

Report No. IITRI-U6003-18
(Quarterly Report)

INVESTIGATION OF LIGHT SCATTERING
IN HIGHLY REFLECTING PIGMENTED COATINGS

National Aeronautics
and Space Administration

IIT RESEARCH INSTITUTE

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Prepared by

G. A. Zerlaut, B. H. Kaye
and M. R. Jackson

of

IIT RESEARCH INSTITUTE
Technology Center
Chicago, Illinois 60616

for

National Aeronautics and Space Administration
Office of Advanced Research and Technology
Washington 25, D. C.

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FOREWORD

This is Report No. IITRI-U6003-18 (Quarterly Report) of IITRI Project U6003, Contract No. NASr-65(07), entitled "Investigation of Light Scattering in Highly Reflecting Pigmented Coatings". This report covers the period from February 1, 1965 to May 1, 1966. Previous Quarterly Reports were issued in October 1963, February 1964, May 1964, September 1964, January 1965, March 1965, May 1965, August 1965, November 1965, and February 1966. The project is under the technical direction of the Research Projects Laboratory of the George C. Marshall Space Flight Center, and Mr. Daniel W. Gates is project manager.

Major contributors to the program include G. A. Zerlaut, project leader; Dr. S. Katz and Dr. B. H. Kaye, theoretical analysis; and M. R. Jackson, experimental investigator. Experimental data are recorded in logbooks C16369, and C16765.

Respectfully submitted,
IIT RESEARCH INSTITUTE

G. A. Zerlaut (B&K)

G. A. Zerlaut
Group Leader
Polymer Research

Approved by:

T. H. Meltzer (AL)

T. H. Meltzer
Manager
Polymer Research

GAZ/am/jb

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INVESTIGATION OF LIGHT SCATTERING
IN HIGHLY REFLECTING PIGMENTED COATINGS

I. INTRODUCTION

The objective of this program is the application of light-scattering theories to polydisperse, highly reflecting, highly pigmented coatings. The program is aimed at a definition of the light scattering parameters associated with the maximum reflection of solar radiation. The definition of these factors should facilitate the eventual development of more efficient solar reflectors and, perhaps more important, may extend the applications of light-scattering theory to the solution of other problems.

Previous work has involved (1) a review of classical light-scattering theory with emphasis on that portion having the most promise for application to multiple scattering events, (2) the generation of data on the optical properties of carefully prepared arrays of silver bromide particles dispersed in gelatin, and (3) the conception of theoretical approaches and random-walk techniques with which to treat the problem of multiple scattering.

The adaptation of classical Mie theory to multiple scattering and the experimental studies on silver bromide dispersions have been discussed in several of the previous Quarterly Reports. The complete review of classical light-scattering theory will be given in the summary Final Report which is currently planned

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for distribution in July of this year. This report will also contain a complete review of the studies pertaining to the silver bromide dispersions.

This report continues the Monte Carlo investigations which have been performed during the last year (Reports No. IITRI-C6018-14, IITRI-C6018-15, IITRI-U6003-16, and IITRI-U6003-17). This discussion is primarily concerned with the growth of clusters of pigment particles as the pigment volume concentration is increased. Also presented is a critique of current concepts and studies pertaining to Mie theory, a subject of special relevance to these studies.

II. MONTE CARLO STUDIES OF CLUSTER FORMATION OF PIGMENT PARTICLES IN A PAINT FILM

In simulation experiments reported previously the growth of cluster formation was simulated in two dimensions using a two dimensional grid-plotting experiment (ref. 1). Although the validity of the quantitative data on cluster formation deduced from these experiments is limited by the fact that clusters are growing in three dimensions, the qualitative deductions (such as the presence of a maximum in the total number of scattering centers) would appear to correlate with known empirical data on the changes in opacity of a paint film at various solids concentrations. A criticism of the simple cubic pigment-particle model used in the first Monte Carlo experiment is that the particle shape assumed is too symmetrical and that results from the plotting experiment are not valid because real pigment particles have non-symmetrical shapes. To explore the implications of this possible criticism the following plotting experiment was carried out. A square grid containing 70 x 70 square subdivisions was marked out. On this grid particles consisting of two squares were plotted using three random numbers.

The first two random numbers selected ranged between 1 and 70 to find a location on the plotting grid. The third random was a single digit and if it was even the particle was plotted with its left-side-lower-corner on the co-ordinate and longer side lying horizontally. If it was odd, the left-side-lower-

