Reports

Precision Measurement of Particulates by Light Scattering at Optical Resonance

Abstract. *The efficiency of light scattering from individual aerosol spheres is markedly enhanced for certain combinations of the particle size and refractive index with the incident wavelength. Differential scattering patterns observed at these "optical resonances" are so sensitive to particle characteristics that relative optical diameters of an approximately 3-micrometer sphere can be determined to within about 2 angstroms (0.007 percent). Such resonances for 1- to 30-micrometer spheres, cylinders, and regular nonspherical particles should be generally accessible with tunable lasers.*

When a plane wave of light strikes a particle, energy can be removed by scattering and by absorption. The efficiency Q of light extraction from the incident beam is the ratio between the total flux removed and the flux falling on the geometric area of the particle (*1*). Peaks, or "optical resonances," occur in Q for certain combinations of particle size, refractive index, and incident wavelength (*1, 2*).

Examples for nonabsorbing spheres are shown for several values of refractive index m in Fig. 1. The plots of Q against $\alpha = \pi d/\lambda$ are equivalent to plots of Q against particle diameter d for fixed incident wavelength λ. The first resonance encountered with increasing size for spheres with refractive index $m = 9$ (that is, water droplets scattering long-wavelength radiation) shows remarkably high Q (*2, 3*). As m decreases, the "first" resonance is reduced in both magnitude and sharpness and moves to larger values of α. The resonances shown for $m = 1.581$ correspond to polyvinyltoluene (PVT) spheres scattering from a He-Ne laser. The first resonance has now moved out to $\alpha \approx 2$ and is indistinct. However, as α increases further, the resonances become extremely sharp. One of these resonances, having only a 15-Å half-width, is the subject of this report.

While the resonances in Q are well known to theorists (*4*), they appear to have eluded experimental characterization. In addition, description of the differential (360°) scattering patterns at these resonances seems limited to the comment of van de Hulst (*2*, p. 158), "The [360°] scattering diagram in the resonance region assumes many weird forms."

We have explored the latter issue using PVT aerosol particles. A resonance occurs for 632.8-nm light at $\alpha = 14.774$ ($d = 2.9760$ μm) with a half-width $\Delta\alpha = 0.0075$ ($\Delta d = 0.0015$ μm). We find that scattering

