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13. ABSTRACT (Maximum 200 words) Researchers have developed techniques required for a more thorough analysis of nonlinear optics in droplets and have developed models for some nonlinear processes. They have continued to develop techniques for modeling the resonances of perturbed spheres, based primarily on perturbation methods. Falling droplets deviate from perfect sphericity, and laser beams can induce inhomogeneities in the droplets. The techniques developed for perturbed spheres now allow us to model dispersive and absorptive optical bistability in droplets. They have modeled linear time-dependent internal fields in droplets illuminated with laser pulses, as a prelude to analyzing the nonlinear optical effects usually observed with pulsed laser illumination. They have developed techniques for computing the internal and scattered fields of droplets illuminated with Gaussian beams. Since droplets are often illuminated with Gaussian beams which may be focused on or outside the droplets, this capability is needed for a thorough modeling of nonlinear optics in droplets.					
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**FINAL REPORT:
COMPUTATION OF
NONLINEAR OPTICAL SCATTERING BY DROPLETS**

Army Research Office Contract Number DAAL03-90-G-0143 to Clarkson University with
subcontract to New Mexico State University

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1 THE PROBLEM STUDIED

Laser beams are being used to characterize the atmosphere [1]. Lidar (Light Detection and Ranging) is increasing in importance. Lightwaves are also used for free-space communications. In using lightwaves to characterize or to modify the atmosphere, and in using lightwaves to communicate through the atmosphere, it is important to understand the interactions between optical radiation and the droplets or particles in the air. Linear and nonlinear optical scattering in droplets is also used to study growth and evaporation of droplets, adsorption of materials onto droplets, and chemical reactions in droplets. Light scattering is also being used to characterize droplets in sprays.

The scattering of light by homogeneous spheres composed of linear media is well known [2]. However, laser beams used for characterization of the atmosphere often have intensities large enough to perturb the refractive indices of the droplets. Consequently, the usual analyses which assume linear optical properties are not always valid, and methods for computing the nonlinear optical scattering by droplets are required.

Nonlinear optical effects such as phase modulation lineshape broadening [3], stimulated Raman scattering (SRS) [4-7], stimulated Brillouin scattering (SBS) [8], third harmonic generation (THG) and third-order sum frequency generation [9], and lasing [10-11] have been observed in droplets. The effects typically occur in droplets with lower intensities than are required in bulk materials. The primary reason for the lower thresholds is the optical feedback provided by morphology dependent resonances, MDR's [12-13]. If the refractive index of the droplet is a function of both time and space, because of its dependence on the internal intensity, the MDR's and quality factors, Q 's, are also time varying.

Our objectives were to develop methods for computing the nonlinear optical scattering by droplets. Two classes of problems were considered: elastic scattering where the real part of the refractive index is intensity-dependent, and inelastic scattering where light is generated at new frequencies. An additional objective was to use the calculated results to help explain experimental measurements and to suggest new experiments.

2 SUMMARY OF MOST IMPORTANT RESULTS

We have developed techniques required for a more thorough analysis of nonlinear optics in droplets and have developed models for some nonlinear processes. We have continued to develop techniques for modeling the resonances of perturbed spheres, based primarily on perturbation methods. Falling droplets deviate from perfect sphericity, and laser beams can induce inhomogeneities in the droplets. The techniques developed for perturbed spheres now allow us to model dispersive and absorptive optical bistability in droplets. We have modeled linear time-dependent internal fields in droplets illuminated with laser pulses, as a prelude to analyzing the nonlinear optical effects usually observed with pulsed laser illumination. We have developed techniques for computing the internal and scattered fields of droplets illuminated with Gaussian beams. Since droplets are often illuminated with Gaussian beams which may be focused on or outside the droplets, this capability is needed for a thorough modeling of nonlinear optics in droplets.

2.1 Results in Journal Articles Published or Submitted

2.1.1 Angular Fine Structures in Stimulated Raman Scattering

Paul Chen, William Acker, and Richard Chang at Yale University measured the angular distribution of the SRS emitted by droplets. These angular distributions were used to identify the MDRs which support the SRS. When the incident laser intensity is focused on the edge of the droplet then lower-order MDRs are more likely to support the SRS. This work was largely completed prior to the initiation of the ARO contract, but was completed during the period of the contract. This work was also discussed by Richard Chang at the SPIE meeting in Dallas, TX May 8-10, 1991.

