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by

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ANGULAR SCATTERING FROM IRREGULARLY SHAPED PARTICLES WITH APPLICATION TO ASTRONOMY

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1. INTRODUCTION

Small, solid particles play an important role in current theories of star formation (Burbidge and Burbidge, 1958) and the origin of the solar system (Urey, 1957). It also appears that the accumulation of the planets can only be explained by the coagulation of small particles whose shapes are irregular rather than spherical (Donn and Sears, 1962).

Some information concerning the mass and momentum of particles near the earth has recently been obtained from rockets, satellites and probes (Alexander *et al.*, 1962). Information about the composition and size of dust particles collected on earth is summarized by Fireman and Kistner (1961), but the relation of these particles to cosmic grains is not clearly established.

Until rocket techniques become more sophisticated, most of our knowledge concerning the specific nature of the particulate matter in space will be based upon observations of the angular scattering from the zodiacal light particles, comet grains, and the *F* corona of the sun; and upon the extinction characteristics of the interstellar medium.

The usual procedure in astronomy is to compare observed scattering data with theoretical computations for a distribution of spheres in order to obtain three characteristics of the particle cloud: (1) effective size and size distribution; (2) index of refraction and (3) space density (van de Hulst, 1949; Liller, 1960). That distribution of spheres showing the best fit to the observed data is adopted as representing the scattering material.

Three major uncertainties affect the conclusions. These are: (1) the assumption of spherical scatterers; (2) the incompleteness of astronomical observations, i.e. small range of angle and wavelength and (3) the incompleteness of scattering calculations for a large range of spherical particle sizes and indices of refraction. The analysis is further handicapped by the labor necessary to carry out a large number of comparisons for many different size distributions.

In order to alleviate the incompleteness of the present tables for spheres

and reduce the labor in making comparisons with many size distributions, we have prepared a table of scattering intensities and an integrated function $F(\alpha, \theta, m)$ for the following intervals in the index of refraction, the scattering angle measured from the forward direction, and the size parameter:

$$\begin{aligned} m &= 1.2(0.2)2.4 \\ \theta &= 0^\circ(5^\circ)180^\circ \\ \alpha &= 2\pi r/\lambda = 0.1(0.1)100 \end{aligned}$$

The integrated function is given by:

$$F(\alpha, \theta, m) = \int_{0.1}^{\alpha} I(\alpha', \theta, m) d\alpha' \quad (1)$$

This function permits quick and simple comparison of the scattering from many size distributions. The method will be discussed further in this paper and in more detail elsewhere.

Our primary objective in this investigation is to study the scattering properties of irregularly shaped particles in order to evaluate the consequences of comparing the observed data with spheres and, thereby, gain a better understanding of which scattering parameters are most definitive from the viewpoint of gathering optimal astronomical data.

Typical results concerning the scattering from irregularly shaped zinc oxide particles are presented and compared with equivalent theoretical results for spheres.

2. FORMULATION OF THE PROBLEM

There are several reasons for suspecting that the particles in space are non-spherical in shape. First, light scattered by interstellar dust is polarized in the forward direction. Aligned, asymmetric grains would polarize in this manner. Secondly, irregular particles, which may be related to the cosmic grains, have been collected at high altitudes (Hemenway *et al.*, 1961a, b; Fireman and Kistner, 1961). Thirdly, any zodiacal particles produced by fragmentation in the asteroid belt (Pietrowski, 1953; Fessenkov, 1959) will be angular—not spherical. Finally, experimental and theoretical research on crystal growth (Donn and Sears, 1962) indicates that crystals forming in space would tend to resemble plates, needles or snowflakes rather than spheroids. Cometary grains are expected to fall into this category.

For our preliminary measurements we picked an irregular particle which was easy to grow, whose shape was always the same, with a high, real index of refraction, and whose size was easy to control. Zinc oxide crystals fulfill these requirements. Also, zinc oxide crystals can easily be made to cluster in loose aggregate form so that scattering from porous particles may be studied. An electron micrograph of particles collected from one of our scattering samples is shown in Fig. 1.

3. EXPERIMENTAL ARRANGEMENT

The zinc oxide particles are grown by vaporizing zinc in a mixture of dry nitrogen and oxygen. The size of the particles is controlled by adjusting the temperature of the zinc, the ratio of nitrogen to oxygen, and the flow rates.

The resulting zinc oxide smoke is diluted and time is allowed for the larger particles to settle out. The resulting size distribution is shown in Fig. 2.

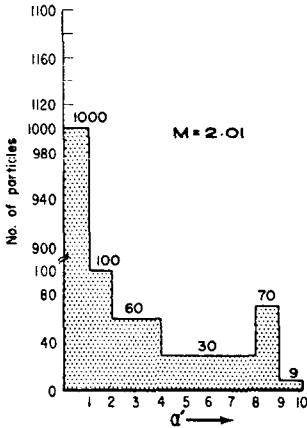


Fig. 2 - observed size distribution of zinc oxide particles.

