using concepts and results of random-media theory. Random-media theory is a broad field that attempts to relate bulk properties of disordered heterogeneous media to the details of the microstructure.[13] We shall begin by considering two-dimensional models consisting of distributions of opaque particles of arbitrary shape and size and of variable penetrability. A two-dimensional distribution of particles is a reasonable first approximation of thin films. Using our general formula, we compute, for the first time known to us, the granularity of a distribution of impenetrable disks for arbitrary densities and aperture sizes.

This paper should be regarded as the first step in a systematic study of the granularity of real materials. Effects of film thickness (i.e., three-dimensional effects), size distribution of the grains, etc. must also be taken into account in the same rigorous manner. We intend to study such effects in future papers.

In Section 2, we present some basic definitions and equations. In Section 3, we derive a general expression for the granularity of arbitrary, statistically anisotropic media and arbitrary-shaped apertures in terms of the microstructure, using a new approach. We then review recent advances made in the quantitative characterization of the microstructure of random media and give an explicit relation for the granularity of a broad class of anisotropic media. In Section 4, we compute the granularity for distributions of impenetrable disks and compare our results with the well-known, random-dot model. Finally, we make concluding remarks in Section 5.

2. DEFINITIONS AND BASIC RELATIONS

We shall generally model the photographic emulsion or electrophotograph as a two-dimensional array of N arbitrary-shaped opaque bodies distributed throughout an otherwise transparent surface. In the language of random-media theory,[13] the random medium is a domain of space $\mathcal{A}(\omega) \in \mathcal{R}^2$ (where the realization ω is taken from some probability space Ω) of area A, which is composed of two regions: the transparent region $\mathcal{A}_1$ of area fraction ϕ_1 and the opaque grain region $\mathcal{A}_2$ of area fraction ϕ_2.

A. Point Transmittance

The characteristic function $I(\mathbf{x})$ of the transparent region is defined by

$$I(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \in \mathcal{A}_1 \\ 0, & \mathbf{x} \in \mathcal{A}_2 \end{cases}. \tag{2.1}$$

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Now the point transmittance at position $\mathbf{x}$, $t(\mathbf{x})$ is simply equal to the characteristic function, i.e.,

$$t(\mathbf{x}) = I(\mathbf{x}). \tag{2.2}$$

It is useful to introduce the n-point probability function S_n (Ref. 13), which is defined according to the relation

$$S_n(\mathbf{x}_1, \ldots, \mathbf{x}_n) = \left\langle \prod_{i=1}^{n} I(\mathbf{x}_i) \right\rangle, \tag{2.3}$$

in which angle brackets denote an ensemble average. S_n gives the probability of simultaneously finding n points at positions $\mathbf{x}_1, \ldots, \mathbf{x}_n$ all in the transparent region $\mathcal{A}_1$. If the medium is statistically homogeneous, then the n-point probability function is translationally invariant and hence just depends on the relative displacements, i.e., $S_n = S_n(\mathbf{x}_{12}, \ldots, \mathbf{x}_{1n})$, where $\mathbf{x}_{1i} = \mathbf{x}_i - \mathbf{x}_1$. Invoking an ergodic hypothesis for statistically homogeneous media, one can equate the area (or volume) average $\bar{f}$ of some function f,

$$\bar{f} = \lim_{A \to \infty} \frac{1}{A} \int_A f \mathrm{d}A, \tag{2.4}$$

to the ensemble average $\langle f \rangle$. If the medium is also statistically isotropic, then S_n depends on the relative distances $x_{12}, \ldots, x_{1n}$, where $x_{1i} = |\mathbf{x}_i - \mathbf{x}_1|$. For simplicity, we shall henceforth consider statistically anisotropic but homogeneous media.

Let us consider the first two n-point probability functions in more detail. S_1, the probability of finding a point at $\mathbf{x}_1$ in region $\mathcal{A}_1$, is just the expected value of I or t, i.e.,

$$S_1(\mathbf{x}_1) = \langle I(\mathbf{x}_1) \rangle = \langle t(\mathbf{x}_1) \rangle. \tag{2.5}$$

For statistically homogeneous media, S_1 is independent of $\mathbf{x}_1$, and we simply have

$$S_1 = \phi_1 = \langle t \rangle. \tag{2.6}$$

The two-point probability function

$$S_2(\mathbf{x}_{12}) = \langle I(\mathbf{x}_1) I(\mathbf{x}_2) \rangle \tag{2.7}$$

is just the autocorrelation function of I. It contains considerably more information than S_1, such as the average interparticle spacing and average coordination number.[14–19] When $x_2 \to x_1$, then it is clear that

$$S_2(\mathbf{x}_{12}) \to \phi_1. \tag{2.8}$$

When the relative distance between the points becomes very large, then

$$S_2(\mathbf{x}_{12}) \to \phi_1^2, \tag{2.9}$$

assuming no long-range correlations.

The fluctuations associated with the transmittance t can be measured by the variance σ_t^2 given by

$$\begin{aligned} \sigma_t^2 &\equiv \langle t^2 \rangle - \langle t \rangle^2 \\ &= \phi_1 - \phi_1^2 = \phi_1 \phi_2, \end{aligned} \tag{2.10}$$

where σ_t is just the standard deviation.

