UNCLASSIFIED — MASTER COPY — FOR REPRODUCTION PURPOSES

SECURITY CLASSIFICATION OF THIS PAGE

# REPORT DOCUMENTATION PAGE

AD-A217 432

| | |
|---|---|
| 4. PERFORMING ORGANIZATION REPORT NUMBER(S) | 1b. RESTRICTIVE MARKINGS |
| | 3. DISTRIBUTION/AVAILABILITY OF REPORT<br>Approved for public release; distribution unlimited. |
| | 5. MONITORING ORGANIZATION REPORT NUMBER(S)<br>ARO 23909.4-GS |
| 6a. NAME OF PERFORMING ORGANIZATION<br>Texas A&M University<br>6b. OFFICE SYMBOL (If applicable) | 7a. NAME OF MONITORING ORGANIZATION<br>U. S. Army Research Office |
| 6c. ADDRESS (City, State, and ZIP Code)<br>College Station, TX 33843 | 7b. ADDRESS (City, State, and ZIP Code)<br>P. O. Box 12211<br>Research Triangle Park, NC 27709-2211 |
| 8a. NAME OF FUNDING/SPONSORING ORGANIZATION<br>U. S. Army Research Office<br>8b. OFFICE SYMBOL (If applicable) | 9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER<br>DAAL03-86-K-0168 |
| 8c. ADDRESS (City, State, and ZIP Code)<br>P. O. Box 12211<br>Research Triangle Park, NC 27709-2211 | 10. SOURCE OF FUNDING NUMBERS<br>PROGRAM ELEMENT NO. / PROJECT NO. / TASK NO. / WORK UNIT ACCESSION NO. |

11. TITLE (Include Security Classification)

Experimental and Theoretical Studies of Light Scattering by Small Particles

12. PERSONAL AUTHOR(S)

Edward S. Fry, George W. Kattawar, Chia-Ren Hu

| 13a. TYPE OF REPORT | 13b. TIME COVERED | 14. DATE OF REPORT (Year, Month, Day) | 15. PAGE COUNT |
|---|---|---|---|
| Final | FROM 9/15/86 TO 11/14/89 | December 1, 1989 | 12 |

16. SUPPLEMENTARY NOTATION The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

| 17. COSATI CODES | | |
|---|---|---|
| FIELD | GROUP | SUB-GROUP |
| | | |
| | | |

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)

Light Scattering; Laser Pulses; Plane Wave Scattering; Mueller Matrices; Optical Scattering, Dielectric Particles . (jhd)

19. ABSTRACT (Continue on reverse if necessary and identify by block number)


The following studies were made:

(1) Scattering of ultra-short laser pulses by microscopic particles both experimentally and theoretically.
(2) Plane wave scattering by small clusters of spheres in various configurations.
(3) Symmetries of Mueller matrices for forward (0 deg) and backward (180 deg) scattering by dielectric particles of various shapes.
(4) Optical scattering at zero degrees, and measurements of absorption in the presence of high densities of scatterers. Keywords:


| | |
|---|---|
| 20. DISTRIBUTION/AVAILABILITY OF ABSTRACT<br>☐ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS | 21. ABSTRACT SECURITY CLASSIFICATION<br>Unclassified |
| 22a. NAME OF RESPONSIBLE INDIVIDUAL | 22b. TELEPHONE (Include Area Code) / 22c. OFFICE SYMBOL |

DD FORM 1473, 84 MAR — 83 APR edition may be used until exhausted. All other editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE
UNCLASSIFIED

DTIC FILE COPY

90 01 23 186

ARO 23909.4-GS

# EXPERIMENTAL AND THEORETICAL STUDIES OF LIGHT SCATTERING BY SMALL PARTICLES

FINAL REPORT

EDWARD S. FRY
GEORGE W. KATTAWAR
CHIA-REN HU

DECEMBER 1, 1989

U. S. ARMY RESEARCH OFFICE

DTIC COPY INSPECTED 1

CONTRACT NUMBER / MODIFICATION NUMBER
DAAL03-86-K-0168 / P00011

TEXAS A&M UNIVERSITY
PHYSICS DEPARTMENT
COLLEGE STATION, TX 77843

Codes
/or
A-1

APPROVED FOR PUBLIC RELEASE;
DISTRIBUTION UNLIMITED.

