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Demonstration of the Measurement of Light Scattering at an Angle of Zero Degrees from a Single Particle

FINAL REPORT

Period Covered by Report: 4 Months; June 1, 1991 to September 30, 1991

Edward S. Fry

Physics Department Texas A&M University October 23, 1991



U. S. Army Research Office ARO Proposal Number: P-29054-GS ARO Grant Number: DAA L03-91-G-0200

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A. STATEMENT OF THE PROBLEM STUDIED

An experimental demonstration of an innovative new idea that permits light scattering measurements at an angle of zero degrees was proposed. The idea is based on the use of coherent beam coupling in a nonlinear, photorefractive crystal of $BaTiO_3$ to separate the scattered light from the unscattered direct beam.

B. SUMMARY OF THE MOST IMPORTANT RESULTS

Measurements of light scattering by a glass fiber at an angle of 0° were successfully made at a wavelength of 0.5145 µm. Data were obtained as a function of fiber radii from 1 µm to 35 µm and are in excellent agreement with theory. This new measurement technique is based on the fanning of a coherent light beam in a photorefractive BaTiO₃ crystal. The use of fanning is a variant on the beam coupling technique originally proposed; and since it is a much more robust appreach, it is a significant additional accomplishment under this grant.

The results were recently submitted to Optics Letters and the paper was accepted for publication in early October. A preprint is attached.

C. LIST OF ALL PUBLICATIONS AND TECHNICAL REPORTS

- G. G. Padmabandu, Choonghoon Oh, and Edward S. Fry, "Measurement of Light Scattering at 0° by Micron Size Glass Fibers", submitted to Optics Letters, September, 1991; accepted October, 1991.
- G. G. Padmabandu, Choonghoon Oh, and Edward S. Fry, "Application of Beam Fanning in a Photorefractive BaTiO₃ Crystal; Measurement of Light Scattering at Zero Degrees by a Single Glass Fiber," in <u>Technical Digest on Photorefractive Materials. Effects.</u> <u>and Devices</u>, (Optical Society of America, Washington, D. C.) Vol. <u>14</u>, pp. 314-316 (1991).
- 3. Edward S. Fry, G. G. Padmabandu, and Choonghoon Oh, "Scattering At and Near 0° by Spheres and Glass Fibers", submitted to the Proceedings of the 1991 CRDEC Scientific Conference on Obscuration and Aerosol Research, October, 1991.

D. LIST OF ALL PRESENTATIONS

- Edward S. Fry, G. G. Padmabandu, and Choonghoon Oh, "Scattering At and Near 0° by Spheres and Glass Fibers", at the 1991 CRDEC Scientific Conference on Obscuration and Aerosol Research, Aberdeen, Md, June 24-27, 1991.
- G. G. Padmabandu, Choonghoon Oh, and Edward S. Fry, "Application of Beam Fanning in a Photorefractive BaTiO₃ Crystal; Measurement of Light Scattering at Zero Degrees by a Single Glass Fiber," at Photorefractive Materials, Effects, and Devices Topical Meeting, Beverly, Massachusetts, July 29-31, 1991.
- G. G. Padmabandu, Choonghoon Oh, and Edward S. Fry, "Measurement of Light Scattering at Zero Degrees by a Single Glass Fiber", at the LEOS Annual Meeting, San Jose, CA, Nov. 3-8, 1991.
- Choonghoon Oh, G. G. Padmabandu, and Edward S. Fry, "Measurement of the Forward Light Scattering from Polystyrene Spheres with a Barium Titanate Crystal", at the Optical Society of America Annual Meeting, San Jose, CA, Nov. 3-8, 1991.

E. LIST OF ALL PARTICIPATING SCIENTIFIC PERSONNEL SHOWING ANY ADVANCED DEGREES EARNED BY THEM WHILE EMPLOYED ON THE PROJECT

Edward S. Fry

G. G. Padmabandu

Shifang Li

Choonghoon Oh

No advanced degrees were earned during the 4 month duration

of this project.