B. Aperture Transmittance and Granularity

Consider $\tau(\mathbf{x})$ the transmittance through an arbitrary-shaped aperture of area A_a centered at $\mathbf{x}$ with some fixed orientation:

$$\begin{aligned} \tau(\mathbf{x}) &\equiv \frac{1}{A_a} \int t(\mathbf{z}) \theta(\mathbf{z} - \mathbf{x}) \mathrm{d}\mathbf{z} \\ &= \frac{1}{A_a} \int I(\mathbf{z}) \theta(\mathbf{z} - \mathbf{x}) \mathrm{d}\mathbf{z}, \end{aligned} \tag{2.11}$$

where

$$\theta(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \in D_a \\ 0, & \text{otherwise} \end{cases} \tag{2.12}$$

is the aperture-indicator function and D_a is the aperture region. Note that aperture transmittance $\tau(\mathbf{x})$ is, in general, a random variable. Observe furthermore that as $A_a \to \infty$,

$$\tau(\mathbf{x}) \to \langle t \rangle = \phi_1. \tag{2.13}$$

Also, in the limit of a very small aperture ($A_a \to 0$), $\tau(\mathbf{x})$ becomes the point transmittance, i.e.,

$$\tau(\mathbf{x}) \to t(\mathbf{x}) = I(\mathbf{x}). \tag{2.14}$$

The expected value of τ is easily shown to be given by

$$\langle \tau \rangle = \langle t \rangle = \phi_1. \tag{2.15}$$

The granularity G, a measure of the fluctuations in the aperture transmittance, is defined here to be

$$G = \frac{\sigma_\tau}{\langle \tau \rangle} = \frac{\sigma_\tau}{\phi_1}, \tag{2.16}$$

where

$$\sigma_\tau^2 = \langle \tau^2 \rangle - \langle \tau \rangle^2 = \langle \tau^2 \rangle - \phi_1^2 \tag{2.17}$$

is the variance associated with the aperture transmittance. From relations (2.13) and (2.14), we determine that the granularity for infinitesimally small and infinitely large apertures is given, respectively, by

$$G = \frac{\sigma_t}{\phi_1} = \frac{\sqrt{\phi_1 \phi_2}}{\phi_1} \tag{2.18}$$

and

$$G = 0. \tag{2.19}$$

The dependence of the granularity on the aperture area A_a is, in general, nontrivial because it depends on the details of the microstructure of the random medium. A new derivation of this relationship is described in Section 3.

3. MATHEMATICAL FORMULATION OF THE GRANULARITY FOR GENERAL CASES

We shall derive a general expression for the granularity for arbitrary, statistically anisotropic microstructures and arbitrary-shaped apertures in terms of the two-point probability function $S_2(\mathbf{r})$. Although the general formula that we derive is obtained by using a new approach (namely, random-media theory), an equivalent expression in a different form was given by O'Neill.[5] Using the explicit series representations of S_2 obtained by Torquato and Stell[13] for a broad class of anisotropic media, we then explicitly give the granularity relation for such heterogeneous materials.

A. General Granularity Expression

Consider the variance expression (2.17) for an arbitrary-shaped aperture with some fixed orientation. Substitution of Eqs. (2.11) and (2.15) into Eq. (2.17) yields

$$\sigma_\tau^2 = \frac{1}{A_a^2}\left\langle \int I(\mathbf{z})\theta(\mathbf{z}-\mathbf{x})\mathrm{d}\mathbf{z} \int I(\mathbf{y})\theta(\mathbf{y}-\mathbf{x})\mathrm{d}\mathbf{y}\right\rangle - \phi_1^2. \quad (3.1)$$

Since the ensemble average operator and the area integral operators commute, Eq. (2.1) can be rewritten as

$$\sigma_\tau^2 = \frac{1}{A_a^2}\int\int \mathrm{d}\mathbf{y}\mathrm{d}\mathbf{z}\theta(\mathbf{y}-\mathbf{x})\theta(\mathbf{z}-\mathbf{x})S_2(\mathbf{z}-\mathbf{y}) - \phi_1^2, \quad (3.2)$$

where $S_2(\mathbf{z}-\mathbf{y}) = \langle I(\mathbf{y})I(\mathbf{z})\rangle$ is the two-point probability function as defined by Eq. (2.7). Note that since we are treating statistically homogeneous media, $S_2(\mathbf{z}-\mathbf{y}) = S_2(\mathbf{y}-\mathbf{z})$. Finally, we observe that because Eq. (3.1) is independent of position, we can integrate over $\mathbf{x}$ instead of $\mathbf{y}$ and find the equivalent expression

$$\sigma_\tau^2 = \frac{1}{A_a^2}\int S_2(\mathbf{r})A_a^{int}(\mathbf{r})\mathrm{d}\mathbf{r} - \phi_1^2, \quad (3.3)$$

where

$$A_a^{int}(\mathbf{r}) = \int \theta(\mathbf{y}-\mathbf{x})\theta(\mathbf{z}-\mathbf{x})\mathrm{d}\mathbf{x} \quad (3.4)$$

is the intersection area of two aperture regions in which the centroids are separated by the displacement $\mathbf{r} = \mathbf{z} - \mathbf{y}$. Dividing Eq. (3.3) by A_a^2 and integrating over $\mathbf{r}$ gives

$$\frac{1}{A_a^2}\int A_a^{int}(\mathbf{r})\mathrm{d}\mathbf{r} = 1. \quad (3.5)$$

Substitution of Eqs. (3.3) and (3.5) into Eq. (2.16) yields

$$G = \frac{1}{\phi_1 A_a}\left\{\int [S_2(\mathbf{r}) - \phi_1^2]A_a^{int}(\mathbf{r})\mathrm{d}\mathbf{r}\right\}^{1/2}. \quad (3.6)$$

This is the desired granularity expression, which is valid for statistically anisotropic media and given in terms of the two-point probability function $S_2(\mathbf{r})$ (the evaluation of which we discuss below) and the intersection area $A_a^{int}(\mathbf{r})$.