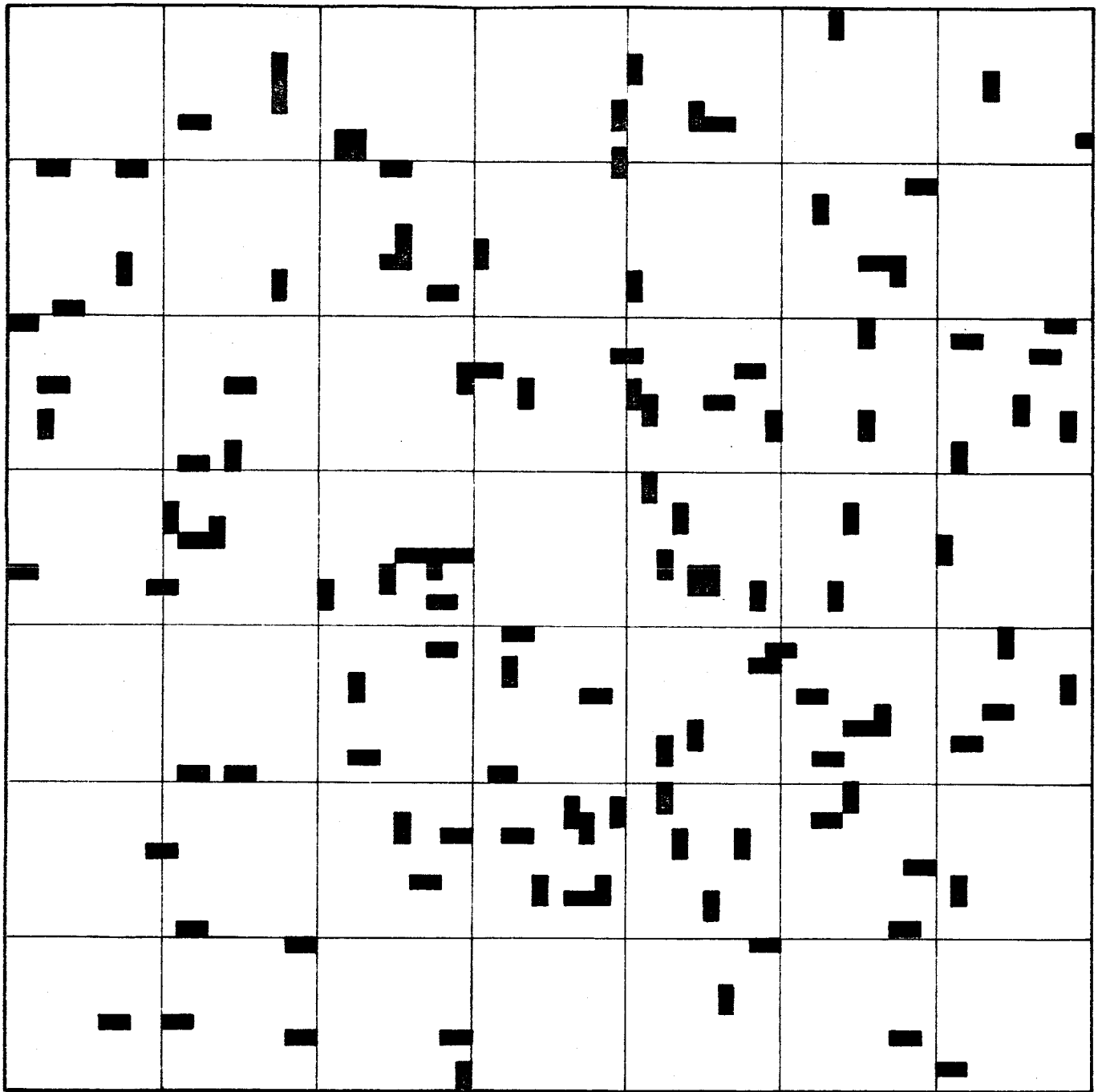


Figure 1

5% COVERAGE BY PARTICLES HAVING 2:1 SHAPE RATIO

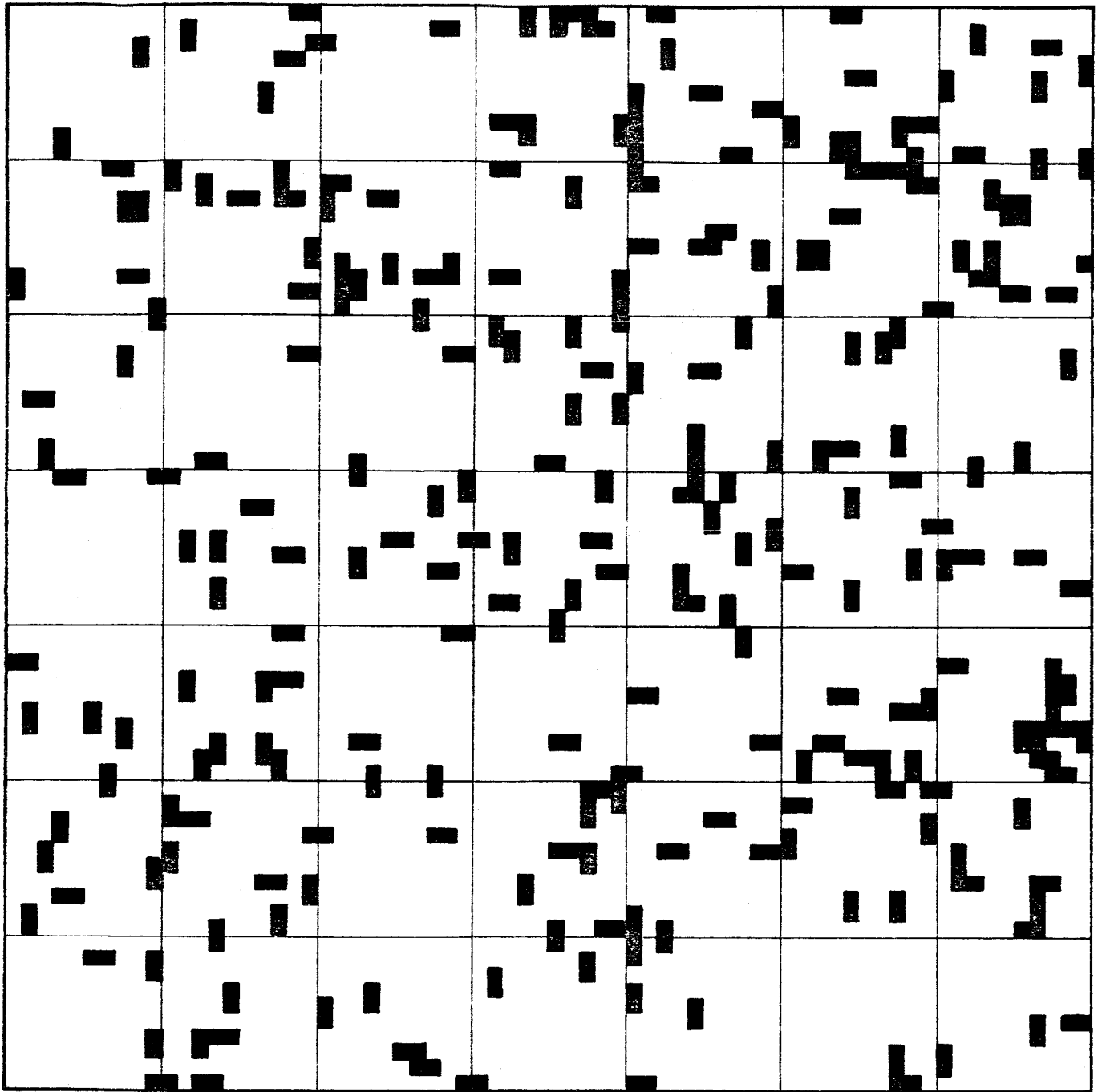


Figure 2