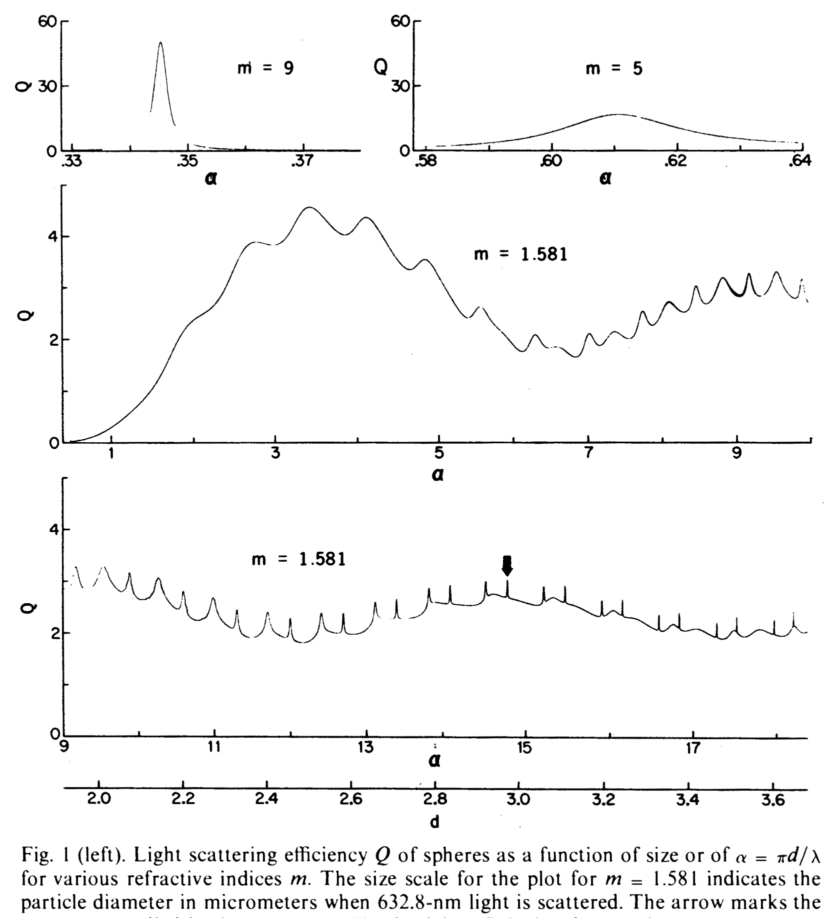

Fig. 1 (left). Light scattering efficiency Q of spheres as a function of size or of $\alpha = \pi d/\lambda$ for various refractive indices m. The size scale for the plot for $m = 1.581$ indicates the particle diameter in micrometers when 632.8-nm light is scattered. The arrow marks the resonance studied in this work.

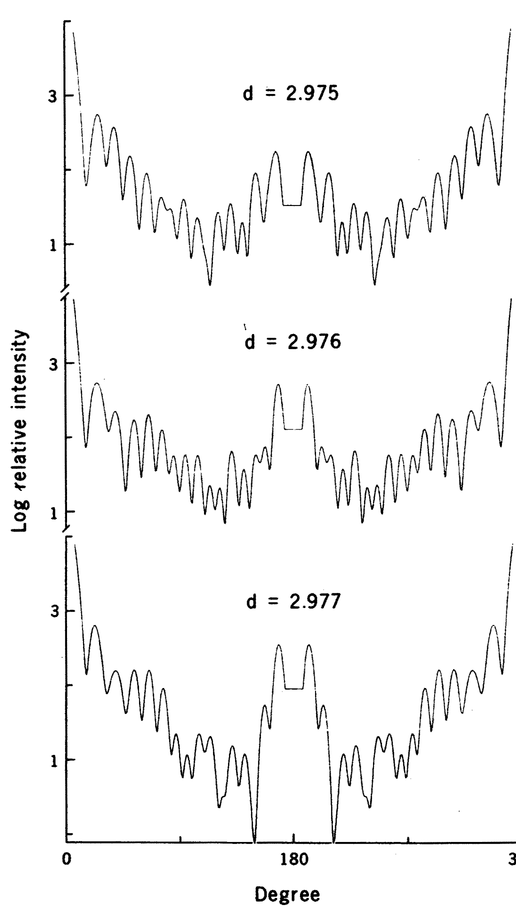

Fig. 2 (right). Calculated scattering patterns (5° resolution) for a sphere ~3 μm in diameter ($m = 1.581$), which almost exactly matches the resonance $\alpha = 14.774$ in Fig. 1. The spheres have assumed diameters differing by only 10 Å.

patterns are extremely responsive to size at sharp resonances. In fact, they are so sensitive that micrometer particles differing in diameter by a few angstroms can be distinguished.

Theoretical scattering patterns for three assumed diameters within this resonance are shown in Fig. 2. One of the diameters was chosen to match the resonance peak and the others are larger and smaller by 10 Å. Notwithstanding the small size difference, the theoretical patterns are easily distinguished by eye. Similar findings have been noted for other resonances occurring with $\alpha > 12$ in the $m = 1.581$ plot of Fig. 1.

The scattering patterns were computed using a Mie theory program with the refractive index set at the manufacturer's value, 1.581. The theoretical curves were degraded to 5° resolution and were blanked out ± 7° about 0° and 180° to match experimental conditions.

Scattering at resonance was recorded by the Gucker differential scattering photometer developed in this laboratory (*5, 6*). A schematic is given in Fig. 3. The instrument acquires in 20 msec a full 360° scattering pattern from a single particle in an aerosol flowing through a laser beam. With this instrument, hundreds of individual patterns can be recorded quickly. Comparisons with theoretical patterns establish the size and refractive index of each particle. When away from resonance, computer comparisons fix d and m for a particle in the range 1 to 3 μm to within 0.5 percent (*5–8*).