Note that Eq. (3.6) involves $S_2(\mathbf{r})$ minus its long-range value of ϕ_1^2 [compare expression (2.9)]. Hence $[S_2(\mathbf{r}) - \phi_1^2]$ generally oscillates about zero for small r and decays to zero for large r. We refer to the range over which $[S_2(\mathbf{r}) - \phi_1^2]$ is nonnegligibly small as the correlation length, l. If the characteristic size of the aperture is much larger than l, then $A_a^{int}(\mathbf{r})$ is approximately equal to $A_a^{int}(0) = A_a$. Thus Eq. (3.6), in such instances, yields

$$G = \frac{1}{\phi_1 A_a^{1/2}}\left\{\int_{r<l} [S_2(\mathbf{r}) - \phi_1^2]\mathrm{d}\mathbf{r}\right\}^{1/2}. \quad (3.7)$$

Since the integral above is a constant, then for such large apertures

$$G = KA_a^{-1/2}, \quad (3.8)$$

where

$$K = \frac{1}{\phi_1}\left\{\int_{r<l} [S_2(\mathbf{r}) - \phi_1^2]\mathrm{d}\mathbf{r}\right\}^{1/2} \quad (3.9)$$

is a constant (having dimensions of length) that depends on ϕ_1 or, equivalently, on the particle area fraction ϕ_2. Equation (3.9) is reminiscent of the compressibility equation of liquid-state theory,[20] which relates the D-dimensional volume integral over the total correlation function to density fluctuations in the system. $K\sqrt{\phi_1}$ gives a measure of the nonuniformity of coverage of the transparent (void) region. Equation (3.8) is in agreement with experimental observations[5] and, in the limit $A_a \to \infty$, agrees with Eq. (2.19).

For a circular aperture of diameter σ_a (see Fig. 1), the intersection area is given by

$$A_{int}(r) = \frac{\sigma_a^2}{2}\left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]H(\sigma_a - r), \quad (3.10)$$

where $H(x)$ is the Heaviside step function. For a rectangular aperture having sides of length a and b (as indicated in Fig. 1), the intersection area is given by

$$A_{int}(x, y) = (a-x)(b-y)H(a-x)H(b-y). \quad (3.11)$$

Here x and y are the distances between the centroids of two rectangular aperture regions in the x_1 and x_2 directions, respectively.

B. Equations for the n-Point Probability Functions

In order to apply Eq. (3.6), one must know the two-point probability function S_2 for the particular microstructure of interest. Until recently, a means of representing and computing the n-point probability function for realistic model microstructures had been lacking. Torquato and Stell[13] have provided such a formalism for distributions of D-dimensional spheres and, as a result, S_2 has been computed for such models.[14–18] The formal generalization of these results

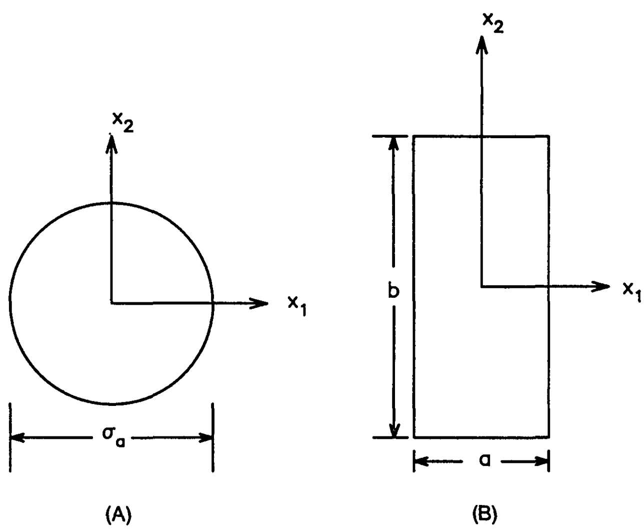

Fig. 1. (A) Circular aperture of diameter σ_a; (B) Rectangular aperture with sides of length a and b.

to anisotropic media with identical particles is actually trivial.[19] We shall briefly review some of these results.

Consider a distribution of N identical particles whose positions are completely specified by center-of-mass coordinates $\mathbf{r}^N \equiv \{\mathbf{r}_1, \ldots, \mathbf{r}_N\}$. In two dimensions this specification includes circular disks, oriented rectangles, oriented ellipses, etc. The particles are distributed over an area A according to the N-particle probability density $P_N(\mathbf{r}^N)$, which characterizes the probability of finding the particles labeled $1, \ldots, N$ with a particular configuration $\mathbf{r}^N$, respectively. P_N normalizes to unity, i.e.,

$$\int P(\mathbf{r}^N)\mathrm{d}\mathbf{r}^N = 1, \tag{3.12}$$

where $\mathrm{d}\mathbf{r}^N \equiv \mathrm{d}\mathbf{r}_1 \ldots \mathrm{d}\mathbf{r}_N$. The function P_N depends on the interparticle interactions (impenetrability effects, Coulombic interactions, etc.) and, in principle, is given for the ensemble under consideration. The ensemble average of any many-body function $f(\mathbf{r}^N)$ is given by

$$\langle f(\mathbf{r}^N)\rangle = \int f(\mathbf{r}^N)P_N(\mathbf{r}^N)\mathrm{d}\mathbf{r}^N. \tag{3.13}$$

The reduced n-particle probability density $\rho_n(n < N)$ is defined by

$$\rho_n(\mathbf{r}^n) = \frac{N!}{(N-n)!}\int P_N(\mathbf{r}^N)\mathrm{d}\mathbf{r}_{n+1}\ldots \mathrm{d}\mathbf{r}_N. \tag{3.14}$$

The quantity $\rho_n(\mathbf{r}^n)$ characterizes the probability of simultaneously finding the center of a particle in the area element $\mathrm{d}\mathbf{r}_1$ about $\mathbf{r}_1$, the center of another particle in area element $\mathrm{d}\mathbf{r}_2$ about $\mathbf{r}_2$, etc. If the medium is statistically homogeneous, the $\rho_n(\mathbf{r}^n)$ depend on the relative displacements $\mathbf{r}_2 - \mathbf{r}_1, \ldots, \mathbf{r}_n - \mathbf{r}_1$. In such instances, it is implied that the thermodynamic limit has been taken; i.e., $N \to \infty$, $A \to \infty$ such that the number density $\rho = N/A = \rho_1(\mathbf{r}_1)$ is some finite constant, and it is convenient to introduce the n-body distribution function g_n defined by

$$g_n(\mathbf{r}^n) = \frac{\rho_n(\mathbf{r}^n)}{\rho^n}. \tag{3.15}$$

When the relative distances between each of the n particles is large, the n-body distribution function has the simple property

$$g_n(\mathbf{r}^n) \to 1, \tag{3.16}$$

assuming no long-range correlations.