10% COVERAGE BY PARTICLE HAVING 2:1 SHAPE RATIO

Figure 3

14.8% COVERAGE BY PARTICLE HAVING 2:1 SHAPE RATIO

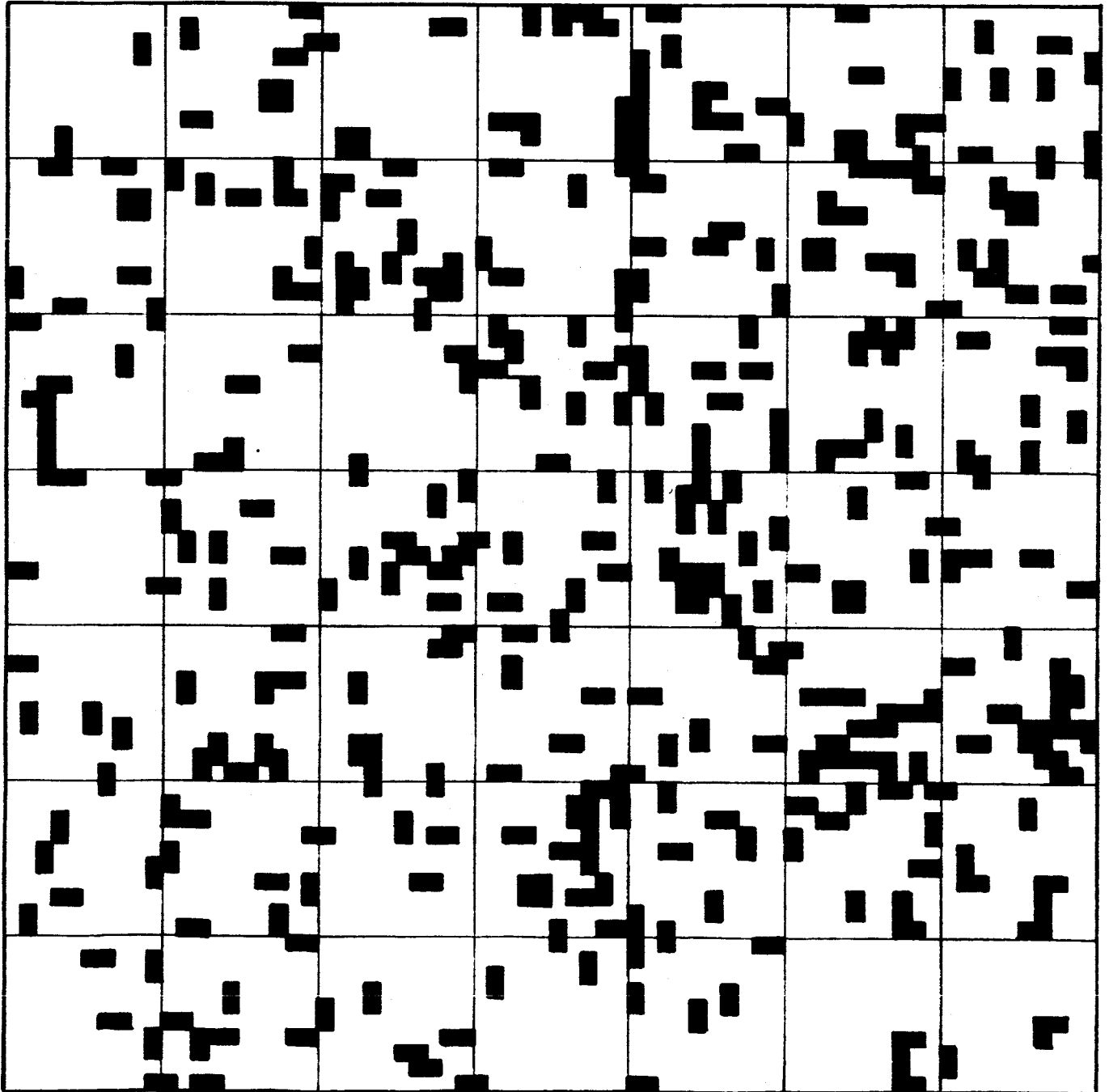
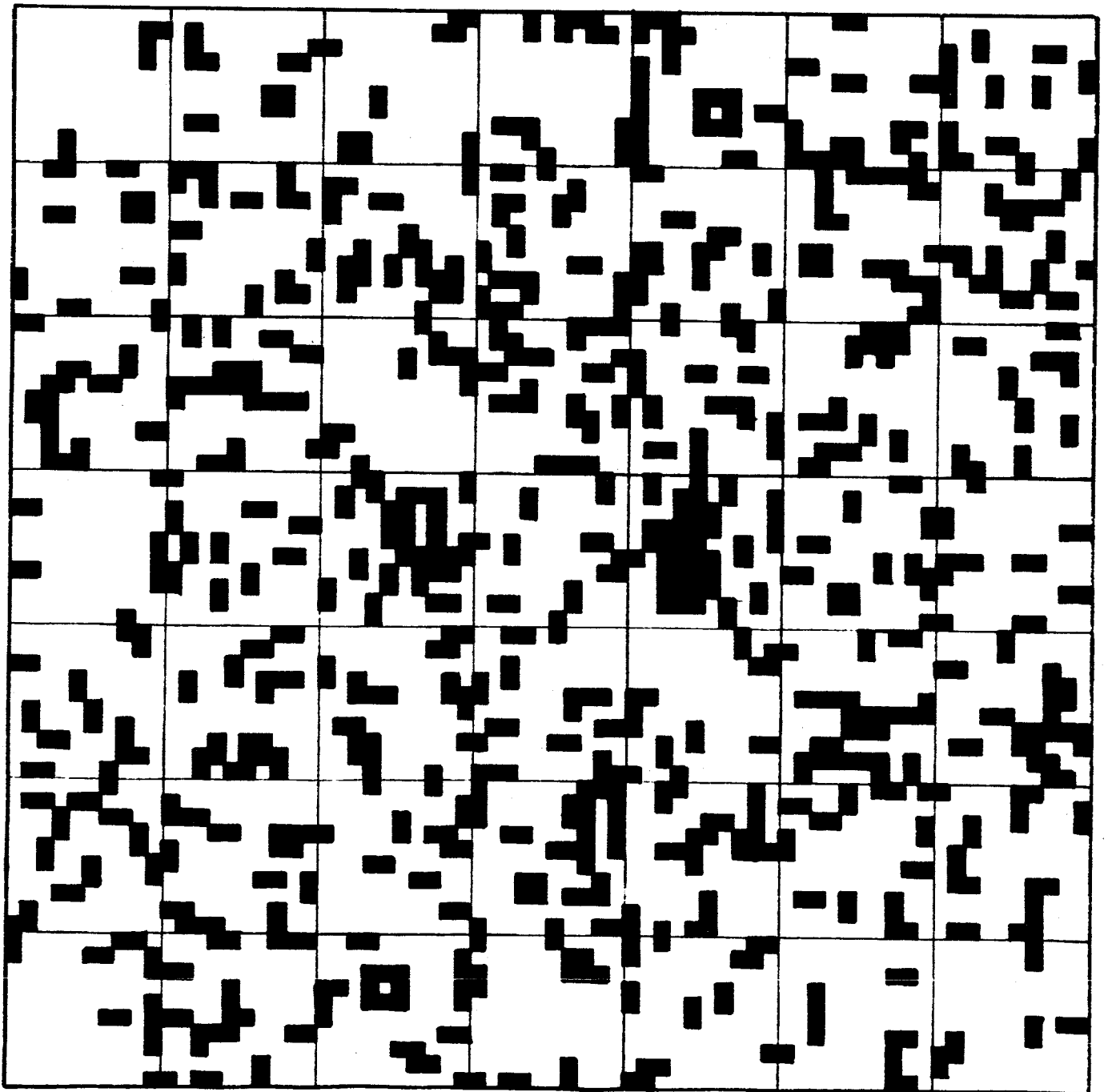