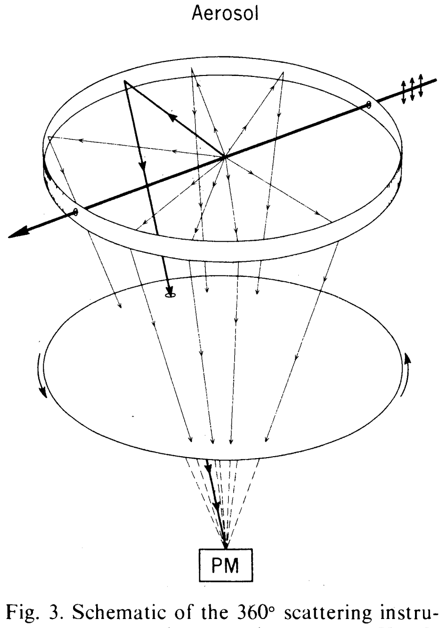

Fig. 3. Schematic of the 360° scattering instrument. An aerosol stream intercepts a He-Ne laser beam ("vertical" polarization) at one of two foci of an ellipsoidal mirror. Light scattered in the "horizontal" plane is intercepted by a segment of that mirror and directed to a photomultiplier (*PM*) at the second focus. A rotating aperture limits detection of the scattered light to a 5° segment. The aperture rotates at about 3000 rev/min so that a 360° scan is completed in 20 msec. The photomultiplier signal is fed through a logarithmic amplifier to a computer for storage and processing.

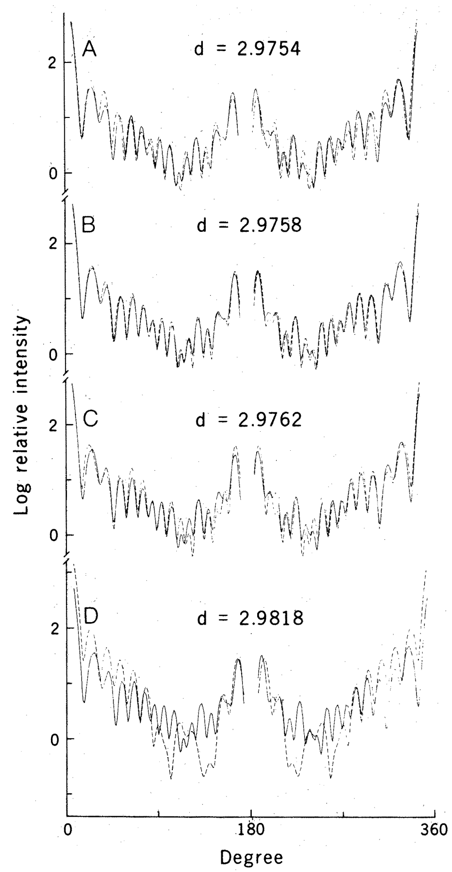

Fig. 4. (A to C) Comparisons of calculated scattering patterns (dashed curves) with the pattern observed from a particle whose diameter places it almost exactly at the resonance $\alpha = 14.774$. The best fit of calculated patterns is found to occur for an assumed $d = 2.9758$ μm (B). Assumed diameters 4 Å larger or smaller give poorer fits in these visual comparisons. (D) Comparison of the experimental pattern in A to C with a curve calculated for a particle whose diameter is 60 Å larger than the best-fit diameter.

The size dispersion of the PVT aerosol ($\bar{d} \approx 3$ μm) is large enough to encompass the resonance at $\alpha = 14.774$ (*9*). Among the hundreds of individual scattering patterns observed from this aerosol were a few generated by particles whose size falls within the resonance half-width. They were recognized by comparison of the calculated and observed scattering patterns. A pattern obtained from a particle which appears to be almost precisely at the resonance peak is compared in Fig. 4 with calculated patterns. The "best fit" $d = 2.9758$ μm was chosen by computer comparisons with patterns calculated for sizes differing by 0.0002-μm (2 Å) increments. This fit can also be found by visual inspection, but only when the trial fits differ by at least 4 Å.

The special character of resonance scattering diagrams is further displayed in Fig. 4, where the experimental diagram from the particle at resonance is compared with a pattern calculated for a size 0.006 μm (60 Å) larger than the optimum fit. While such increments are the minimum which can be used for visual discrimination when away from resonance, the two curves look almost unrelated in the resonance region.