It has been shown[13,19] that for such a distribution of particles the S_n are related to the g_n according to the following relation:

$$S_n(\mathbf{x}^n) = 1 + \sum_{k=1}^{\infty} \frac{(-1)^k \rho^k}{k!}\int \cdots \int g_k(\mathbf{r}^k) \times \prod_{j=1}^{k}\left\{1 - \prod_{i=1}^{n}[1 - m(\mathbf{x}_i - \mathbf{r}_j)]\right\}\mathrm{d}\mathbf{r}_j, \tag{3.17}$$

where

$$m(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \in D_p \\ 0, & \text{otherwise} \end{cases} \tag{3.18}$$

is the particle indicator function and D_p denotes a particle region. Given the g_n, one can in principle compute the S_n for any n.

Fully Penetrable Particles

The fully penetrable particle model employed in random-media theory is precisely the same as the so-called random-dot model used in photographic science; i.e., this model assumes that the particle centers are Poisson distributed, and hence spatial correlations between particles are completely neglected. For fully penetrable particles, the g_n are trivial:

$$g_n(\mathbf{r}^n) = 1 \qquad \text{for all } \mathbf{r}^n. \tag{3.19}$$

In such instances, Eq. (3.17) is formally easy to evaluate. For two-dimensional systems, it is found that

$$S_n(x^n) = \exp[-\rho A_n(\mathbf{x}^n)], \tag{3.20}$$

where $A_n(\mathbf{x}^n)$ represents the union area of n particle areas centered at $\mathbf{x}^n$.

In the special case of equisized circular disks of diameter σ, for example,

$$m(x) = \begin{cases} 1, & x < \sigma \\ 0, & \text{otherwise} \end{cases}, \tag{3.21}$$

and the first two union areas are, respectively,

$$A_1 = \frac{\pi\sigma^2}{4} \tag{3.22}$$

and

$$A_2(r) = \frac{\pi\sigma^2}{2} - \frac{\sigma^2}{2}\left[\cos^{-1}\frac{r}{\sigma} - \frac{r}{\sigma}\left(1 - \frac{r^2}{\sigma^2}\right)^{1/2}\right]H(\sigma - r). \tag{3.23}$$

Lower-order S_n were evaluated for fully penetrable spheres,[14] circular disks,[17,18] and oriented circular cylinders of finite aspect ratio.[19]

Totally Impenetrable Particles

For totally impenetrable particles, Torquato and Stell[13] showed that the infinite series [Eq. (3.17)] exactly truncates after the nth term in the sum. For instance, for $n = 1, 2$, we have

$$S_1 = 1 - \rho A_1, \tag{3.24}$$

$$S_2(\mathbf{x}_{12}) = 1 - \rho A_2(\mathbf{x}_{12}) + \rho^2 \int\int g_2(\mathbf{r}_{12})m(\mathbf{x}_1 - \mathbf{r}_1)m(\mathbf{x}_2 - \mathbf{r}_2)\mathrm{d}\mathbf{r}_1\mathrm{d}\mathbf{r}_2. \tag{3.25}$$

Note that there are two-body terms in Eq. (3.17) (second term in the sum) that are identically zero for $n = 2$. Comparison of Eq. (3.20) for $n = 1$ with Eq. (3.24) reveals that the void-volume fraction $\phi_1 = S_1$ for the fully penetrable particle model is always greater than ϕ_1 for the totally impenetrable particle model at the same number density. This is true since there is no interparticle overlap permitted in the former model. Lower-order S_n have been computed for impenetrable equisized spheres[15] and disks.[16]

In Fig. 2, we plot the two-point probability function $S_2(r)$ for an isotropic distribution of fully penetrable disks (random-dot model)[17] and totally impenetrable disks[16] as a func-

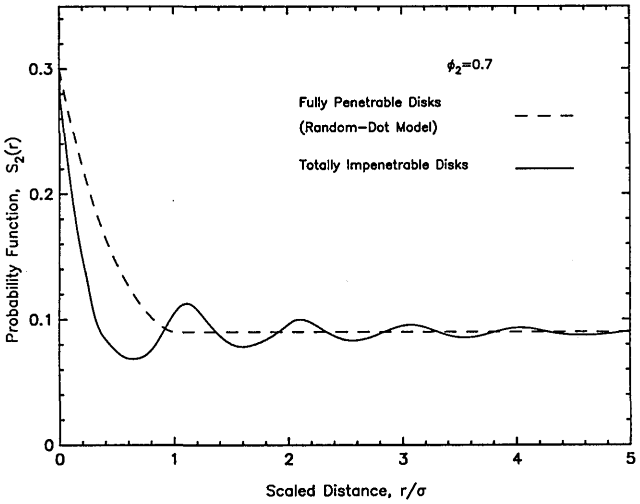

Fig. 2. Two-point probability function $S_2(r)$ as a function of the distance r for distributions of disks of diameter σ at a disk area fraction $\phi_2 = 0.7$.

tion of the distance r for a particle area fraction $\phi_2 = 0.7$. The function $S_2(r)$ for the random-dot model decays exponentially until it achieves its long-range value at $r = \sigma$, but the corresponding function for impenetrable disks oscillates around its long-range value for small r, indicating short-range order due to exclusion-volume effects that are totally absent in the random-dot model.