Figure 1

20% COVERAGE BY PARTICLE HAVING 2:1 SHAPE RATIO



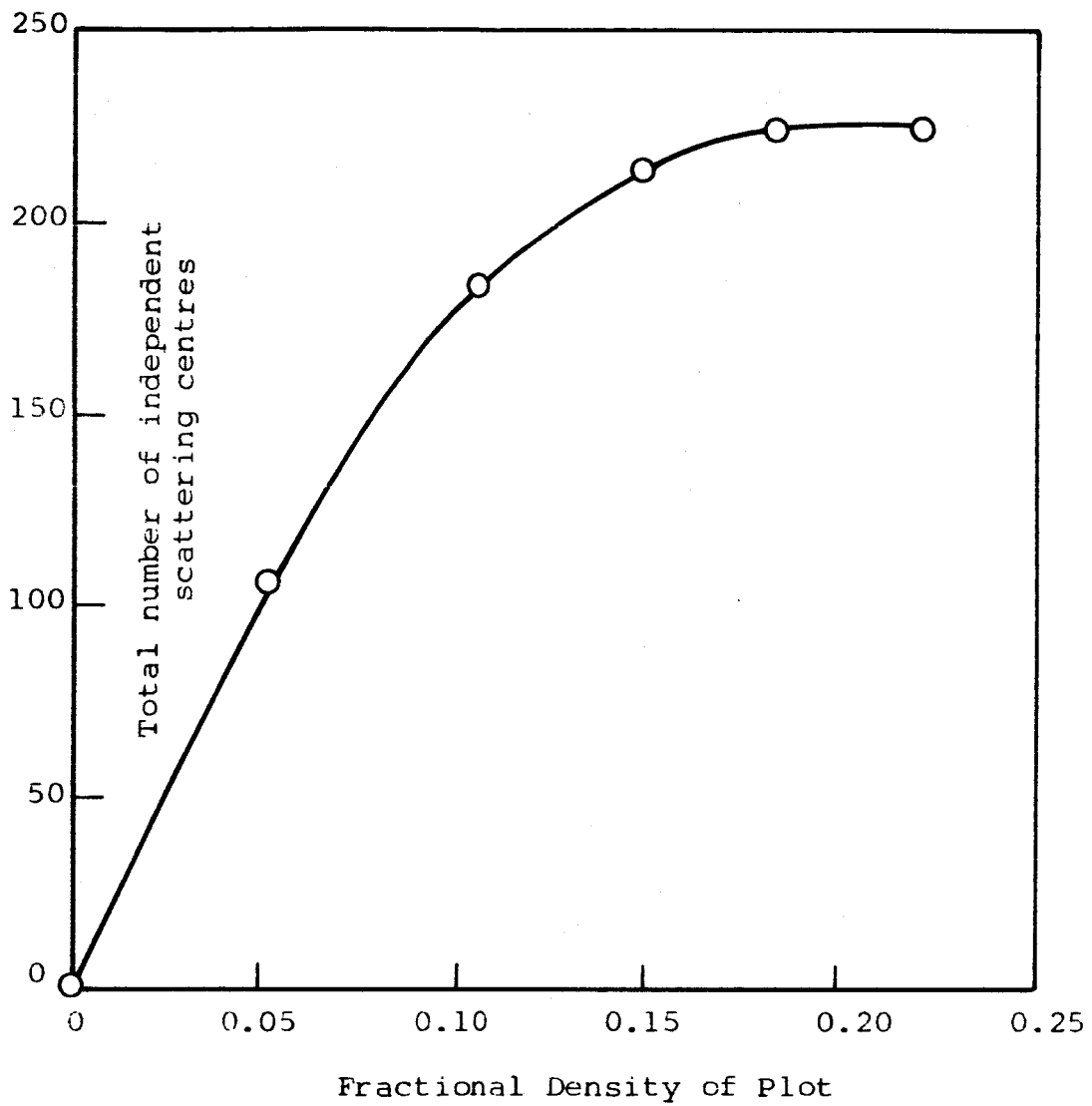


Figure 5

ABSOLUTE NUMBER OF SCATTERING CENTERS
PER UNIT VOLUME FOR PARTICLES
HAVING 2:1 SHAPE RATIO

slightly higher concentrations for the particles with a shape factor of 2:1. This phenomena may not be real in the sense that statistical fluctuations between repeat Monte Carlo plotting experiments could demonstrate that the difference between it and 20% for the two different shaped particles could arise purely from chance mechanisms. The results of the plotting experiments with the 2:1 shaped particles have important implications for paint reflectance studies.

First of all, irrespective of the significance of the small shift from 17 to 20%, these experiments have confirmed that cluster formation imposes a limit on the number of scattering centers achieved and that 20% by volume is the order of magnitude for pigment concentrations beyond which there is a loss in effective scattering power.

The concentration at which the number of independent scattering centers starts to fall off is approximately the same both for the square model and the 2:1-ratio model particles and can be understood from an examination of the structure of Figures 1 to 4 and the following qualitative reasoning. As long as the spaces between particles is several particles wide, the chances that a particle 2-diameters wide will touch another particle is not much higher than if it is one diameter wide. For instance, consider a sphere of $\frac{n}{2}$ -diameters radius such that if a small particle is placed within this sphere it can be considered to touch another particle if it at least touches the surface of the sphere. In one sense the sphere can be considered an abstract

model of an unpopulated region within a pigmented material and in the following discussion it will be referred to as the location sphere. Now the chance that a center of a particle of diameter d will be located within the larger sphere so that it will touch its surface or extend beyond it is the ratio:

$$P_d = \frac{1/6 \pi d^3 n^3 - 1/6 \pi d^3 (n-1)^3}{1/6 \pi d^3 n^3} \quad (1)$$

Thus:

$$P_d = 1 - (1-1/n)^3$$

It is important to notice that this relationship does not involve d and this is why the relative locations within a paint film are not a function of particle size.

If we now consider the particle which has a shape factor of 2 to 1, this can be located in a larger volume in space and still protrude through the surface of the location sphere to make contact with a particle to form a cluster. For particles of shape factor 2:1 with their long axis lying along the radius of the location sphere, the probability of being in a position to form a cluster is

$$P_d (2:1) = 1 - (1-2/n)^3$$

However, only a fraction of the particles will lie along the radius of the location sphere and the probability for particles

occupying all possible orientations within the location sphere will be

$$P_d(2:1) = 1 - (1 - \alpha/n)^3 \quad (2)$$

where α is some number between 1 and 2. From a comparison of the probabilities expressed in equations 1 and 2 it can be seen that the probability of cluster formation for the non-circular particle is higher than the spherical particle (from spatical considerations evaluated in isolation from other factors influencing cluster formation). It can also be seen from such a comparison that the difference between them is small when n is large but increases rapidly as n tends towards α . There is however a competing factor which tends to reduce the probability of cluster formation for the particles of shape factor 2:1 compared with the particles of 1:1 symmetry. For a given mass of particles, the number of points in space at which a particle can occur for systems containing particle 2:1 is half that for particles 1:1 symmetry. This means that the effective value of n to be used in Equations 1 and 2 is larger for particles of 2:1 shape factor than for the 1:1 symmetrical particles at a given volume concentration. Therefore, we have two competing factors -- one tending to reduce cluster formation and one tending to increase it for the particles of shape factor 1:2. The competitions between these two factors could offer a qualitative explanation in the shift of the peak for maximum number of scattering centers towards the higher volume concentrations for the 2:1 shaped particles

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should subsequent investigation confirm this shift.

It should be noted that this possible shift towards higher concentrations for achieving maximum number of scattering centers is not an argument for trying to achieve pigment particles with 2:1 shape factors because if one compares the number of scattering centers achieved for a given mass of particles the number is always superior for the 1:1 symmetry particles. It is also interesting to note that if for instance both types of particles were measured by sieving techniques* then, since in sieving techniques the particles are classified by their minimum diameter, both types of particles we have been considering would have the same measured particle size yet one set of particles would have a much higher number of scattering centers per unit mass. This suggests that it may not be possible to solve some problems associated with the optical properties of paint films until quantitative methods of shape analysis are developed.

At the end of the discussion of the relation between Mie Theory predictions of the scattering power of single particles

* This is not possible for normal pigment particles at the current stage of size analysis technology but the fact that we are discussing a set of hypothetical measurements does not affect the validity of the points which are being made concerning particle size and paint film properties.

and the optical properties of paint films it was postulated that it may be advantageous to achieve maximum pigment surface per film thickness provided that the individual particles were still effective scattering centers. From this point of view it might be argued that 2:1 or higher shape factors might be advantageous from the point of view that they have more surface per mass than a sphere. Again this is a superficial argument which has no real meaning unless one defines particle size very carefully. For example, consider the simple cubic model and the simple particle of 2:1 shape factor formed by fusing two cubes together over one surface. Now if we consider the cube and the particle formed by fusing two together as having the same particle size, which they would have from sieving techniques, then, although the particle formed from two cubes has more surface than a particle of the same mass, it has less surface than the two cubes from which it was formed. Thus, from this point of view the cubical particles have more scattering centers and more surface area per unit mass than do particles of 2:1 shape factor when one dimension is identical for the two particles. This apparent paradox arises from the fact that one normally talks about surface area per unit mass whereas in discussing the properties of pigment particles the property of interest is surface area for a given particle size where the meaning of particle size has to be defined carefully. Again this discussion underlines the need for a knowledge of shape factors in conjunction with accurate and well defined particle size analysis.