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

## TABLE OF CONTENTS

## A. STATEMENT OF PROBLEMS STUDIED

Scattering of ultra- short laser pulses by microscopic particles has been studied both experimentally and theoretically. The interesting regime is that for which the physical length of the laser pulse is less than the particle size. Both our experimental and theoretical efforts concentrated on the scattering by a single sphere and included both time averaged and time resolved studies.

Since a femtosecond laser had to be constructed for this work, and since this was a new technology, we made studies of chirp and phase modulation in such lasers that led to two publications and a thesis.

Plane wave scattering by small clusters of spheres in various configurations has been studied. Definitive theoretical results have been obtained and have been compared with the microwave analog measurements of the upper-left 2×2 submatrix of the complete Mueller matrix. Laser levitation and manipulation techniques to produce arbitrary clusters of two, three, or four spheres have been investigated so that complete Mueller matrix measurements can be made in the visible.

Studies have been made of the symmetries of Mueller matrices for forward (0°) and backward (180°) scattering by dielectric particles of various shapes.

Other problems include investigations of optical scattering at zero degrees, and measurements of absorption in the presence of high densities of scatterers

## B. SUMMARY OF MOST IMPORTANT RESULTS

1) Time Averaged Scattering of Femtosecond Laser Pulses. Time averaged data with superb signal to noise has been obtained for ultrashort laser pulse scattering by spheres. All work was done at intensities sufficiently low that nonlinear effects were not important. Theoretical predictions were obtained by a Fourier decomposition of the incident pulse and then summing the plane wave Mie solutions with the appropriate weights. There was excellent agreement with the experimental data under all conditions. The results using pulses of similar bandwidth but different pulse durations show conclusively that time averaged results depend only on the spectral content of the pulses and not on the temporal duration.

2) Time Resolved Scattering (Experimental). In a sense this is optical radar on microscopic particles. Experimental work has been limited to time resolved scattering from thin plates and fibers. Both interference and auto-correlator techniques have been used. Two internal reflections in a 240 μm glass plate have been observed and time resolved in both forward and backscattering. Observing two internal reflections in forward scattering corresponds to observing a signal that is down in intensity by a factor of $\approx 3\times10^{-6}$ from the incident beam. Two time resolved reflections from a 75 μm fiber have also been observed. It is straight forward to extend this work to time resolved scattering by a single levitated microscopic particle. Amplification of the reference beam for the auto correlator will increase the sensitivity by $\approx 10^5$; an excimer laser will soon be available that can pump a bow-tie amplifier and provide this gain of $10^5$.

3) Time Resolved Scattering (Theoretical). We have written a set of programs to calculate the time-resolved scattering of ultra-short pulses from a dielectric sphere at any scattering angle. For the special case of backscattering, our results are in agreement with the earlier work by Rheinstein for this case. For sufficiently short pulses, the

scattered intensity is a set of discrete peaks occurring at characteristic delay times. Previous studies have succeeded in identifying many of the major peaks as due to axial rays, bounce rays, stationary rays and surface waves. The surface waves include the possibility of taking short cuts through the dielectric sphere. Refraction into the sphere occurs at the critical angle and internal reflection can occur many times before the ray exits. However, the identifications made by previous workers are at most semi-quantitative, leaving clearly observable discrepancies between the calculated peak positions and predicted positions based on some simple geometric-optics formulae. These discrepancies are bigger for the later (and therefore much weaker) peaks, which correspond to longer travel times inside the dielectric sphere. Also some peaks were not identified.

We have done calculations to much higher precision in order to include many more peaks in our analysis and thus enable us to analyze these discrepancies in more detail. Calculations of the scattered intensity vs delay time have been plotted on a semi-log scale covering 12 orders of magnitude. Programs have also been written to give the predicted positions of all four types of peaks based on geometric optics. Identifications made in previous works were based mainly on the positions of the back-scattering peaks. We have also looked for other signatures, such as the angular dependence of the scattered pulses. Our much more detailed study has enabled us to uncover several new features not discovered and/or understood in previous work. (Some are specifically related to forward scattering which was not studied previously.) We have reported some of these new findings in the 1988 and 1989 CRDEC conferences, and have also written a technical report for the 1988 conference proceedings.