The two-point probability function computed by Torquato and Lado[16] for impenetrable disks (without attractive interactions) assumed an equilibrium ensemble. For an equilibrium distribution, the two-body distribution functioning (called the radial-distribution function for isotropic media) can be obtained by solving the Ornstein–Zernike integral equation.[20] Torquato and Lado used the solution of this integral equation in the Percus–Yevick approximation as obtained by Lado.[20] In general, the impenetrability condition alone does not uniquely determine the distribution (ensemble). One may assume an equilibrium distribution or some nonequilibrium distribution, such as random-sequential addition of hard particles, for which Smith and Torquato[21] computed S_2.

Penetrable-Concentric-Shell Model

Interpenetrable-particle models lie between the two extremes of fully penetrable and totally impenetrable particles. One such model is the penetrable-concentric-shell model,[22] which considers particles that possess a smaller, internal hard core encompassed by a perfectly penetrable concentric shell. The size of the internal hard core is proportional to an impenetrability index λ, $0 \le \lambda \le 1$, with $\lambda = 0$ and $\lambda = 1$ corresponding to fully penetrable and totally impenetrable particles, respectively. This model enables one to change continuously the degree of overlap and hence the degree of connectedness of the particle phase. This more general model may be of value in photographic science applications. The two-point probability function for disks in the penetrable-concentric-shell model (cf. Fig. 3) has been computed by Smith and Torquato.[21] Equation (3.17) for S_n is general enough to treat the penetrable-concentric-shell model.

C. Explicit Relation for G for a Class of Anisotropic Media

Using the results of the previous two subsections, we can state an explicit relation for the granularity of the class of anisotropic media described in Subsection 3.B. Specifically, we substitute Eq. (3.17) for $n = 2$ into the general granularity expression from Eq. (3.6) and find that

$$G = \left[\frac{1-{\phi_1}^2}{{\phi_1}^2} - \frac{\rho}{{\phi_1}^2{A_a}^2}\int A_2(\mathbf{r})A_a^{int}(\mathbf{r})d\mathbf{r} + \frac{1}{{\phi_1}^2{A_a}^2}\int \sum_{k=2}^{\infty} {S_2}^{(k)}(\mathbf{r})A_a^{int}(\mathbf{r})d\mathbf{r}\right]^{1/2}, \quad (3.26)$$

where

$${S_2}^{(k)}(\mathbf{r}) = \frac{(-1)^k\rho^k}{k!}\int \ldots \int g_k(\mathbf{r}^k) \times \prod_{j=1}^{k}\left\{1-\prod_{i=1}^{n}[1-m(\mathbf{x}_i-\mathbf{r}_j)]\right\}d\mathbf{r}_j \quad (3.27)$$

and $A_2(\mathbf{r})$ is the union area of two particle regions in which the centroids are separated by the displacement $\mathbf{r}$. Note that anisotropy appears in terms involving two- and higher-body effects, i.e., in the sum of Eq. (3.26). It is useful to recall the class of anisotropic media for which Eq. (3.26) is applicable. Equation (3.26) is valid for distributions of identical particles whose positions are completely specified by center-of-mass coordinates $\mathbf{r}_1, \mathbf{r}_2, \ldots$, which in two dimensions include circular disks, oriented rectangles, and oriented ellipses. The interparticle interactions that one can consider are perfectly general; hence, the particles may

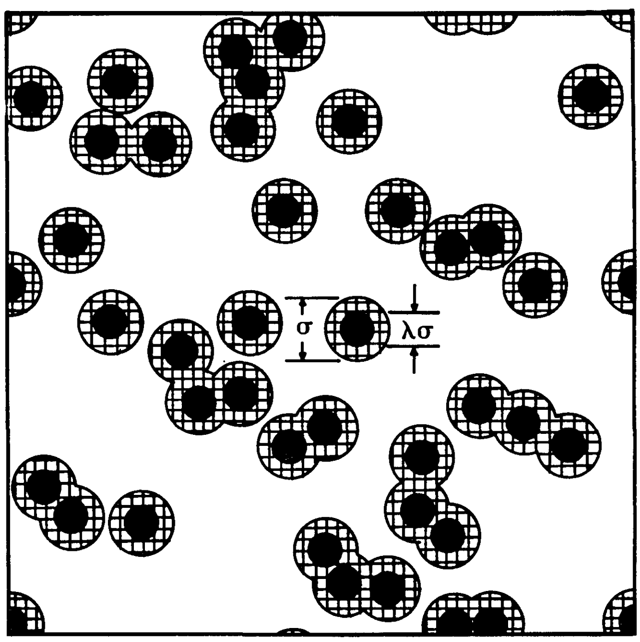

Fig. 3. Computer-generated realization of a distribution of disks of diameter σ (hatched and black regions) in the penetrable-concentric-shell model.[22] The disks have a hard core of diameter $\lambda\sigma$ indicated by the smaller, black circular region. Here $\lambda = 0.5$ and $\phi_2 \simeq 0.3$.

partially overlap one another, interact through repulsive (e.g., impenetrable cores and coulombic forces) as well as attractive forces, etc.

4. CALCULATION OF GRANULARITY FOR DISTRIBUTIONS OF DISKS AND A CIRCULAR APERTURE

We shall apply previous results to obtain specific expressions for the granularity for distributions of equisized circular disks of diameter σ in both the fully penetrable disk (random-dot) model and the totally impenetrable disk model with a circular aperture of diameter σ_a. We then compute these expressions for arbitrary aperture sizes. The calculations for impenetrable disks are believed to be new.

