Particle characteristics from light scattering measurements[1]

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(Manuscript received February 2, 1968; revised version July 5, 1968)

ABSTRACT

The effects of the physical characteristics of particles that influence light scattering have been examined. Measurements were made of the polarization and angular distribution of 6328 Å He-Ne laser light scattered by various latex-sphere hydrosols. Some measurements were compared with Mie theory calculations, and additional calculations were made to determine the effect on scattering of variation in particle size and index of refraction. Laser and incoherent light scattering patterns are the same. Measurements are also given showing the nature of polarization of the scattered light for various types of incident polarization. Results of this work indicate that laser light scattering techniques can be used to obtain useful information on the properties of atmospheric aerosols.

Particle characteristics from light scattering measurements

Measurement of light scattering is a useful technique for determining physical properties of particles in the atmosphere (Hodkinson, 1966, Dobbins, 1967 and Bullrich 1964). The natural aerosols, when their light-scattering pattern is observed, show a complex spectrum of effects that depend on their wide variation in index of refraction, size, shape, and relative size concentration. The long-range objective of the work reported here is to determine size and concentration of remote aerosols by means of coherent light sources. Because atmospheric aerosols are highly complex, the initial phase of the work consisted of a study of controlled concentrations of spherical latex particles suspended in water.

The intensity, angular distribution and polarization of the scattered light are influenced by several physical parameters. The physical factors determining the light scattering pattern are as follows:

1. The diameter (d) of the particle (the diameter is usually combined with the wavelength in the size parameter ratio $\alpha = \pi d/\lambda$)
2. Particle shape
3. The material's index of refraction (m), which may vary with a layer or shell of differing material, or which may be complex if there is absorption
4. The total number of particles in the scattering volume
5. The size (or other parameter) distribution of the particles

In the following discussion, the scattering particles were assumed to be uniform spheres with real index of refraction. The scattering parameters that were varied include size, index of refraction, concentration, size distribution, and coherent or incoherent source illumination. Theoretical calculations based on Mie theory were used along with experimental results.

The best available controlled suspensions of monodispersed (all same size) and polydispersed (mixture of many sizes) latex spheres in water were studied with conventional and laser light sources. Some results and a description of the apparatus have been previously reported (Harris, Sherman, and Morse, 1967). Every effort was made to keep the system simple and free from extraneous light and electrical noise, and to make the scatterers as uniform as possible.

Fig. 1 shows scattering intensity as a function of the angle of deviation from the incident light direction. Both the parallel polarized light (scattered with polarization parallel to the plane of scattering) and perpendicular polarized

[1] Presented at International Association of Meteorology and Atmosphere Physics. Lucerne 5 Oct. 1967.

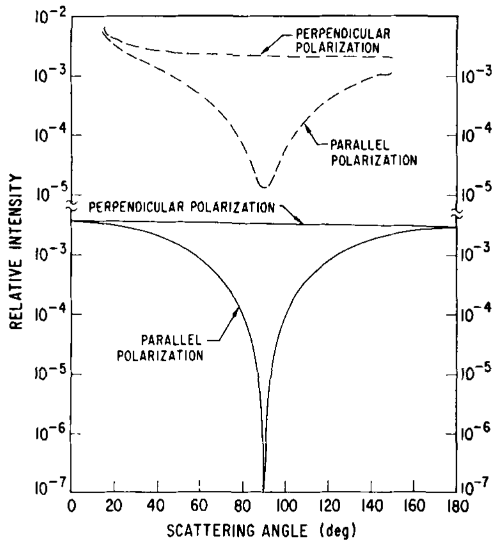

Fig. 1. Angular scattering diagram for 0.088 μ nominal diameter Dow latex particles. Dashed curves indicate experimental data; solid curves indicate calculated data.

light are shown as calculated from Mie theory and as measured for the smallest latex sphere available (0.088 μ diameter with size parameter $\alpha = 0.582$). This size is in the transition region where the simplified Rayleigh equation offers a good approximation to Mie theory. This more recently obtained sample from Dow Chemical Co. had a more uniform size distribution than the previous sample and gave marked improvement over our previously published results for this particle size.