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It is possible to discuss the possible influence of dispersion on the optical properties of a paint film using the statistical considerations outlined in the foregoing paragraphs. Consider a mono-sized pigment. If the particles are not well dispersed in the paint film, this is equivalent to saying that the units of pigment to be dispersed are not single particles but groups of particles containing 2, 3, 4 up to n particles. A cluster of two particles can be considered to be a particle of shape factor 2:1. The case of a cluster containing a number of particles is more complex because of the possible configurations in space which they can achieve; however, all clusters in fact represent basic units of increasing particle-shape factor as the cluster size increases. From the statistical considerations given above one would anticipate that in a badly dispersed pigment the concentration of solids for maximum scattering centers is displaced towards the higher concentration but that the total number of scattering centers is low. Therefore, increasing the degree of dispersion should increase the overall opacity and should shift the maximum of the opacity/concentration curve towards lower concentrations. This conclusion is based on tentative postulation and very simple models but its implications are sufficiently interesting to warrant investigation. If the conclusion proves to be a real description of the properties of a paint film, then an interesting corollary to the hypothesis is that the location of the maximum in the concentration/maximum

scattering centers curve will always be a function of the shape factor of a well dispersed pigment.

It is pertinent at this point to discuss the relevance of the speculations outlined above based on statistical reasoning. Even if all of the above speculations proved to be irrelevant, at least this theoretical study has indicated the possible phenomena occurring within the paint film. Knowing these possible phenomena, we can design experiments efficiently so that the importance of the possible mechanisms effecting the opacity of paint films can either be substantiated or eliminated. In Figure 6 the growth of the different sized cluster in the plotting experiment using 2:1 shape factor particles are presented.

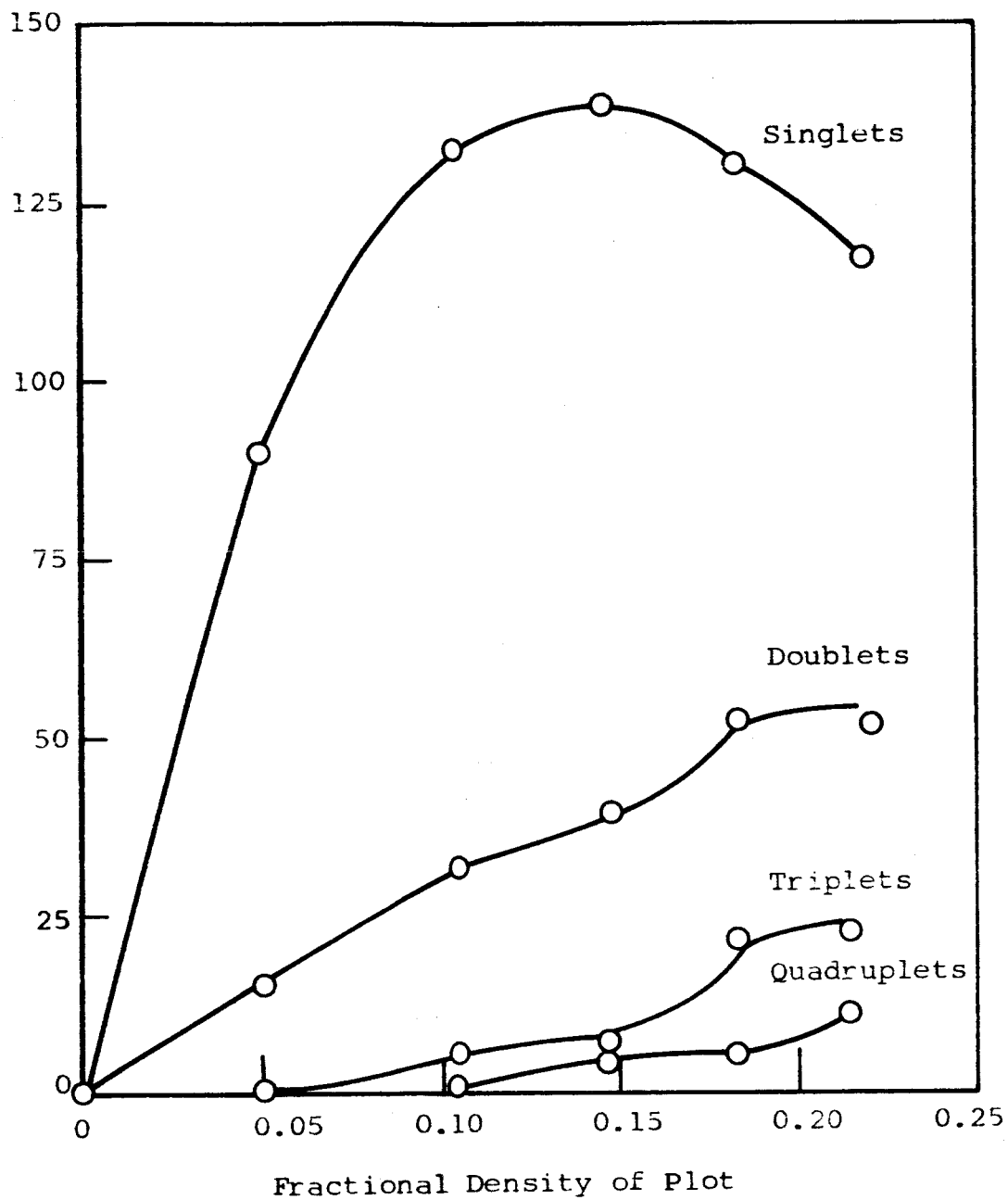


Figure 6

NUMBER OF DIFFERENT SIZED CLUSTERS AT VARIOUS VOLUME CONCENTRATIONS FORMED FROM PARTICLES HAVING 2:1 SHAPE RATIO

III. THE RELEVANCE OF MIE THEORY STUDIES FOR THE PREDICTION OF THE REFLECTIVE PROPERTIES OF PAINT FILMS

The scattering power of a single smooth spherical particle placed in the path of a plane parallel beam of noncoherent monochromatic light can be studied by the theory developed by Mie (ref. 2). The variations in scattering power of the single sphere with refractive index and particle diameter-wavelength ratio has been computed using the Mie theory and this curve is often used to justify the claim that optimum opacity is obtained using a pigment-particle size of about $1/5$ the wavelength of the incident light. When the relevance of the Mie theory to the physical phenomena occurring within the paint film is considered, it is found that even if the maximum-opacity pigment size eventually proves to be $1/5$ of the wavelength, this fact cannot possibly be deduced from the Mie theory. Any agreement between fact and speculation stimulated by considering Mie theory can only be fortuitous. The relationship between Mie theory and phenomena occurring within a paint film can be understood by considering several aspects of the interference and Fraunhofer diffraction patterns of long single slits for plane-parallel beams of monochromatic noncoherent light. Consider first the Fraunhofer diffraction pattern of single slits of various widths. These patterns are shown in Figure 7 (ref. 4). These patterns are observed on a screen at infinity (simulated using lenses). Consider now if the screens were replaced by a photoelectric device which could receive all scattered light. We would not be concerned with the spatial distribution of the energy and our observation would be that "the total ener-