Unfortunately, the computers we have been using in our calculations, a VAX 11/782 main frame and a VAX 2000 work station, do not have the accuracy we require. (After all, we need to analyze peaks which have at least ten orders of magnitude less intensity than the most intense peak!). We recently concluded that it was absolutely necessary to switch to a new machine-- a DEC station 3100 which is roughly 11.5 times faster then the VAX 11/782 and the VAX 2000 work station, and have now managed to obtain a large number of results which are much more reliable than those previously obtained. We have presented some of these results at the 1989 CRDEC conference. In order to reach reliable conclusions about all the discrepancies and new features we have observed, a more thorough study using a computer such as the new IBM 25 MIPS machine (or perhaps even a supercomputer) is required. Fortunately, Texas A&M University has recently acquired a new CRAY Y-MP2/116 supercomputer which we anticipate using for a more definitive study of this problem.

4) Femtosecond Laser Development. We have discovered and developed a simple modification that greatly enhances the stability of colliding pulse mode-locked(CPM) femtosecond dye lasers. It basically consists of a spectral filter that is placed within the four prism sequence that is used to provide negative group velocity dispersion in the laser cavity. It enables one to run the laser reliably for long periods of time at much shorter temporal widths than was previously possible.

We have also completed rigorous calculations of the time dependence of the frequency shifts arising from the collisions of femtosecond optical pulses both in nonresonant and in saturable media. The results are particularly applicable to the gain and absorber jets in CPM lasers and show the conditions for short, stable pulse formation. It was shown that there are conditions for which the pulses produced would be spread by either sign of group velocity dispersion.

5) Optical Scattering by Sphere Multiplets. We developed an order-of-scattering technique to study the light scattering of two or more interacting spheres. This new method provides, what is to our knowledge, the first complete physical description of the classical processes involved in cooperative EM scattering by an aggregate of spheres. Comparisons were made between microwave analog experimental results (kindly provided to us by Dr. Ru Wang) and our theoretical calculations for linear chains of three and five spheres. The effects of particle orientation were also investigated theoretically.

The order-of-scattering method was extended to the more difficult problem of scattering by clusters of spheres, the centers of which need no longer lie on a common axis. Comparisons were made between theoretical and experimental results for triangular and tetrahedral arrays of spheres and the agreement was very good.

Experimental work on this problem has proceeded to the point that we have developed laser levitation techniques to levitate and individually manipulate up to three solid spheres into arbitrary configurations. This would have been much simpler with liquid spheres, but it is necessary to use solid spheres because they will be in contact in some configurations. Complete Mueller matrices will be measured so a much more thorough test of the calculations is possible than with the microwave analog measurements.

6) Mueller Matrix Symmetries in Forward/Backward Scattering. By determining the symmetries in the Mueller matrix for scattering by dielectric particles of various shapes in the forward (0°) and backward (180°) directions, we have been able to develop general classifications of particle morphologies. For forward scattering there are six distinct symmetry classes with distinct scattering properties; for back scattering there are three. Each includes a class whose Mueller matrix has no signature indicative of a particle symmetry. We believe this would provide an ideal classification scheme for nonspherical

particles, particularly in view of the promising new approach to the measurement of scattering at zero degrees described in the following.

7) Scattering at an Angle of Zero Degrees. We have designed a scheme which for the first time permits measurement at zero degrees of the optical scattering by a single particle. It involves modulating the phase in one arm of a Mach-Zehnder interferometer and modulating the position of the particle in the other arm. Both the amplitude and phase of the zero degree scattering can be measured using lock-in detectors at linear combinations of the modulation frequencies and their harmonics. The first direct verification of the Optical Theorem in the visible region of the spectrum will now be possible. The method was described at the 1989 CRDEC conference.