Dow Chemical Co., the manufacturer of the latexes, uses an electron microscope to measure the diameters of the spheres. The diameters measured by electron microscope at Dow were slightly larger than those obtained by light scattering experiments. Though different wavelengths were used, our measurements by light scattering techniques agreed with those of Kratohvil and Smart (1965) on particles of the same nominal size. In Figs. 2 and 3, the scattering curves as measured for a particle of nominal diameter 1.305 μ (Dow) or $\alpha = 8.623$ are compared with Mie theory calculations for size parameters α from 8 to 9. The experimental curve is inserted in what is probably the right position in the theoretical sequence of curves, but a minimum mean square analysis was not performed. The apparent size can be determined to about 1% by this comparison. The Dow value is about 3% higher than our measurements. The curve shows the size precision of the scattering technique when the spheres are very nearly monodispersed. The actual small distribution of sizes and other effects have the result of smoothing the maxima and minima fluctuations (Kratohvil and Smart, 1965).

The angular distribution of scattered intensity depends on the size parameter and index of refraction. For a very small particle, the distribution is almost that given by Rayleigh, as shown in Fig. 1, but as the size increases a large number of maxima and minima are produced and more and more of the total scattered intensity remains close to the original direction of propagation. Computed values for α's of

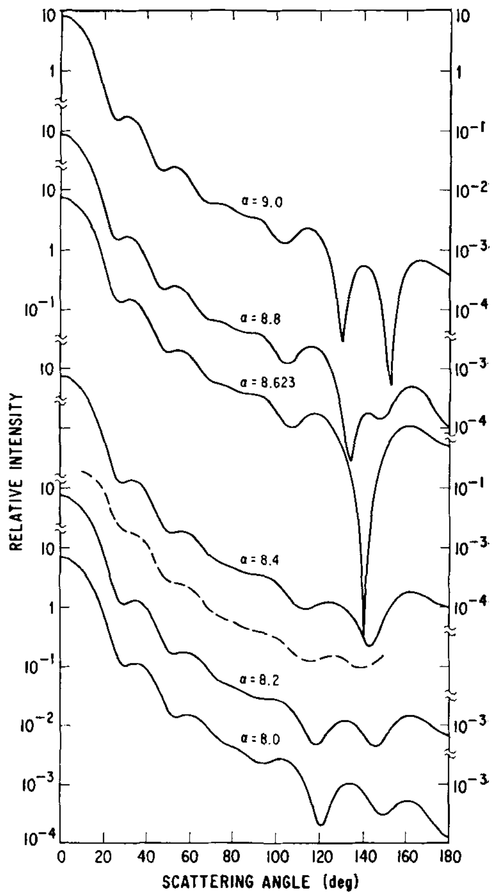

Fig. 2. Angular scattering diagram for 1.305 μ nominal diameter ($\alpha = 8.623$) Dow latex particles (parallel polarization). Dashed curve indicates experimental value; solid curves indicate calculated values for various α.

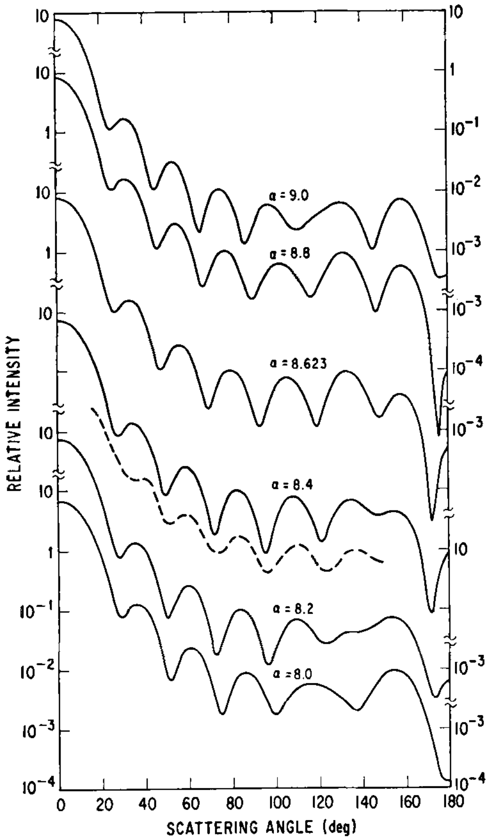

Fig. 3. Angular scattering diagram for 1.305 μ nominal diameter (α = 8.623) Dow latex particles (perpendicular polarization). Dashed curve indicates experimental value; solid curves indicate calculated values for various α.