A. Expressions for Circular Disk and a Circular Aperture

Fully Penetrable Disks

For fully penetrable disks, it is useful to substitute Eqs. (3.10), (3.20), (3.22), and (3.23) into the general expression, Eq. (3.6), to yield

$$G = \frac{4}{\sqrt{\pi}\sigma_a}\left\{\int_0^{\sigma_a} r\{\exp[\rho A_2^{int}(r)] - 1\} \times \left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]\mathrm{d}r\right\}^{1/2}, \quad (4.1)$$

where

$$A_2^{int}(r) = \frac{\sigma^2}{2}\left[\cos^{-1}\frac{r}{\sigma} - \frac{r}{\sigma}\left(1 - \frac{r^2}{\sigma^2}\right)^{1/2}\right]H(\sigma - r), \quad (4.2)$$

which agrees with the expression given by Bayer[6] (apart from a trivial constant due to a difference in our definitions of G).

Totally Impenetrable Disks

For an anisotropic distribution of totally impenetrable disks, Eq. (3.26) gives

$$G = \frac{\phi_2}{\phi_1}\left\{-1 + \frac{16\rho}{\phi_2^2\pi\sigma_a^2}\int_0^{\sigma_a} rA_2^{int}(r) \times \left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]\mathrm{d}r + \frac{8}{\phi_2^2\pi^2\sigma_a^2}\int_{r<\sigma_a} S_2^{(2)}(\mathbf{r}) \times \left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]\mathrm{d}\mathbf{r}\right\}^{1/2}, \quad (4.3)$$

where

$$S_2^{(2)}(\mathbf{r}) = \rho^2\int\int g_2(\mathbf{r}_{34})m(r_{13})m(r_{14})\mathrm{d}\mathbf{r}_3\mathrm{d}\mathbf{r}_4. \quad (4.4)$$

and m, as noted earlier, is simply given by Eq. (3.21). We have changed the variables $\mathbf{r}_{12}$, $\mathbf{x}_1 - \mathbf{r}_1$, and $\mathbf{x}_2 - \mathbf{r}_2$ to $\mathbf{r}_{34}$, $\mathbf{r}_{13}$, and $\mathbf{r}_{14}$, respectively, with $\mathbf{r} \equiv \mathbf{r}_{12}$. Note that anisotropy enters through the last integral of Eq. (4.3), i.e., through the term involving the two-body distribution function $g_2(\mathbf{r}_{34})$.

In the case of an isotropic distribution of impenetrable disks, Eq. (4.3) yields

$$G = \frac{\phi_2}{\phi_1}\left\{-1 + \frac{16\rho}{\sigma_2^2\pi\sigma_a^2}\int_0^{\sigma_a} rA_2^{int}(r) \times \left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]\mathrm{d}r + \frac{16}{\phi_2^2\pi\sigma_a^2}\int_0^{\sigma_a} rS_2^{(2)}(r) \times \left[\cos^{-1}\frac{r}{\sigma_a} - \frac{r}{\sigma_a}\left(1 - \frac{r^2}{\sigma_a^2}\right)^{1/2}\right]\mathrm{d}r\right\}^{1/2}. \quad (4.5)$$

B. Calculations for Impenetrable and Fully Penetrable Disks

We compute the granularity Eqs. (4.1) and (4.5), using a trapezoidal rule. In evaluating Eq. (4.5), we make use of the results of Torquato and Lado[16] for the two-point probability function of an equilibrium distribution of impenetrable disks without attractive interactions. Torquato and Lado provided results up to a disk volume fraction $\phi_2 = 0.7$, which corresponds to approximately 87% of the random-close-

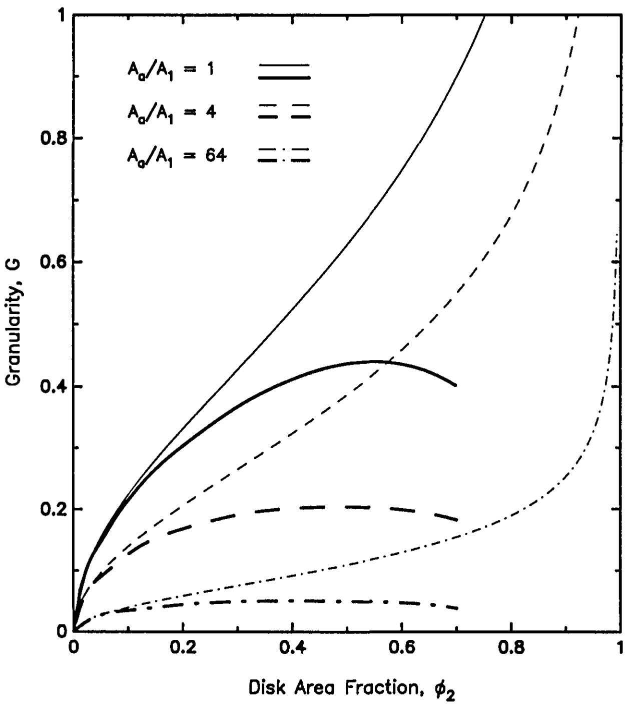

Fig. 4. Granularity G versus the disk-area fraction ϕ_2 for three different values of the scaled aperture area A_a/A_1 in both the fully penetrable disk (random-dot) model (lighter curves) and the totally impenetrable disk model (heavier curves). A_a is the aperture area ($\pi\sigma_a^2/4$), and A_1 is the disk area ($\pi\sigma^2/4$).