8) Absorption Measurements in the Presence of Scattering. We have developed a new instrument for accurately measuring the optical absorption of a medium even in the presence of a high density of scatterers. Measurements of the absorption of pure water have been made to an accuracy of 1% to 2% even in the presence of nonabsorbing scatterers at a density that gives an extinction length due to scattering of ≈5 cm. The device was described at the 1989 CRDEC conference and in the proceedings. Application of the method to the measurement of the absorption of aerosols and smokes is being pursued.

## C. LIST OF ALL PUBLICATIONS, REPORTS, ETC.

Publications (Refereed):

1. "Mueller Matrix Calculations for Dielectric Cubes: Comparison With Experiments", G. W. Kattawar, C-R. Hu, M. E. Parkin, and P Herb, Appl. Opt. **26**, 4174 (1987).

2. "Symmetry Theorems on the Forward and Backward Scattering Mueller Matrices for Light Scattering from a Nonspherical Dielectric Scatterer", C-R. Hu, G. W. Kattawar, M. E. Parkin, and P. Herb. Appl. Opt. **26**, 4159 (1987).

3. "Colliding Pulse Mode-Locked Dye Laser Stabilization Using an Intracavity Spectral Filter", K. L. Schehrer, E. S. Fry and G. T. Bennett, Appl. Opt. **27**, 1908 (1988).

4. "Consummate Solution to the Problem of Electromagnetic Scattering by an Ensemble of Spheres. I: Linear Chains", K. A. Fuller and G. W. Kattawar, Optics Letters, **13**, 90 (1988).

5. "Consummate Solution to the Problem of Classical Electromagnetic Scattering by an Ensemble of Spheres. II: Clusters of Arbitrary Configuration", K. A. Fuller and G. W. Kattawar, Optics Letters, **13**, 1063 (1988).

6. "Colliding-Pulse Phase Modulation and Chirping of Ultrashort Optical Pulses in Thin Slabs of Nonresonant and of Saturable Media", K. L. Schehrer and E. S. Fry, Optical Society of America B, **6**, 1182 (1989).

7. "Time Averaged Scattering of Femtosecond Laser Pulses", W. E. White, C. Wang, and E. S. Fry, Submitted to Appl. Opt., December, 1989.

Publications (Proceedings):

8. "The Scattering of Femtosecond Optical Pulses by Small Dielectric Spheres", G. T. Bennett, E. S. Fry, W. E. White, P. Herb, G. W. Kattawar, and W. Xu, in Proceedings of the 1987 CRDEC Scientific Conference on Obscuration and Aerosol Research, 387-393.

9. "Light Scattering by an Arbitrarily Configured Cluster of Spheres", K. A. Fuller and G. W. Kattawar, in Proceedings of the 1987 CRDEC Scientific Conference on Obscuration and Aerosol Research, 631-644.

10. "Calculation of Time-Resolved Scattering of Femtosecond Light Pulses from Dielectric Spheres" C.-R. Hu, G. H. Rose and G. W. Kattawar, Submitted to the Proceedings of the 1988 CRDEC Scientific Conference on Obscuration and Aerosol Research.

11. "Time Resolved Measurements of the Scattering of Femtosecond Laser Pulses", W. E. White, C. Wang, and E. S. Fry, Submitted to the Proceedings of the 1988 CRDEC Scientific Conference on Obscuration and Aerosol Research.

12. "Scattering of Femtosecond Optical Pulses by Small Dielectric Particles", E. S. Fry, W. E. White, and C. Wang, Conference on Lasers and Electro-Optics, 1989 Technical Digest Series **11**, 202-203 (1989)

13. "Scattering of Femtosecond Laser Pulses", W. E. White, C. Wang, and E. S. Fry, Submitted to the Proceedings of the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research.

14. "Absorption Measurements Using an Integrating Cavity", R. M. Pope, E. S. Fry, and R. L. Montgomery, Submitted to the Proceedings of the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research.

Theses:

15. "Cooperative Electromagnetic Scattering by Ensembles of Interacting Spheres", Kirk A. Fuller, PhD Dissertation, Texas A&M University, 1987.

16. "Scattering of Femtosecond Laser Pulses", William E. White, PhD Dissertation, Texas A&M University, 1989.