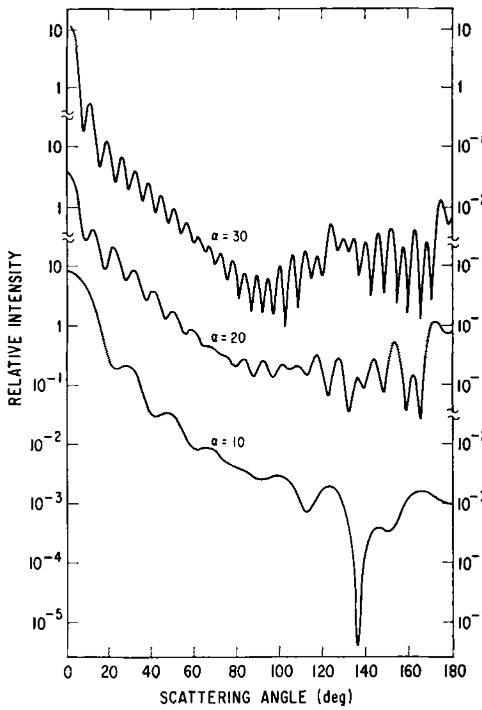

Fig. 4. Angular scattering diagram for $m = 1.20$ for three calculated values of α (parallel polarization).

10, 20, and 30 are represented in Figs. 4 and 5. In a mixture, there is superposition of the appropriate pattern due to each individual particle. Figs. 6, 7, 8, and 9, computed from Mie theory, show the effect of a variation in index of refraction from 1.2 to 1.6 for a non-absorbing material for two different size parameters $\alpha = 4.0784$ and 6.2275. Increasing the index of refraction moves the maxima closer together in a somewhat similar manner to increasing the size.

An initial question was whether there would be a difference in scattering behavior between

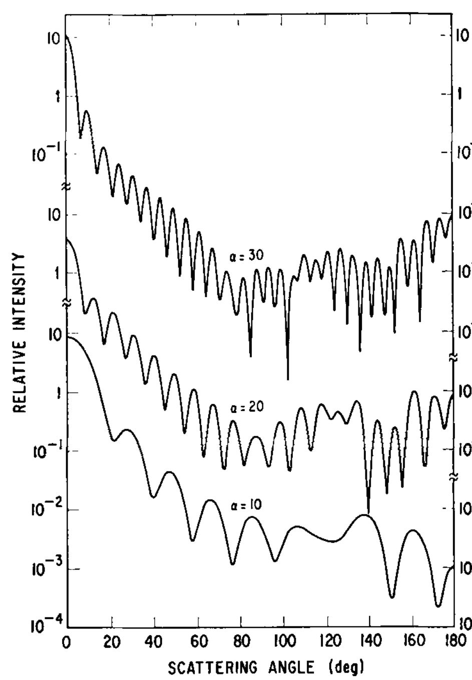

Fig. 5. Angular scattering diagram for $m = 1.20$ for three calculated values of α (perpendicular polarization).

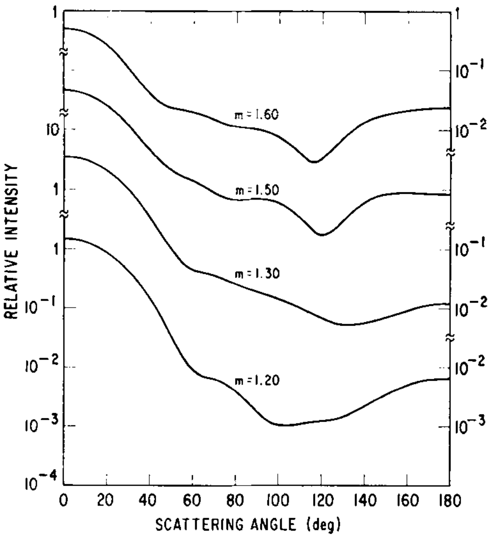

Fig. 6. Angular scattering diagram for $\alpha = 4.0784$ for four calculated values of m (parallel polarization).