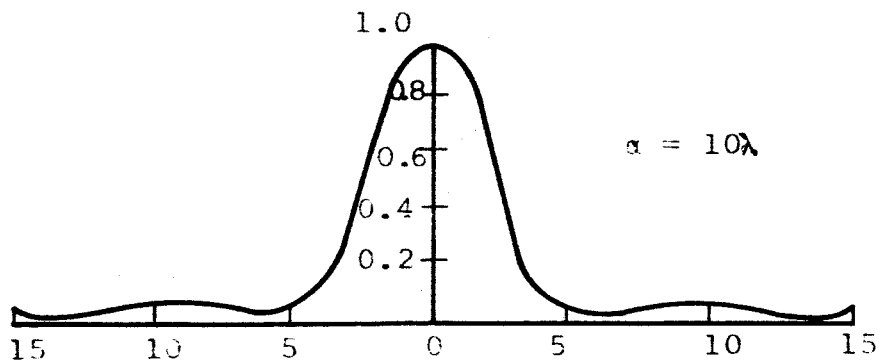
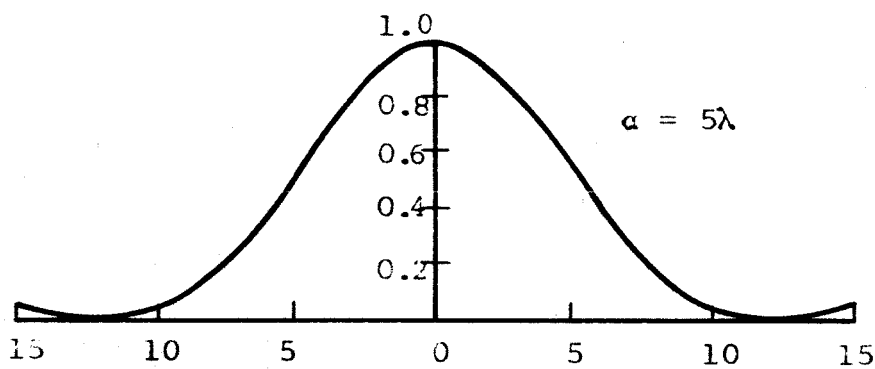
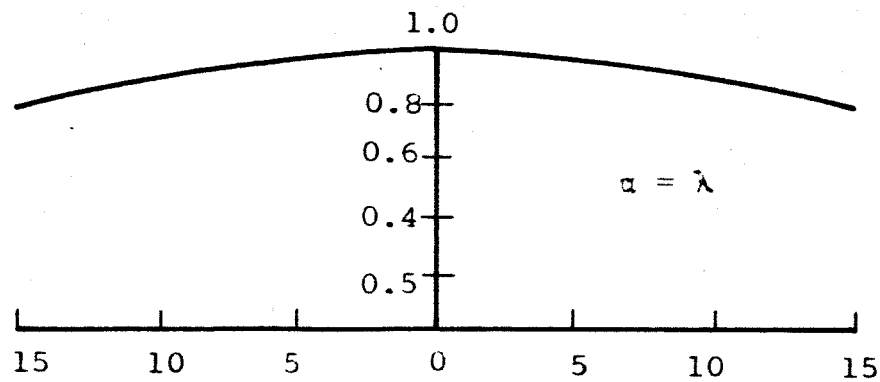


Figure 7

THE RELATIVE INTENSITY IN SINGLE SLIT DIFFRACTION
FOR THREE VALUES OF SLIT-WIDTH OF THE RATIO α/λ WHERE
 λ IS THE WIDTH OF THE SLIT

gies from the three slits was simply proportional to the slit widths, i.e., 1:5:10". From this point of view we are not troubled by the fact that geometric optics does not apply to the system. Consider now the diffraction pattern of a set of slits each 5 wavelengths wide but 50 wavelengths apart. The resultant pattern is as shown in Figure 8 (ref. 4). The interference fringes lie within the envelope of the diffraction pattern for the single slit. Now, familiarity with these diagrams in school text books on optics tends to obscure the important point. Although the relative intensity pattern is dominated by the system predicted from the single slit, the total energy passing through the slits is proportional to the number of slits. Again if the screen is replaced by a photoelectric detector capable of receiving all the diffracted light, then the measurement device would not be able to differentiate between 20 slits, 2 wavelengths wide, or 40 slits, 1 wavelength wide, even though the spatial distribution of the energy for the two systems would be very different. Consider now what would happen if we had 50 parallel slits each 5 wavelengths wide but which were spaced randomly across the diffraction screen. Any pair of lines would produce an interference pattern modulated by the diffraction envelope, but the peaks for each pattern would be separated by a factor related to the separation of the two slits. The argument holds for any pair of slits so that the peak for each pair would fall at different points within the same diffraction envelope. Averaged