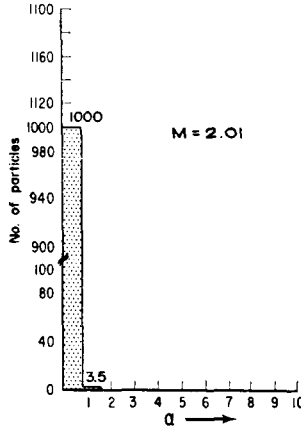


Fig. 3a - computed size distribution for equivalent spheres.

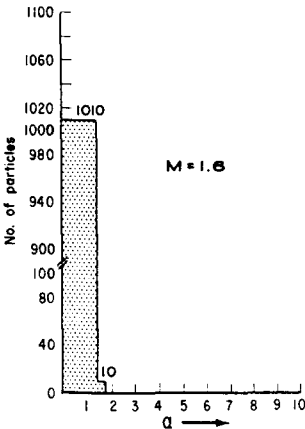


Fig. 3b - computed size distribution for equivalent spheres.

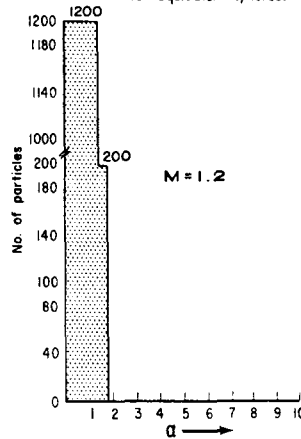


Fig. 3c - computed size distribution for equivalent spheres.

After the smoke has become relatively stable, it is irradiated with a beam of unpolarized mercury green or mercury blue light ($\lambda 4360, \lambda 5468$). Angular scattering for both polarizations is recorded from $\theta = 150^\circ$ to $\theta = 50^\circ$.

The smoke density is monitored during the entire measurement process. After a stable run the smoke is sampled with a standard Cascela thermal precipitator modified to accept electron microscope grids. The size distribution is then counted from the electron micrographs. A typical scattering diagram for the size distribution of Fig. 2 is shown in Fig. 4.

4. THEORETICAL ANALYSIS OF ZINC OXIDE SCATTERING DIAGRAM

The observed scattering diagram shown in Fig. 4 was compared to an enormous number of scattering diagrams for spheres in various size distributions by using the integrated function of equation (1). Any distribution of sizes may be approximated by a horizontal or vertical histogram. The scattering which corresponds to each interval, $\Delta\alpha = \alpha_i - \alpha_{i-1}$, in the histogram is then given by $F(\alpha_i) - F(\alpha_{i-1})$ with an appropriate weighting factor. In general, then, the scattering from any size distribution, p , is given by:

$$I(p, \theta, m) = \sum_{i=0}^n A_i [F(\alpha_i) - F(\alpha_{i-1})] \quad (2)$$

Each interval, $\alpha_i - \alpha_{i-1}$, may usually be taken quite large because the scattering is relatively insensitive to changes in the shape of continuous distributions except at the extreme forward and backward scattering angles.

In practice, one can obtain a closest fit to any scattering diagram by first referring to the scattering diagrams of, say, fifty basic functions $F(\alpha_i)$ where α_i is taken from very small values to arbitrarily large ones in equal steps. Each basic function exhibits its own peculiar characteristics. For instance, $F(2)$ may exhibit a polarization ratio $I_1/(I_1 + I_2) < 1/2$ whereas $F(4)$ may exhibit $I_1/(I_1 + I_2) > 1/2$. On the basis of these peculiar characteristics most of the basic functions may be discarded from further consideration. Those which remain may then be weighted and combined by trial and error until the desired scattering diagram is constructed. With a little experience one can accomplish this quite easily and, for most distributions, be assured of the uniqueness of the fit.

There are, of course, several different criteria available by which one judges which distribution of spheres most nearly fits the observed scattering diagram.

One choice is to find a distribution of spheres which reproduces only one scattering parameter; for instance, the vertical component of the intensity. Another distribution can then be found which reproduces the horizontal component and so on until one has several different distributions of spheres, each of which accurately portrays one of the characteristic scattering parameters. Among the scattering parameters which may be used in this manner are: (1) slope of vertical component as a function of angle; (2) same for horizontal component; (3) polarization ratio as a function of angle; (4) ratio of forward

to back scattering or ratio at any fixed angles; (5) ratio of maximum to minimum scattering over some angular interval. (All of these should be considered as a function of wavelength as well.) The differences between the resulting set of "equivalent" sphere distributions may then be used to characterize the scattering from a particular irregular particle. Various different

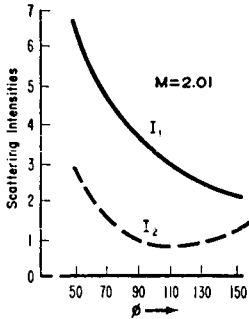


Fig. 4—scattering observed from zinc oxide particles.