F. REPORT OF INVENTIONS

None

THE VIEWS, OPINIONS, AND/OR FINDINGS CONTAINED IN THIS REPORT ARE THOSE OF THE AUTHOR AND SHOULD NOT BE CONSTRUED AS AN OFFICIAL DEPARTMENT OF THE ARMY POSITION, POLICY, OR DECISION, UNLESS SO DESIGNATED BY OTHER DOCUMENTATION.

Measurement of Light Scattering at 0° by Micron Size Quartz Fibers

G. G. Padmabandu, Choonghoon Oh and Edward S. Fry Department of Physics, Texas A & M University College Station, TX 77843-4242

ABSTRACT

Measurements of light scattering by a quartz fiber at an angle of zero degrees have been made using the 0.5145 μ m line from an Ar⁺ laser. Data have been obtained as a function of fiber radius from 1 μ m to 32 μ m and are in excellent agreement with theory. This new measurement technique is based on the fanning of a coherent light beam in a photorefractive BaTiO₃ crystal. The scattering of light at and near a scattering angle of 0° has several important aspects: (1) The scattering probability is so large that it significantly affects light propagation through dust, smoke, ocean water, etc.^[1-3] Consequently, it plays a critical role in imaging and LIDAR. (2) It can produce erroneous results in experimentally measured optical extinction coefficients and several algorithms have been used to overcome these difficulties.^[4, 5] (3) It is fundamentally related to the extinction via the optical theorem, but has never been explicitly checked.^[3, 6] (4) It provides a classification scheme for grouping particle shapes into six symmetry classes.^[7] (5) For a collection of identical particles in the same orientation, the 0° scattering is coherent. This produces an intensity at 0° that is proportional to the square of the number of particles.^[3] (6) Finally, it can provide a relatively accurate measure of particle size that is independent of the index of refraction.

The calculated results shown in Fig. 1 illustrate this latter point. Specifically, the scattering amplitude at 0° from a 30 μ m radius fiber is confined to a narrow range of values for all indices of refraction above approximately 1.1. Furthermore, this range has no overlap with that of, for example, a 20 μ m fiber. This radius discrimination is even more significant for spheres; 0° scattering characterizes a sphere radius to within ≈3% independent of any index of refraction above 1.1.

Despite its importance, light scattering in the forward direction has received very little experimental and theoretical attention. This work focuses on measurements of light scattering at 0° by a quartz fiber when it is illuminated at normal incidence.

For light scattering by a fiber at normal incidence, the relation between the incident and scattered field amplitudes is

$$\begin{pmatrix} \mathbf{E}_{||s} \\ \mathbf{E}_{\perp_{s}} \end{pmatrix} = e^{i3\pi/4} \sqrt{\frac{2}{\pi \mathbf{k} \mathbf{r}}} e^{i\mathbf{k} \mathbf{r}} \begin{pmatrix} \mathbf{S}_{1}(\theta) & \mathbf{0} \\ \mathbf{0} & \mathbf{S}_{2}(\theta) \end{pmatrix} \begin{pmatrix} \mathbf{E}_{||i} \\ \mathbf{E}_{\perp_{i}} \end{pmatrix}$$
(1)

where subscripts || and \perp refer to field components parallel and perpendicular, respectively, to the axis of the fiber; subscripts i and s indicate incident and scattered fields, respectively; and the scattering amplitudes $S_1(\theta)$ and $S_2(\theta)$ are complex functions of the scattering angle θ .^[3] The scattered intensities for the respective polarizations can be found by taking the absolute square of Eq. (1) and is given by

$$\begin{pmatrix} I_{II_{s}} \\ I_{\perp_{s}} \end{pmatrix} = \frac{2}{\pi kr} \begin{pmatrix} \left| S_{1}(\theta) \right|^{2} & 0 \\ 0 & \left| S_{2}(\theta) \right|^{2} \end{pmatrix} \begin{pmatrix} I_{II_{i}} \\ I_{\perp_{i}} \end{pmatrix}$$
(2).