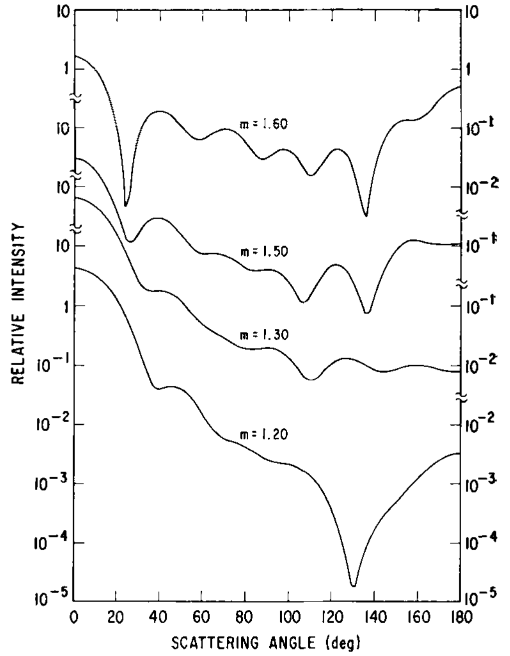

Fig. 8. Angular scattering diagram for $\alpha = 6.2275$ for four calculated values of m (parallel polarization).

conventional incoherent light and coherent laser light sources (Carrier and Nugent, 1965). Experimental work (Harris, Sherman, and Morse, 1967) indicates that there is no source effect under the experimental conditions examined. Coherent light at 6328 Å from a He-Ne laser and incoherent light from a high-pressure xenon lamp centered in a 100 Å band at the

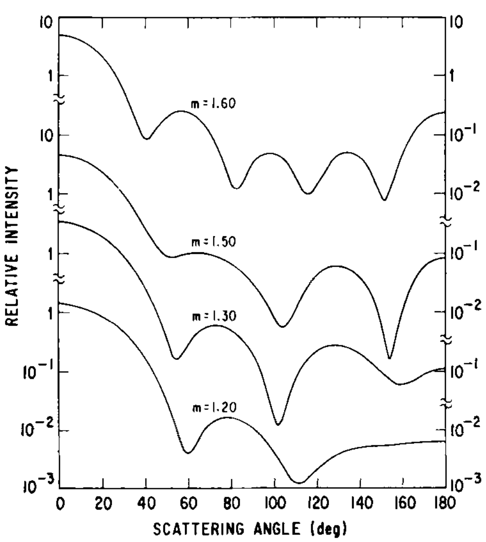

Fig. 7. Angular scattering diagram for $\alpha = 4.0784$ for four calculated values of m (perpendicular polarization).

same wavelength were compared by scattering from hydrosol targets of latex spheres. The results indicate that aerosols should scatter independently of the coherence of the light source. This is true also for concentrations of particles with multiple scattering and also for a mixture of sizes (Sherman, Harris, and Morse, 1968). Since the size and concentrations studied are not exceeded in the atmosphere, no coherence of light effect should be expected there either. Recent experimental work on artificial fogs has given the same result (Reisman, Cumming, and Bartky, 1967 and Zuev, in press).

The variation of light intensity with angle, polarization, and wavelength increases the possibilities of discrimination, as shown by calculations and actual atmosphere aerosol measurements (Bullrich 1964, Deirmendjian 1964, Eiden 1966, Hodkinson 1966, Holland and Draper 1967). Linear polarizations have been commonly used, but other types of polarization might be even more useful. The incident light may be "diagonally" polarized (linear at 45° to the plane of scattering). The scattered component of incident diagonally polarized light can be analyzed for a polarized component