Table 1. Granularity, G, for Totally Impenetrable Disks as a Function of the Scaled Aperture Area A_a/A_1 for Several Values of the Disk Area Fraction ϕ_2[a]

	ϕ_2						
$\frac{A_a}{A_1}$	0.1	0.2	0.3	0.4	0.5	0.6	0.7
0.0	0.333	0.500	0.655	0.816	1.000	1.225	1.528
0.5	0.251	0.362	0.452	0.527	0.589	0.632	0.643
1.0	0.215	0.303	0.367	0.412	0.437	0.435	0.400
1.5	0.188	0.261	0.308	0.335	0.340	0.323	0.284
2.0	0.169	0.231	0.268	0.285	0.285	0.270	0.249
2.5	0.155	0.201	0.239	0.253	0.253	0.244	0.234
3.0	0.144	0.192	0.219	0.231	0.232	0.228	0.219
3.5	0.134	0.179	0.203	0.214	0.217	0.213	0.201
4.0	0.127	0.168	0.191	0.201	0.204	0.199	0.182
4.5	0.121	0.159	0.181	0.190	0.192	0.185	0.166
5.0	0.115	0.152	0.172	0.181	0.182	0.173	0.154
5.5	0.110	0.145	0.164	0.172	0.172	0.164	0.147
6.0	0.106	0.139	0.157	0.165	0.164	0.156	0.142
6.5	0.102	0.134	0.151	0.158	0.156	0.150	0.139
7.0	0.099	0.130	0.146	0.153	0.152	0.145	0.135
8.0	0.093	0.122	0.137	0.143	0.142	0.136	0.127
9.0	0.088	0.115	0.129	0.134	0.134	0.129	0.117
10.0	0.084	0.110	0.123	0.128	0.127	0.122	0.109
15.0	0.070	0.090	0.101	0.104	0.103	0.099	0.090
20.0	0.061	0.078	0.087	0.090	0.089	0.085	0.077
25.0	0.055	0.070	0.078	0.081	0.080	0.076	0.068

[a] A_a is the aperture area ($\pi\sigma_a^2/4$) and A_1 is the disk area ($\pi\sigma^2/4$).

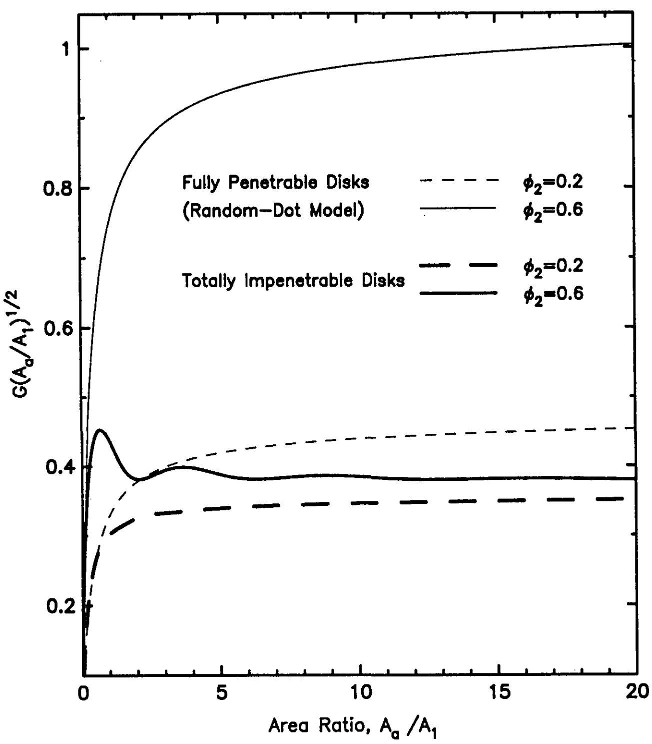

Fig. 5. Granularity G multiplied by the square root of the scaled aperture area, $(A_a/A_1)^{1/2}$, versus the scaled aperture area, A_a/A_1, for both fully penetrable disks (lighter curves) and totally impenetrable disks (heavier curves) at $\phi_2 = 0.2$ and 0.6. A_a is the aperture area ($\pi\sigma_a^2/4$), and A_1 is the disk area ($\pi\sigma^2/4$).

packing fraction.[23] We include the results for fully penetrable disks (random-dot model), first given by Bayer,[6] in order to compare them with the impenetrable-disk model.

In Fig. 4, we plot the granularity G as a function of the disk-area fraction ϕ_2 for various values of the scaled aperture area A_a/A_1 for both fully penetrable and totally impenetrable disks. For the same value of the scaled aperture area A_a/A_1, the granularity is always smaller for impenetrable disks than for fully penetrable disks at the same value of ϕ_2. This effect becomes more pronounced at higher values of the disk area fraction. Physically, this is true because impenetrable disks (unlike fully penetrable disks) cannot overlap one another and, in this sense, constitute a less random distribution than the random-dot model. For fixed A_a/A_1, the granularity for fully penetrable disks is a monotonically increasing function of ϕ_2. This dependence is in contrast to that of the impenetrable-disk model in which G increases with increasing ϕ_2 for small to moderate ϕ_2, reaches some maximum, and then decreases with increasing ϕ_2. Finally, we note that G decreases with increasing area ratio A_a/A_1 for either model.

In Table 1, we tabulate G for totally impenetrable disks as a function of A_a/A_1 for $0.1 \le \phi_2 \le 0.7$ in increments of 0.1. The same trends described above are seen in the table.