In principle, both $|S_1(\theta)|$ and $|S_2(\theta)|$ can be obtained from intensity measurements with two linear polarizers.

Most experimental studies on light scattering by small particles are limited to angles from the near forward direction to the backscattering direction. Scattering at precisely 0° is manifested via a shift in phase. The factor limiting experimental measurements at $\theta = 0^{\circ}$ is the unscattered plane wave which is superimposed on the scattered spherical wave.^[8] Separation of the two waves is, in fact, not "impossible". However, it is difficult and there has been little previous experimental success.^[1, 2, 9]

Spinrad used a special low angle scattering meter to measure volume scattering functions down to angles as small as 0.1° from the forward direction.^[2, 10] He studied water suspensions of spheres and of two types of phytoplankton. *Amphidinium carterae* and *Thalassiosira fluviatilis*. Forward scattering measurements from an isolated sphere have been recently made using the Guoy phase shift that occurs at the waist of a focused Gaussian beam.^[9] However, it had a different motivation and is applicable only to particles so small that the scattering phase shift can be neglected in the analysis. By contrast, the present work is directed towards all particles including larger ones for which there are appreciable scattering phase shifts.

Specifically, we have developed a new approach to the measurement of light scattering at 0° from particle suspensions as well as isolated scatterers. The approach is based on the fanning of a coherent light beam in a photorefractive $BaTiO_3$ crystal.^[11]

Beam fanning or asymmetric self defocusing of a laser beam in a photorefractive BaTiO₃ crystal has been described and several applications have been demonstrated.^[12-18] Briefly, the phenomenon arises because a coherent light beam passing through a BaTiO₃ crystal creates an anisotropic electric field via optically induced charge migration. This field in turn produces an index gradient that forms along a direction orthogonal to beam propagation. The grating deflects light several angular degrees away from the incident direction into the fanning direction. A 45° cut BaTiO₃ crystal with dimensions $5 \times 5 \times 5$ mm³ deflects more than 99% of the energy of a normal incidence cw laser beam.

A finite time is required for the photo-induced charge migration and the resulting beam fanning to be established. This time τ is

known as the photorefractive response time; it is inversely proportional to the intensity of the incident laser beam.^[14] A BaTiO₃ crystal exhibits strong beam fanning even with very weak (microwatt) laser beams. The time response becomes quite slow at these low intensities. If the spatial character or the phase of any part of the beam is perturbed rapidly compared to τ , then the perturbed portion does not undergo fanning and is transmitted without deviation. This is the basis of our technique to separate the scattered and unscattered beams. Such photorefractive behavior has been used in several novel devices such as optical limiters.^[14] time integrating optical interferometers.^[15] adaptive spatial filters.^[16] and transient detection microscopes.^[17]

Fig. 2 shows a schematic of the experimental setup. The cw Ar⁺ laser (514.5 nm) is polarized in the plane of the figure, as is the C-axis of each BaTiO₃ crystal. The first crystal is 45° cut and the second is 0° cut. Each crystal was a cube of approximately $5\times5\times5$ mm³. For all measurements the laser power was kept below 10 mW to maintain a relatively long response time ($\tau \approx$ a few sec). Adjustable apertures A1, A2, and A3 shield the two crystals and the detector from stray light. The lens collects the light that is transmitted through the two crystals and focuses it onto the detector.