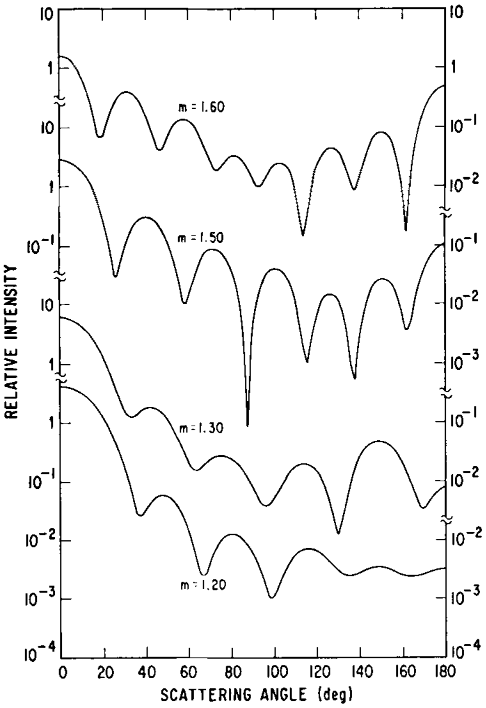

Fig. 9. Angular scattering diagram for $\alpha = 6.2275$ for four calculated values of m (perpendicular polarization).

in any plane, e.g., the same plane as the incident light or at 90° to the original plane of polarization. Incident righthand circularly polarized light, when scattered, may contain some left-hand circularly polarized light that can be detected by using a "crossed" analyzer. Pritchard and Elliott (1960) made some initial studies of polarization in the atmosphere using a conventional light source. Their results indicated, as do ours, that an initial diagonal polarization can be represented by the sum of equal components of parallel and perpendicular polarizations and the scattering of each component treated independently. Furthermore, when only single scattering from 1.305 μ spheres occurred with incident linearly polarized light, the scattered light examined for the same polarization as the incident light was 10^3 times the intensity from crossed polarizers; hence depolarization was negligible. If the particles are not spherical or the material is not isotropic, some depolarization can be expected. Fig. 10 shows scattered light measurements for

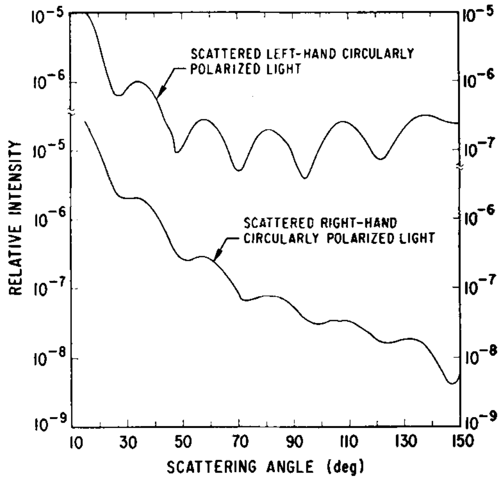

Fig. 10. Experimental angular scattering distribution for incident right-hand circularly polarized incident light.

the same (as the incident) circularly polarized and crossed (with the incident) circularly polarized components for the 1.305 μ particles. Linear polarization measurements for a polydispersed system of 6 to 14 μ ($\alpha = 40 - 92$) are shown in Fig. 11. Circular polarization produced with a Glan–Thompson prism and a Fresnel rhomb gives the curves shown in Fig. 12. Circular polarization when directly reflected changes sense (left $\leftrightarrow$ right); thus, in Fig. 12, "crossed" means that scattered light from incident right-hand polarized light is analyzed for left-hand polarization. Fig. 13 shows the

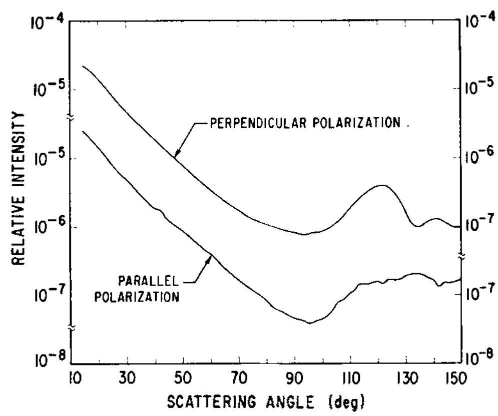

Fig. 11. Experimental angular scattering distribution for linearly polarized light for polydispersed 6 to 14 μ Dow latex particles, $\alpha = 40$ to 92.