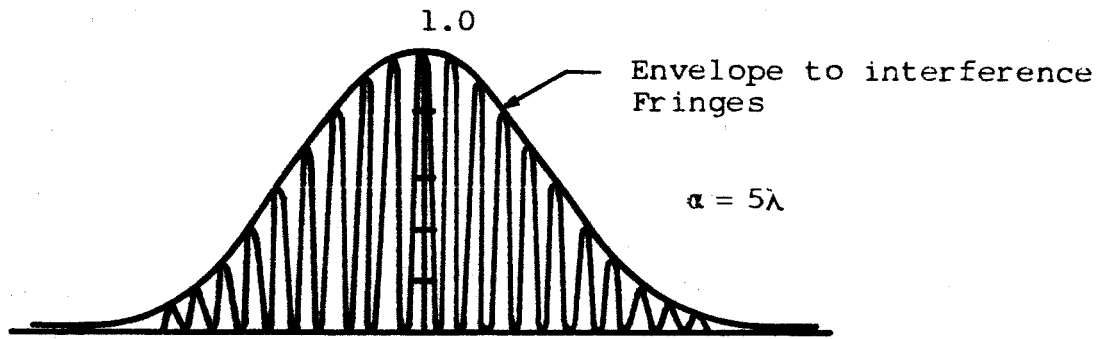


Figure 8

COMBINED INTERFERENCE AND DIFFRACTION PATTERN FOR
SLITS 5 WAVELENGTHS WIDE AND 50 WAVELENGTHS APART

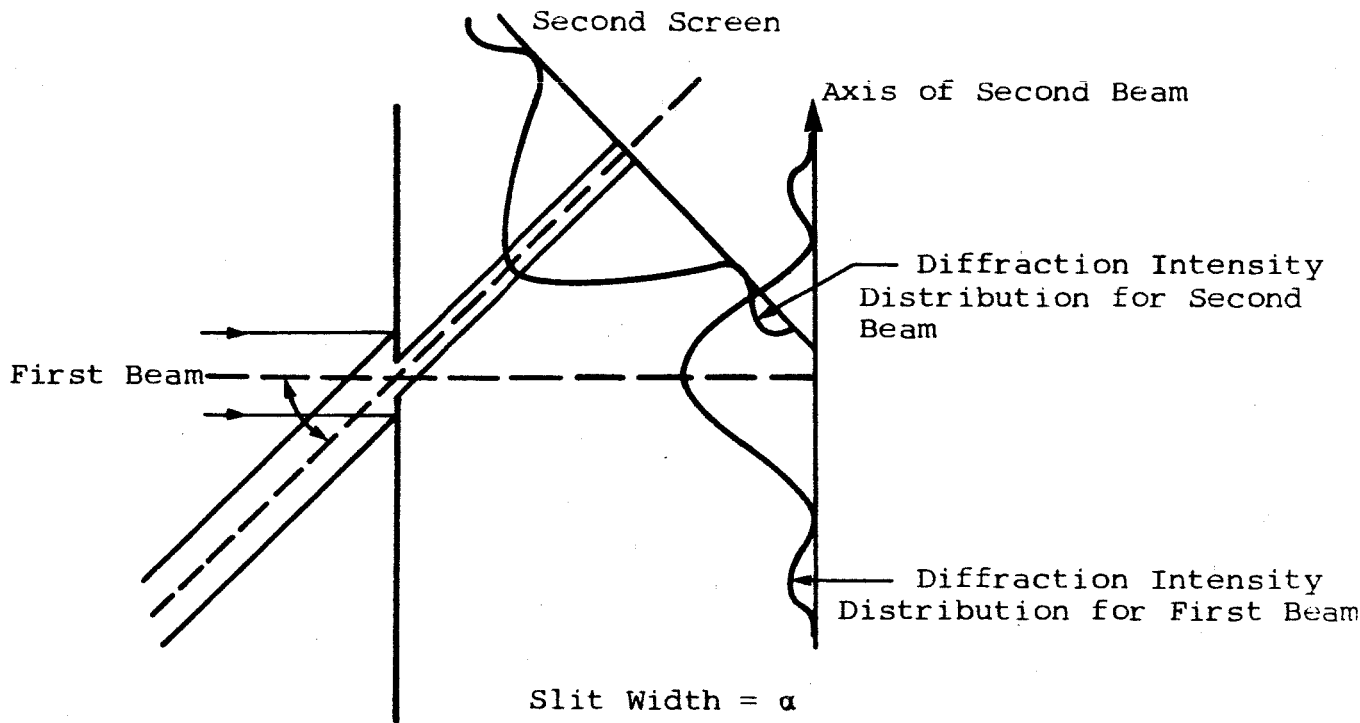


Figure 9

DIFFRACTION PATTERNS FOR TWO BEAM PASSING THROUGH THE SAME SLIT

out for the 50 lines the net effect would be a relatively uniform illumination modulated by the diffraction pattern for the single slit width. This results in the important observation that the diffraction pattern for 50 randomly-spaced slits is undistinguishable from that of 1 slit if the power of the beam for the single slit is 50 times that of the beam passing through the 50 random slits. Physically what happens is that the random positioning of the lines obscures the fine structure of the combined interference diffraction pattern.

So far we have only considered the effect of a single beam. If two beams at two different angles passes through the same slit, then the system will be as outlined in Figure 9. A screen placed on the axis of the first beam would show the diffraction for the specific slit width.* If a second beam at an angle θ is passed through the screen, then a screen placed on the axis of this second beam would have a slightly different pattern since the effective slit width is now $\sin \theta a$, so the prime diffraction lobe will be somewhat wider than that for the first beam. If, however, the screens are replaced by a semi-circular photoconductive device, then the power received by the device will be proportional to $(a + a \sin \theta)$ if the beams are of

* Note: The distances in Figure 9 are not to scale. Either the screen is placed at a relatively great distance from the screen or lenses have to be used to produce the pattern.

equal strength. If now there are beams at a whole range of equally spaced values of θ it can be seen that the calculation of the diffraction pattern on a screen perpendicular to the axis of the prime beam would be very complicated. If again the screen was now moved so that it was only several wavelengths from the screen, the simplified theory of Fraunhofer would no longer apply and the calculation of resultant diffraction patterns would now be exceedingly complex. If the system is now extended to consist of randomly spaced parallel slits of the same width and if all the beams are polychromatic, it can be seen that the calculation of diffraction patterns is now, in practice, impossible. However, the power penetrating the screen is still a function of the available area in the diffracting screen, and if the width of the screen is of the order of the wavelength of light it is probable that we could treat each hole as a self-luminous source of light.

Consider now if we had screens as shown in Figure 10. First let us consider that the 14 slits shown are a wavelength wide, and that the portion between the slits are painted with a completely absorbing black paint. From discussions given earlier, if one screen was placed in the path of a plane parallel beam of monochromatic light then the diffraction pattern observed on a screen placed at a distance large compared to the wavelength of light is the diffraction pattern of a single slit and the power is 14 times that for a single slit.