Reproducibility is judged from the several scattering patterns which are routinely obtained from a particle while it crosses the laser beam. The second pattern obtained from the particle of Fig. 4 was also assigned $d = 2.9758$ μm by computer analysis. Thus, the signal-to-noise ratio and angular resolution in resonance scattering are sufficient to allow size discrimination to the order of a few angstroms (0.007 percent).

Figure 5 documents in another way the sensitivity of size measurements at resonance. We give plots of the least-squares deviations between observed and calculated scattering diagrams as a function of the particle diameter used in the calculation. These "goodness-of-fit" plots show how computer analysis can discriminate among trial size fits in a "resonance region" and also in a region away from optical resonance. The enhancement of size discrimination at resonance is evident when one notes that the size scale on the resonance plot is expanded by a factor of 30.

Two caveats must accompany this dis-

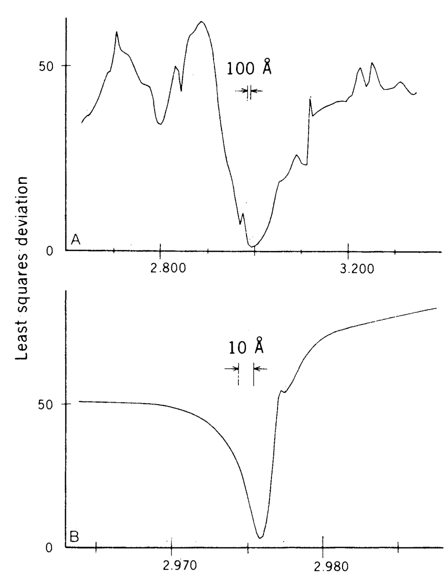

Fig. 5. Plots of the least-squares deviations of calculated scattering diagrams from experimental diagrams as a function of the particle diameter used in calculation. (A) Fits to the scattering diagram of a particle ($d = 3.022$ μm) which is away from an optical resonance. The minimum deviation is 1.04. (B) Fits to the scattering diagram of a particle ($d = 2.9758$ μm) which is almost coincident with the position of optical resonance. The minimum deviation is 2.97.

cussion. First, the remarkable precision of size determination at resonance is on a relative basis only. It presumes that m is fixed, and therefore any uncertainty in m degrades the absolute precision. In trials to determine both m and d simultaneously at resonance, the precision of each seems limited to about 0.5 percent. This is also what we find for nonresonant scattering. Second, the relationship of "optical" diameters determined at resonance to "physical" diameters is not entirely clear, particularly when relative precisions of a few angstroms are considered. In polystyrene aerosols ranging from about 1 to 3 μm, diameters determined by our optical method (away from resonances) match those from physical methods and electron microscopy to within a few percent (*7, 8*).

The sensitivity of scattering to particle parameters at resonance can be either a useful tool or a liability. It is clearly the latter if resonances within the range of aerosol parameters are not recognized. Techniques based on differential scattering in particular may be subject to substantial errors if one is not careful (*8*). On the other hand, the availability of tunable monochromatic light sources makes these resonances generally accessible for many systems. Since the effects of resonance occur for absorbing as well as nonabsorbing particles, and since they should be seen with cylindrical objects (*10*) as well as spheres, numerous applications are possible. The phenomenon might be used to detect extremely small changes in the dimensions of a scatterer due to externally applied stress or modifications to its environment, or to changes in its composition. The effect might also be used as a continuous monitor in the manufacturing of wires or optical fibers requiring a precise maintenance of the diameter or refractive index. It is also possible that resonant scattering may be a useful tool for studying regularly shaped nonspherical particles in biological systems.