The scattering sample is a micron size quartz fiber whose axis is normal to the plane of Fig. 2 (and thus to the incident laser beam). The fiber is mounted on a 12 rpm synchronous motor that rotates it in a circle of radius 3 cm. In each rotation, the fiber crosses the laser beam twice, producing two pulses of scattered light. The forward

scattered portion of these pulses together with the strong unscattered beam then passes through the two successive $BaTiO_3$ crystals. Initially, the crystals were exposed to the direct laser beam for several minutes. The 45° cut crystal produced a very strong beam fanning at normal incidence. However, due to the asymmetry of beam fanning, there was some intensity left on one side of the laser beam. The second crystal further reduced this background. More than 99% of the incident radiation was deviated out of the direct beam path, creating a relatively dark background in the forward direction. Each time the fiber crossed the laser beam, a pulse of light appeared in this dark background. This pulse was detected with a photodiode and the intensity was measured with a sampling oscilloscope. The oscilloscope was triggered by the synchronous motor which rotated the fiber.

Scattering by quartz fibers with radii from 1 μ m to 32 μ m was observed. The observation solid angle for these measurements was a cone with full angle of 0.08° centered at 0°. It was determined by the aperture A3, the collecting lens, and the position of the fiber. Occasionally, small noise signals were observed due to dust particles crossing the laser beam. These perturbations were minimized by averaging over several measurements. The fiber radii were estimated from the angular intensity distributions of their diffraction patterns. The experimental uncertainty of the radius measurement was approximately 0.5%.

Black dots in Fig. 3 show the measured intensity vs. the fiber radius. The rapidly oscillating solid line is the theoretical prediction

for the scattered intensity at zero degrees.^[3] For reference, the smooth solid line is the diffraction term which is proportional to the square of the radius.^[8] One fitting parameter, the normalization for the ordinate, was used. The data shows very good agreement with theory. To elaborate, Fig. 4 shows a section of this data on an expanded scale; the diameter of the black dots approximates the experimental uncertainty in 'he radius. Most experimental data points coincide with the rapidly oscillating theoretical curve. The small deviations for several fiber radii are believed to be due to minor imperfections in the fibers. Specifically, a small nonuniformity in the fiber cross section produces a significant change in the forward scattered signal. This is precisely one of the reasons that scattered intensity measurements at 0° are useful for size determinations.

Fig. 5 shows the variation of the forward scattered intensity as a function of the incident laser power for four different fiber radii. As predicted from Eq. (2), the forward scattered intensity is observed to be linearly proportional to the incident laser intensity.

Fig. 6 shows similar measurements for a fiber mounted on a 1 rpm motor. At this slower speed, the $BaTiO_3$ crystals will be able to correct for phase changes due to scattering at even lower intensities. Indeed, when the incident laser power exceeds only =4 mW, the scattered intensity no longer increases linearly. At this laser intensity the photorefractive response is sufficiently rapid that the crystal is beginning to follow the phase changes created by the fiber motion. Thus, the light scattered by the fiber, as well as the strong incident beam, are fanned out of the forward direction. The result is a reduced

signal at the detector. Controlling the intensity, and thereby the response time of the crystal, determines the minimum object speed necessary for measuring the scattering from it.

In conclusion, we have successfully used this new device to measure the forward scattering from a single micron size quartz fiber. This novel technique should also be applicable to measurements of forward scattering from aerosols as well as particle suspensions. Specifically, since the particles are in constant Brownian motion, they will produce a time dependent phase in the scattered light; beam fanning can then separate the scattered and unscattered light.^[19] We emphasize that this technique also discriminates against the light scattering from imperfections in other optical elements or cell windows is fanned out of the forward beam. Finally, measurements of forward scattering from a single fiber will be used to calibrate instruments for measuring light scattering at 0°.

This concept was initiated with support from the Office of Naval Research (Grant N00014-89-J-1466). Support to obtain the present results was provided by the Army Research Office (Grant DAAL03-91-G-0200).