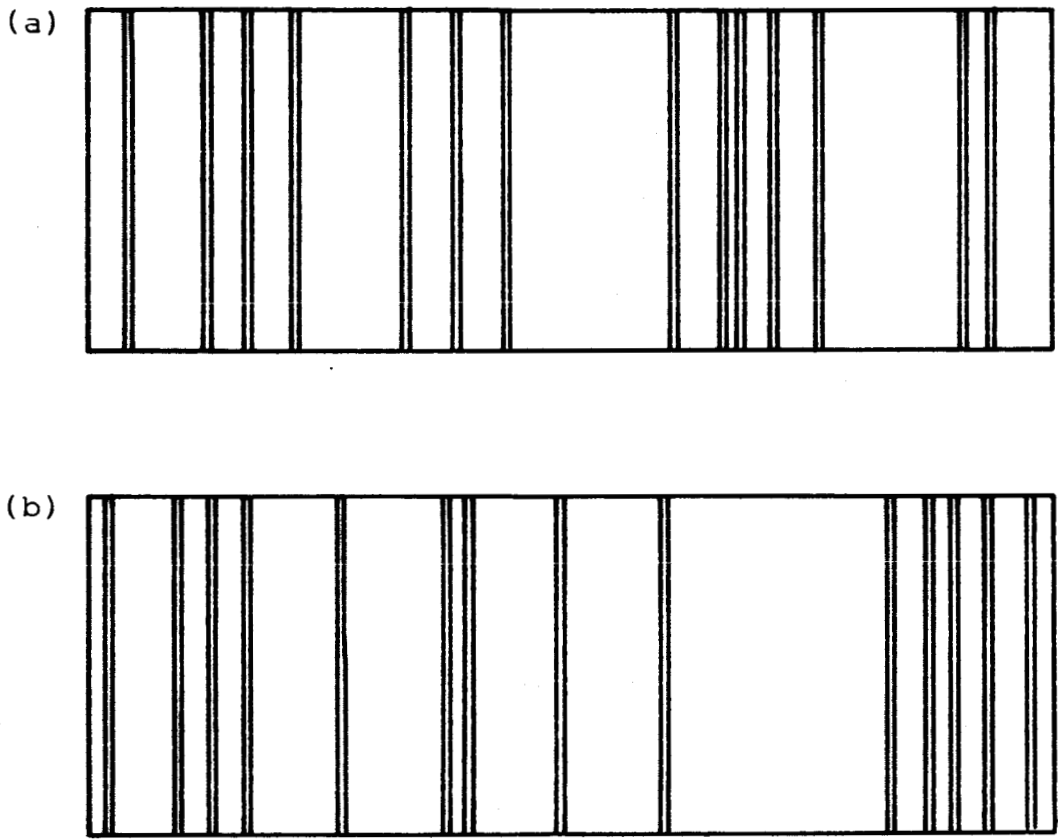


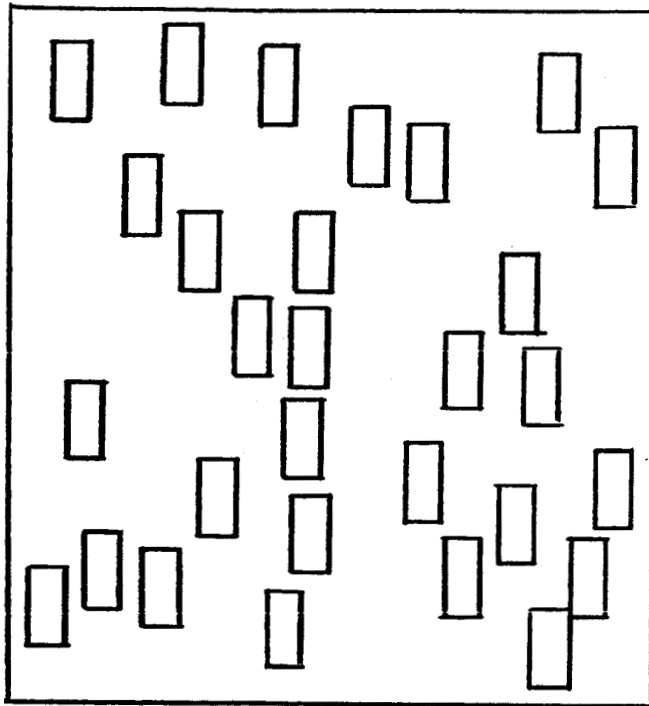
Figure 10

SCREENS CONTAINING RANDOMLY SPACED
PARALLEL SLITS OF EQUAL WIDTH

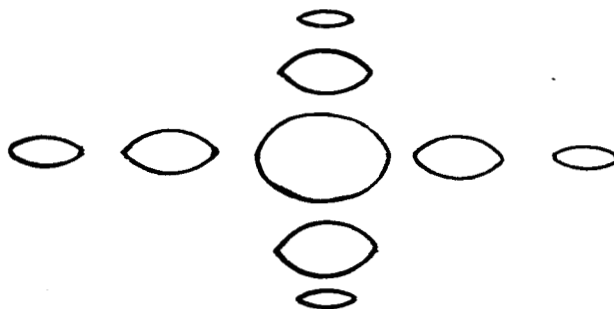
Now, however, consider the problem of studying the passage of diffuse white light through 100 of these screens placed three slit widths apart when the surface of the screen between the slits is 80% reflective. From one point of view the diffuse light can be considered to be a multiplex of parallel equipowered beams of light at equally spaced units of solid angles in space. By considering the complications arising from the combined effects of multiple-wavelength light and multidirectional beams, it is very obvious that a knowledge of the Fraunhofer diffraction pattern of a single screen containing randomly spaced slits will be of very little use in solving the multi, partially reflecting, screen problem involving diffuse white light. However, it can be shown that the Mie theory studies are related to paint film phenomena in approximately the same manner as the Fraunhofer pattern of the single screen containing randomly positioned slits is related to the multi-screen problem.

In order to gain an appreciation of the relevance of some of the published studies of the optical properties of pigment particles to the general problem of paint reflectance, it is necessary to extend the discussion of diffraction phenomena to the case of a screen containing randomly positioned regular apertures. This problem has been discussed by C. L. Andrews (ref.5).

He considered Fraunhofer diffraction for monochromatic non-coherent radiation for the screen shown in Figure 11. He states that, "if a very large number of identical rectangular apertures with identical orientation in a plane screen are randomly scattered about on the screen, the interference pattern of the combination will be smoothed out to constant intensity, but the diffraction pattern will be the same as that for a single rectangular aperture". The bright spots of the Fraunhofer pattern for the single aperture are shown in Figure 11. The screen in Figure 11 is in fact a two dimensional extension of the essentially one dimensional case of the randomly spaced slits discussed earlier and the considerations leading to the conclusions given by Andrews are in fact extensions of those given for the random slits to two dimensions. Andrews goes on to point out that the same type of result is obtained when considering the diffraction pattern produced by random spheres located in one plane which is perpendicular to the forward direction of the forward beam. Thus, if human blood corpuscles are placed on a glass slide and the Fraunhofer diffraction pattern studied, a series of concentric rings typical of the diffraction pattern of the single spheres is obtained (ref. 6). In fact, the structure of the rings is sufficiently well defined that the average diameter of the blood corpuscles can be deduced from from the dimensions of the diffraction rings. A clinical device



Randomly Distributed Identical Apertures



Schematic Representation of Light-Regions in the Fraunhofer Diffraction Patterns of a Rectangular Aperture

Figure 11

DIFFRACTION BY RECTANGULAR APERTURES

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which makes use of this phenomena is called Young's Eriometer. Thus, a random array of uniform spheres confined to a single plane should have a Fraunhofer diffraction pattern the same as for a single sphere (ref. 7).

Now Mie theory represents a general solution for the scattering properties of a single sphere. When the sphere is very small compared to the wavelength of light, the scattering is symmetrical in front of and behind the particle. This type of scattering is known as Rayleigh scattering. Although Mie theory covers all scattering phenomena, the term Mie scattering is usually restricted to the description of scattering by particles in the size range 0.25λ up to 10λ . In fact, the Mie scattering is the growing tendency towards forwards scattering with increasing particle size (ref. 8). The boundary between so-called classical diffraction theory and the restricted Mie theory is not well defined and in one sense the pattern of scattered light produced by particles in the range 0.25λ to 10λ can still be called a diffraction pattern. Now the Mie theory is based upon two explicit conditions. First, the incident beam is a plane parallel beam of noncoherent monochromatic light, and secondly, the radiation detector used to explore the scattering pattern is at infinity, i.e., is at a distance large compared to the wavelength of light. In a sense, therefore, Mie theory corresponds to Fraunhofer diffraction. Now experiments have been carried out using a thin (1 or 2 particle thick)

array of pigment particles using a spectrophotometer and monochromatic radiation (3). Optical equipment of this kind satisfies the condition of plane parallel incident radiation with the detector at a large distance. It is not surprising that these experiments have found maximum scattering power for a given particle size at wavelengths predicted from the Mie theory, since it is a general principle that the diffraction pattern produced by a thin random array of identical particles, perpendicular to the beam, is the same as for a single particle. To conclude from this type of observation that the scattering power of the pigment in a paint film is optimum at this size is to attempt to extrapolate from monochromatic single scattering from a plane parallel beam studied at infinity, to the behavior in diffuse polychromatic light studied at a distance of two or three wavelengths, i.e., from the next pigment particle. This is no more logical than trying to predict the behavior of a series of screens as shown in Figure 10, when placed in diffuse white light at separations of a few wavelengths from Fraunhofer diffraction patterns of a single slit.

We suggested in an earlier report (ref. 8) that a three layer system may prove to be an efficient paint surface for the prevention of white light penetration. This was based on reasoning which extrapolated Mie theory results to the complex interactions occurring in a paint film. If diffuse light radiation is falling on the particle from all directions, then any

diffraction pattern rotated through 360° would give the same resultant pattern. Therefore, with diffuse radiation, it is the fact that the radiation interacts with the surface which is important not the specific diffraction pattern for a single particle in a specific direction. Should a three-layer system prove to have important properties, the properties of the system cannot be regarded as predictable from Mie theory. Having demonstrated the impracticability of pursuing Mie theory as having significance for paint theory and practice we will next study the significance of formulae which have been applied successfully to the properties of paint films.

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