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References and Notes

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9. The values $\bar{d}$ = 2.956 μm and m = 1.581 are given by the manufacturer (Dow Chemical Co., Midland, Michigan). The batch number is unknown.
10. W. A. Farone and M. Kerker [*J. Opt. Soc. Am.* **56**, 481 (1966)] and D. Cooke and M. Kerker [*Rev. Sci. Instrum.* **39**, 320 (1968)] have characterized nonresonance differential scattering from infinite cylinders. Diameters can be found with precisions better than one-half percent.
11. Contribution No. 2632 from the Chemical Laboratories of Indiana University. Supported by the National Science Foundation. C.S.P. appreciates the help of E. C. Hartman in the formative stages of this work

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Magnetotactic Bacteria

Abstract. *Bacteria with motility directed by the local geomagnetic field have been observed in marine sediments. These magnetotactic microorganisms possess flagella and contain novel structured particles, rich in iron, within intracytoplasmic membrane vesicles. Conceivably these particles impart to cells a magnetic moment. This could explain the observed migration of these organisms in fields as weak as 0.5 gauss.*

Few studies have unequivocally revealed effects of the earth's magnetic field on living organisms, although recent work indicates that birds (*1*) and elasmobranchs (*2*) detect and may use geomagnetism as a cue for orientation. I now describe a bacterial tactic response to magnetic fields, a phenomenon for which the term magnetotaxis is appropriate.

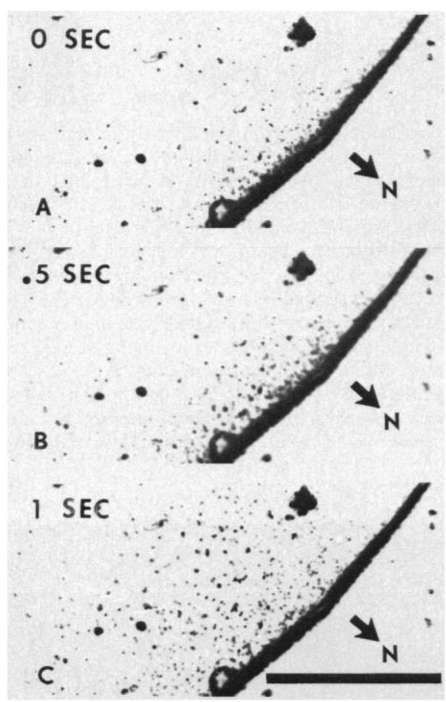

Fig. 1. Cinematographic sequence of bacteria displaying magnetotaxis. Portions of three sequential frames recorded on Kodak Tri-X reversal 16-mm movie film at 18 frames per second, with a Zeiss RA 38 microscope. The images shown were photographically reversed and enlarged. (A) Freely swimming magnetotactic bacteria aggregated at the northern extremity of a water drop by responding to geomagnetism. At the time of recording, a small permanent magnet was used to reverse the magnetic field. The cells then migrated in the opposite direction as recorded in frames (B) (0.5 second or 9 frames later) and (C) (recorded 1 second or 18 frames later). The arrow indicates the direction of the earth's north geomagnetic pole (bar, 100 μm).

During attempts to isolate *Spirochaeta plicatilis* from marine marsh muds (*3*), I observed microorganisms which rapidly migrated (*4*) toward one side of drops of the mud transferred to microscope slides (Fig. 1). I presumed this to be a phototactic response toward light from a northwest laboratory window. It became apparent, however, that light was not the stimulus directing the migration of these organisms as cells aggregated at the same side of mud drops regardless of the distribution of light on the slides, as well as in the dark. The direction in which these organisms moved immediately changed when small magnets were moved about in the vicinity of the microscope preparations. This suggested that geomagnetism was the stimulus for the behavior of the cells. It was experimentally confirmed that the migration of the bacteria was, indeed, directed by the earth's magnetic field (*5*).

Magnetotactic organisms were present in surface sediments collected from salt marshes of Cape Cod, Massachusetts, and in surface layers of sedimentary cores collected from a depth of 15 m in Buzzards Bay. Population densities in these environments ranged from 200 to 1000 cells per milliliter. Mud samples placed under several centimeters of seawater in glass jars and kept in dim light in the laboratory underwent an ecological succession (*3*). Populations of magnetotactic organisms increased to hundreds of thousands of cells per milliliter in many such mud samples after several months to several years of storage. Magnetotactic bacteria were evenly distributed in surface layers of the stored muds even though the jars were positioned for long periods in the same geographic orientation. Apparently factors additional to magnetotaxis also determine the distribution of the organisms in natural environments, since larger populations of magnetotactic bacteria were not detected in the northern areas of a marsh as compared to other locations in the marsh.

The organisms have not yet been iso-