In Fig. 5, the function $G\sqrt{A_a/A_1}$ versus A_a/A_1 is plotted for both models for $\phi_2 = 0.2$ and $\phi_2 = 0.6$. Recall that for large apertures $G \sim A_a^{-1/2}$ [cf. Eq. (3.8)], and thus $G\sqrt{A_a/A_1}$ approaches a constant [equal to K of Eq. (3.9)] for large apertures. At $\phi_2 = 0.6$, $G\sqrt{A_a/A_1}$ for fully penetrable disks is more than twice as great as the corresponding quantity for impenetrable disks for $A_a/A_1 > 2$. An interesting effect is seen to occur in Fig. 5 for impenetrable disks at $\phi_2 = 0.6$. $G\sqrt{A_a/A_1}$ oscillates about its long-range value of K ($\phi_2 = 0.6$) for area ratios in the range $0 \le A_a/A_1 \le 15$. (Note that K for impenetrable disks was computed by Hong.[11]) The oscillations for impenetrable disks actually become more pronounced as ϕ_2 is made to increase, and the amplitude of the oscillations is greatest for sufficiently small area ratios, i.e., $A_a/A_1 < 5$. Oscillations begin to become noticeable at $\phi_2 = 0.4$ (not shown). The oscillations occur because of the short-range ordering that is due to exclusion-volume effects, which become significant at higher densities.

5. CONCLUSIONS

In this paper, we have presented a systematic theoretical procedure to express explicitly the granularity for a wide class of two-dimensional, anisotropic, particulate media in terms of the microstructure for apertures of arbitrary shape and size. Specifically, the granularity is given in terms of the n-body distribution function g_n, which characterizes the positions of n particles in the system. Given the interparticle interactions, one can, in principle, obtain the g_n and thus compute the granularity as a function of the aperture size. For concreteness, we have computed, for the first time to our knowledge, the granularity for a distribution of impenetrable, opaque disks (and a circular aperture) as a function of the aperture size for a wide range of densities. We have found that the effect of impenetrability is to reduce the granularity relative to the random-dot model, especially at high densities. All the results of the present study have been generalized for arbitrary dimensions elsewhere.[24] In a future study, it would be of value to consider the effect of film thickness on the granularity.

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REFERENCES

1. P. G. Nutting, "On the absorption of light in heterogeneous media," Philos. Mag. **26**, 243–426 (1913).
2. E. W. H. Selwyn, "A theory of graininess," Photogr. J. **75**, 571–580 (1935).
3. L. A. Jones and G. C. Higgins, "Photographic granularity and graininess II. The effect of variations in instrumental and analytical techniques," J. Opt. Soc. Am. **36**, 203–227 (1946).
4. J. C. Marchant and P. L. P. Dillon, "Correlation between random-dot samples and the photographic emulsion," J. Opt. Soc. Am. **51**, 641–644 (1961).
5. E. O'Neill, *Introduction to Statistical Optics* (Addison-Wesley, Reading, Mass., 1963).
6. B. E. Bayer, "Relation between granularity and density for a random dot model," J. Opt. Soc. Am. **54**, 1485–1490 (1964).
7. E. A. Trabka, "Crowded emulsions: granularity theory for monolayers," J. Opt. Soc. Am. **61**, 800–810 (1971).
8. J. E. Hamilton, W. H. Lawton, and E. A. Trabka, "Some spatial and temporal point processes in photographic science," in *Stochastic Point Processes*, P. A. W. Lewis, ed. (Wiley, New York, 1972), pp. 817–867.
9. P. E. Castro, J. H. B. Kemperman, and E. A. Trabka, "Alternating renewal model of photographic granularity," J. Opt. Soc. Am. **63**, 820–825 (1973).
10. J. C. Dainty and R. Shaw, *Image Science* (Academic, London, 1974).
11. K. M. Hong, "Granularity of monolayers: hard-disk model," J. Opt. Soc. Am. A **5**, 237–240 (1988).
12. L. B. Schein, *Electrophotography and Development Physics* (Springer-Verlag, Berlin, 1988).
13. S. Torquato and G. Stell, "Microstructure of two phase random media. I. The *n*-point probability functions," J. Chem. Phys. **77**, 2071–2077 (1982).
14. S. Torquato and G. Stell, "Microstructure of two phase random media. III. The *n*-point matrix probability functions for fully penetrable spheres," J. Chem. Phys. **79**, 1505–1510 (1983).
15. S. Torquato and G. Stell, "Microstructure of two phase random media. V. The *n*-point matrix probability functions for impenetrable spheres," J. Chem. Phys. **79**, 980–987 (1985).
16. S. Torquato and F. Lado, "Characterization of the microstructure of rigid rods and disks in a matrix," J. Phys. A **18**, 141–148 (1985).
17. S. Torquato and J. D. Beasley, "Effective properties of fiber-reinforced materials. I. Bounds on the thermal conductivity of dispersions of fully penetrable cylinders," Int. J. Eng. Sci. **24**, 415–434 (1986).
18. C. G. Joslin and G. Stell, "Bounds on the properties of fiber-reinforced composites," J. Appl. Phys. **60**, 1607–1610 (1986).
19. S. Torquato and A. K. Sen, "Conductivity tensor of anisotropic composite media from the microstructure," J. Appl. Phys. **67**, 1145–1155 (1990).
20. F. Lado, "Equation of state of the hard-disk fluid from approximate integral equation," J. Chem. Phys. **9**, 3092–3096 (1968).
21. P. A. Smith and S. Torquato, "Computer simulation results for the two-point probability function of composite media," J. Comput. Phys. **76**, 176–191 (1988).
22. S. Torquato, "Bulk properties of two-phase disordered media. I. Cluster expansion for the effective dielectric constant of dispersions of penetrable spheres," J. Chem. Phys. **81**, 5079–5088 (1984).
23. F. H. Stillinger, E. A. DiMarzo, and R. L. Kornegay, "Systematic approach to explanation of the rigid disk phase transition," J. Chem. Phys. **40**, 1564–1576 (1964).
24. B. Lu and S. Torquato, "Local volume fraction fluctuations in heterogeneous media," J. Chem. Phys. (to be published).