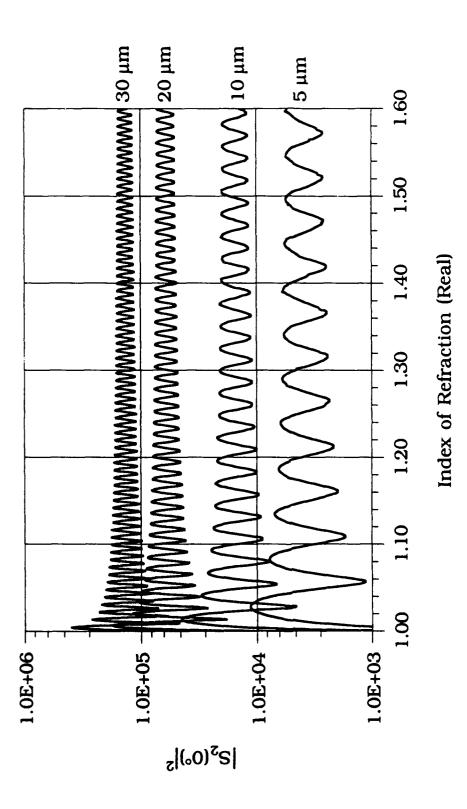
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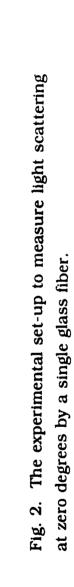
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Figure Captions

- Fig. 1. The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$, as a function of index of refraction for four different fiber radii. The wavelength is 514.5 nm and the absorption is zero.
- Fig. 2. The experimental set-up to measure light scattering at zero degrees by a single glass fiber.
- Fig. 3. The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$, vs. fiber radius. The laser wavelength is 514.5 nm.
- Fig. 4. The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$, vs. fiber radius for fiber radii from 12µm to 19µm. The laser wavelength is 514.5 nm.
- Fig. 5. Forward scattered signal as a function of input laser power. Each graph is for a fiber of a different radius ranging from $2.5\mu - 32\mu$.
- Fig. 6. Forward scattered intensity vs. the input laser power.

Fig.1 The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$, as a function of index of refraction for four different fiber radii. The wavelength is 514.5 nm





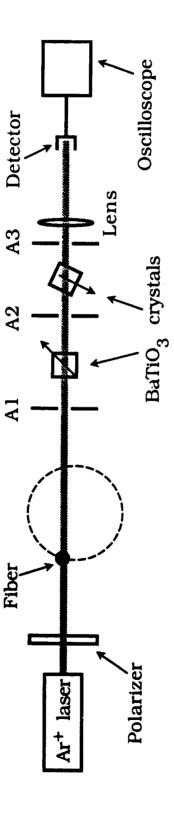


Fig. 3. The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$, vs. fiber radius. Laser wavelength is 514.5 nm.