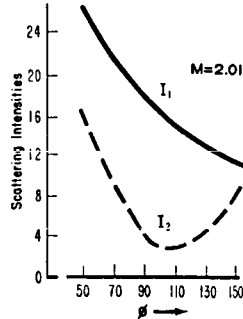


Fig. 5a—scattering computed for "equivalent" spheres.

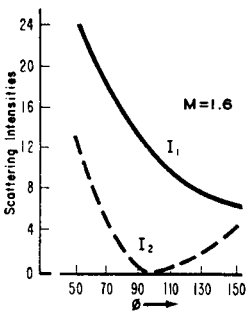


Fig. 5b—scattering computed for "equivalent" spheres.

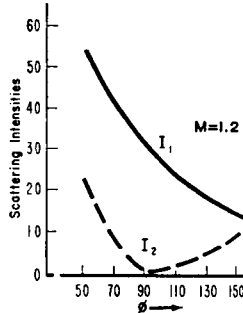


Fig. 5c—scattering computed for "equivalent" spheres.

irregular particles can then be catalogued by comparing the equivalent sphere sets corresponding to each. Under favorable conditions one can then deduce the general shape and refractive index of a particle by observing the relative dominance of the different scattering parameters. We are in the process of investigating the usefulness of this procedure.

For this paper we have chosen a second criterion by which to judge which distribution of spheres is equivalent to the zinc oxide distribution. By using the integrated function we have found a single and unique distribution of spheres for each index of refraction which roughly reproduces the entire zinc oxide scattering diagram. The equivalent scattering diagrams defined on this

basis are shown in Fig. 5a, b, c and the corresponding equivalent size distributions are shown in Fig. 3a, b, c.

5. RESULTS

Using the second criterion mentioned above, we found it impossible to match all parameters exactly with one set of "equivalent" spheres. The general shape of the vertical and horizontal component curves is most nearly reproduced by spheres with index 1.2. The shape of the equivalent horizontal component becomes more distorted as we go to higher indices.

The polarization ratios are shown in Table 1.

TABLE 1. POLARIZATION RATIOS $I_1/(I_1 + I_2)$

θ	Computed (spheres)			Observed (ZnO)
	$m = 1.2$	$m = 1.6$	$m = 2.01$	$m = 2.01$
150°	0.58	0.61	0.57	0.62
90°	1	0.96	0.86	0.81
50°	0.69	0.66	0.62	0.74
Average	0.76	0.74	0.68	0.72

The spheres with $m = 1.6$ match the average observed polarization ratios a little better than the other distributions but only the spheres for $m = 2.01$ match at 90°. The relative slope characteristic defined by:

$$R(\theta) = [I_1(\theta) - I_2(\theta)]/[I_1(150^\circ) - I_2(150^\circ)]$$

is matched most closely by the spheres with $m = 2.01$ as can be seen in Table 2.

TABLE 2. RELATIVE SLOPE CHARACTERISTIC

θ	Computed (spheres)			Observed (ZnO)
	$m = 1.2$	$m = 1.6$	$m = 2.01$	$m = 2.01$
50°	8.5	4.9	3.9	5.4
90°	8.2	5.8	5.4	3.8
Average	8.35	5.35	4.65	4.6

The shape of the vertical component is matched almost equally well by all the distributions. One can, of course, continue to define new characteristics

for purposes of matching, and thus give some sort of rough index of fit to each computed curve. For the present, however, let us just point out the spheres with $m = 1.2$ most nearly approximate the actual size distribution since they give the largest proportion of big particles. Nevertheless, none of the "equivalent" distributions defined by our second criterion show any evidence of particles bigger than $\alpha = 1.8$, whereas 15 per cent of the particles in the observed zinc oxide distribution are larger than $\alpha' = 1.8$.

The size parameter, α' , for the zinc oxide particle was arbitrarily computed using the length, L , between the tips of two spikes. It is, therefore, not surprising that the observed size distribution is different than the deduced ones. In order to obtain a distribution function which roughly approximates those computed for the equivalent spheres, the size parameter defined by $\alpha' = 2\pi L/\lambda$ must be multiplied by a scaling factor β . Then

$$\alpha = \beta\alpha' = \frac{2\pi\beta L}{\lambda}$$

can be compared directly to the size parameter defined for spheres i.e.

$$\alpha = \frac{2\pi r}{\lambda}$$

and the equivalent zinc oxide radius is then given by:

$$r = \beta L$$

The values for β are given in Table 3.

TABLE 3

m	r	which corresponds to:
1.2	0.25L	$\frac{1}{2}$ the length of one spike
1.6	0.16L	$\frac{2}{3}$ the length of one spike
2.01	0.018L	roughly the diameter of the central junction

This is only an estimate of the effective size parameter because we do not know the accuracy of our observed size distribution as counted from the electron micrographs.

Nevertheless, it appears probable that irregular zinc oxide particles exhibit scattering characteristics like those of much smaller spheres of high index of refraction or like only somewhat smaller spheres of low index. Apparently, the "equivalent" spheres are always smaller than the longest length of the irregular particle for indices of refraction lower than, or equal to, that of the irregular particle.

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