17. "Intracavity Evolution of Ultrashort Optical Pulses in a Colliding Pulse Mode-Locked Laser", Kevin L. Schehrer, PhD Dissertation, Texas A&M University, 1989.

Presentations:

18. "A New Method for the Study of Light Scattering by Ensembles of Interacting Spheres", presented at the Joint Fall Meeting of the Texas Sections of the American Physical Society and of the American Association of Physics Teachers, Nacogdoches TX (Nov., 1986).

19. "The Scattering of Femtosecond Optical Pulses by Small Dielectric Spheres", G. T. Bennett, E. S. Fry, W. E. White, P. Herb, and G. W. Kattawar, presented at the 1987 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1987).

20. "Mueller Matrix Calculations for Two Interacting Dielectric Cubes", G. W. Kattawar and P. Herb, presented at the 1987 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1987).

21. "Light Scattering by Ensembles of Interacting Spheres", presented to the Life Sciences Division of Los Alamos National Laboratory, Los Alamos NM (July, 1987).

22. "Scattering of Femtosecond Laser Pulses by a Single Levitated Sphere", G. T. Bennett, E. S. Fry, W. E. White, P. Herb, G. W. Kattawar, and W. Xu, presented at the 1987 Annual Meeting of the American Association for Aerosol Research, Seattle WA (Sept., 1987).

23. "Scattering of Femtosecond Pulses from Dielectric Spheres", W. E. White, E. S. Fry, and G. W. Kattawar, presented at the 1988 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

24. "Calculations of Time-Resolved Scattering of Femtosecond Pulses from Dielectric Spheres", presented at the 1988 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

25. "Mueller Matrix Measurements for Sphere Aggregates", K. Thieme and E. S. Fry, presented at the 1988 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

26. "Scattering of Femtosecond Optical Pulses by Small Dielectric Particles", E. S. Fry, W. E. White, and C. Wang, presented at the Conference on Lasers and Electro-Optics, Baltimore MD (April, 1989).

27. "Analysis of Time-Resolved Scattering of Femtosecond Light Pulses from a Dielectric Sphere", G. H. Rose, C.-R. Hu, and G. W. Kattawar, presented at the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

28. "Recent Femtosecond Scattering Results", C. Wang, W. E. White, and E. S. Fry, presented at the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

29. "Precision Absorption Measurements in a Highly Scattering Medium", R. M. Pope, R. L. Montgomery, and E. S. Fry, presented at the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

30. "Scattering from Sphere Doublets", K. L. Thieme, W. E. White, and E. S. Fry, presented at the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

31. "Scattering by a Single Sphere at an Angle of Zero Degrees", D. F. McCoy, and E. S. Fry, presented at the 1989 CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood MD (Jun., 1988).

32. "Integrating Cavity Absorption Meter", E. S. Fry, G. W. Kattawar, R. M. Pope, R. L. Montgomery, and J. S. Cleveland, **invited** paper at the Annual Meeting of the Am. Optical Society, Orlando FL (Oct., 1989).

## D. LIST OF ALL PARTICIPATING SCIENTIFIC PERSONNEL.

Principle investigators:

Dr. Edward S. Fry, Professor of Physics

Dr. Chia-Ren Hu, Professor of Physics

Dr. George W. Kattawar, Professor of Physics

Post-Doctoral Fellows:

Dr. Glenn T. Bennett

Dr. Pascal Herb

Dr. Juan Carlos Reina

Dr. Greg Rose

Graduate Research Assistants:

Dr. Kirk A. Fuller, received PhD. 1987

Dr. Kevin L. Schehrer, received PhD. 1989

Dr. William E. White, received PhD. 1989

Ms. Xue-Mei Gong, received non-thesis M. S. 1987

Ms. Weihao Xu, received non-thesis M. S. 1988

Mr. Xingfu Li

Ms. Zhangfen Qi

Ms. Kelly Thieme

Mr. Ching Wang

Mr. Eric Yuen

Undergraduate Laboratory Assistants:

Mr. John Knesek

Mr. David F. McCoy

Ms. Madeleine L. Naudeau