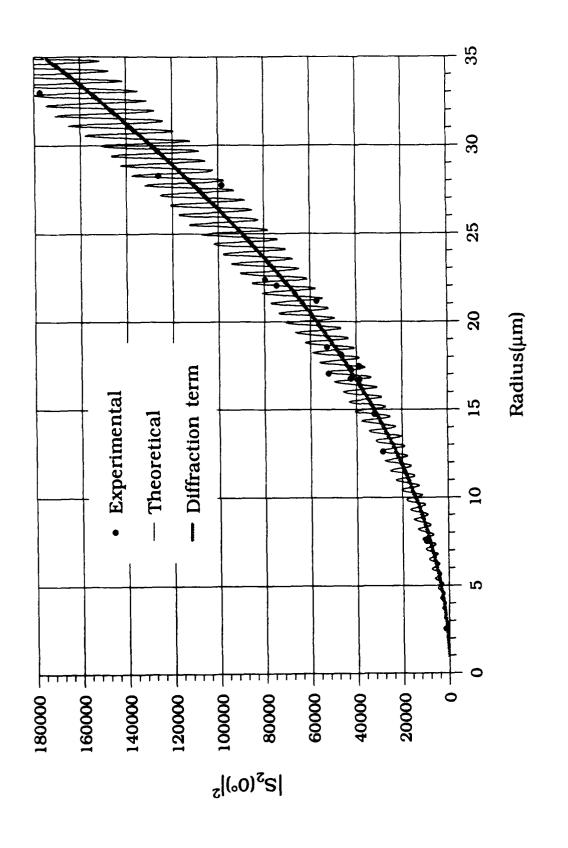
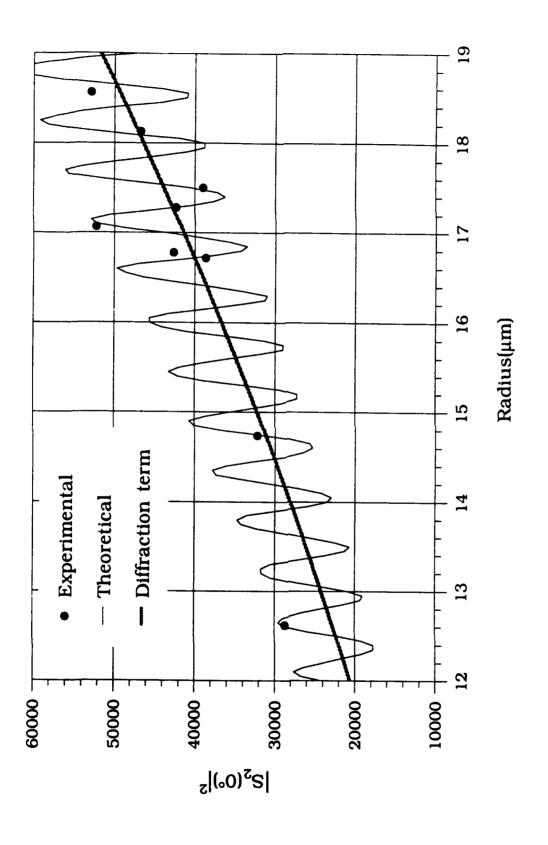
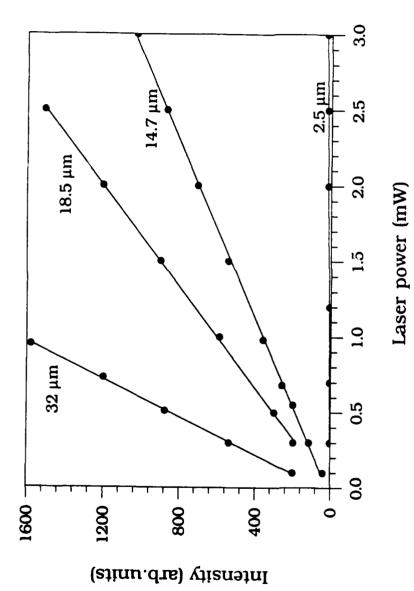


Fig. 4. The absolute square of the scattering amplitude at zero degrees, $|S_2(0^\circ)|^2$. vs. fiber radius. The laser wavelength is 514.5 nm.





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Fig. 5. Variation of the forward scattered intensity as a function of the input laser power.

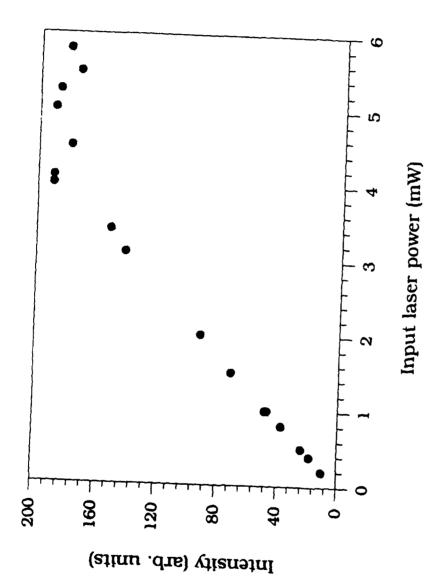


Fig. 6. Forward scattered intensity vs. input laser power (mW).