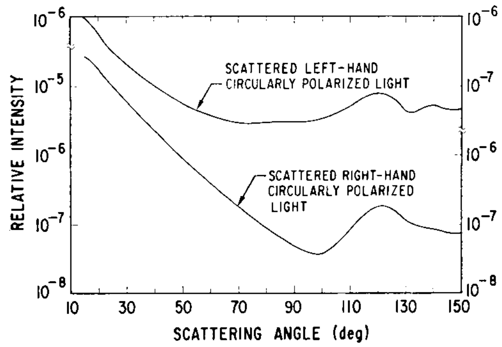

Fig. 12. Experimental angular scattering distribution for right-hand circularly polarized incident light for polydispersed 6 to 14 μ Dow latex particles, $\alpha = 40$ to 92.

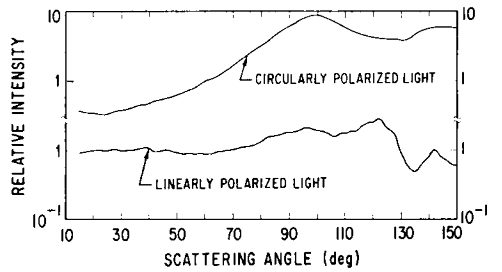

Fig. 13. Angular variation of polarization ratios for 6 to 14 μ Dow latex particles, $\alpha = 40$ to 92, for linear case (perpendicular-to-parallel) and for circular case (left-hand-to-right-hand) for incident right-hand polarized light.

polarization ratio for linear polarization, perpendicular polarization divided by parallel polarization, and the polarization ratio for circular polarization ("crossed" analyzer divided by "same" analyzer).

These results indicate an application of scattering techniques to the atmosphere. The laser light source, with its high intensity, polarization, and coherence, offers a powerful new tool which should greatly increase the useful measurements to add to those so far made with conventional sources (Elterman, 1966). The advantages of the laser source are well illustrated by our laboratory experiments. The best xenon source, after collimation, polarization, and filtering for a 100 Å band at the 6328 Å wavelength, gave the same intensity as obtained when the 1-mW He-Ne laser was operated at 1/30 mW. The laser nephelometer has better sensitivity than nephelometers using conventional sources. However, even with the improved source, there is still at present a problem in remote atmospheric studies in getting sufficient light signal return. Space and time variations of the natural aerosols in concentration, size distribution, index of refraction, shape, and material further complicate the problem. When scattering data is obtained, there still remains the very difficult inversion problem of going from scattered light distributions to the aerosol size distribution. This has not been solved in the general case (Shifrin and Perelman, 1966 and 1967, Shifrin and Chayanoya, 1966, and Twomey and Howell, 1967). The aerosol size distribution as determined by scattering has to be evaluated critically and standardized by computations and comparison with methods that measure the size of the particles individually. Single particle optical detection even has its problems (Podzimek, 1962 and Bricard, 1966).

Laboratory experiments with hydrosols and computations using Mie theory show the laser to be an excellent source for light scattering experiments. Laser light scattering measurements can give us much new information about the material, shape, sizes, number, and size distribution of aerosols, and about the aerosol variation in position and time in the atmosphere. A semi-empirical approach to the aerosol scattering problem using more powerful multiwavelength lasers, good optics and electronics, and computers should give us much valuable information on the atmosphere.

Acknowledgment

Grateful acknowledgment is due to George C. Sherman and Frank L. Morse, Jr. for assistence with all phases of this work.

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СВОЙСТВА ЧАСТИЦ ПО ИЗМЕРЕНИЯМ РАССЕЯНИЯ СВЕТА

Исследуется влияние физических характеристик частиц на рассеяние света. Проведены измерения поляризации и углового распределения излучения He–Ne лазера (6328 Å), рассеянного различными сферическими частицами гидрозоля латекса. Некоторые результаты измерений сравнивались с вычислениями по теории Ми; проведены дополнительные расчеты для выявления зависимости результатов рассеяния от вариаций размеров частиц и показателя преломления. Обнаружено, что диаграммы рассеяния лазерного и некогерентного излучения подобны. Описаны также опыты по определению состояния поляризации рассеянного света для различных типов поляризации падающего излучения. Результаты работы показывают, что эксперименты по рассеянию лазерного излучения можно использовать для получения полезной информации о свойствах атмосферного аэрозоля.