2.1.2 Splitting of Degenerate Modes Observed in Stimulated Raman Scattering Spectra of Falling Droplets

Paul Chen and Richard Chang measured in falling droplets the splitting of MDRs which was predicted [14] to occur in perturbed droplets. The eccentricity of the droplets as determined from the splitting is approximately 0.001. This work was also presented by Paul Chen at the CRDEC meeting at Aberdeen in June, 1991.

2.1.3 Morphology Dependent Resonances in Spherically Symmetric Radially Inhomogeneous Spheres

A Runge-Kutta method was used to compute the Q 's and field distributions of droplets which have a radially inhomogeneous refractive index. There was no dramatic reduction in the Q 's with small changes in the refractive index. However, an intensity-dependent refractive index, in a region where the high intensity of an MDR is largest, could cause the droplet to shift off the resonance. Richard Chang and his students have been concerned about the possible effects of a refractive index profile different from the homogeneous one always assumed in computations. The primary concern was that there might be a large reduction in the Q . Ronald Pinnick also suggested that this was an important question. This work was also presented by Chowdhury at the CRDEC meeting in Aberdeen in June 1991.

2.1.4 Internal Fields of Droplets Computed Using Physical Optics (Mie Theory) and Ray Tracing

Large internal fields are particularly important in determining which nonlinear optical effects occur in droplets. To better understand where the large intensities occur in droplets we computed the energy density using geometrical optics as well as Mie theory. Some of the main features of the full Mie solution are reproduced quite well in the geometrical optics computation of the energy densities. Some of the similarities are remarkable. The cone of high intensity near the forward surface is caused by rays that were bent once at the first droplet surface. The highest intensity region along the central axis is caused by rays that have reflected once internally. In a ray tracing model this highest intensity region is infinitely thin. In the Mie model the width about the axis of this cylindrical region is about one internal wavelength. In a water droplet, the rays that cause the other "hot spots" on the axis are ones that have undergone three internal reflections. This work was also presented

by Hill at the SPIE meeting in Dallas, 1991, and by Chowdhury in a poster session at the CRDEC meeting in Aberdeen, June, 1991.

2.1.5 Near-Resonance Excitation of Dielectric Spheres with Plane waves and Off-Axis Gaussian Beams

The key finding is that modes having high-Q MDRs can dominate the internal energy distribution in spheres even when excited many linewidths from the resonance location. The near-resonances dominance is more pronounced when the incident field is a Gaussian beam focused outside the droplet. These findings are consistent with the measurements of Chen et al, Opt. Lett., 16, 117 (1991). The findings are relevant to discussions about the density of modes which are important in supporting nonlinear optics. This work was stimulated in part by discussions with Richard Chang, David Leach and other members of the Yale group concerning the relevance of frequencies many linewidths from a MDR. This work was presented by Khaled in a poster session at the CRDEC meeting in Aberdeen in June, 1991, and at the IEEE Lasers and Electro-Optics Society annual meeting in San Jose, CA in November, 1991.

2.1.6 Time-Dependent Internal Intensities in Droplets On or Near Resonance

To model the time-dependent nonlinear optical fields, we need to understand the time-dependent linear internal fields, both on and near resonance. The time-dependent fields at a point are the inverse Fourier transform of the frequency spectrum of the fields at that point. The frequency spectrum at a point is the product of the transfer function and spectrum of the incident fields. The paper shows the build-up and decay of the fields for various pulse durations and trains of pulses at internal positions where the resonant fields either dominate or are relatively small. The results are relevant to ongoing experimental work by Professor Richard Chang's group at Yale. This work was presented by Barber in January 1992 at the Radio Science meeting in Boulder, CO.

2.1.7 Effects of Perturbations on Resonances

The time-independent perturbation method (TIPM) [14,15] is applicable for modeling the increase in the refractive index near the high intensity region of an illuminated droplet, or some other types of perturbations. However, it is an approximate method and its range of accuracy had not been determined. We used the T-matrix method for layered structures to determine the accuracy of the TIPM results. The TIPM predicts the resonance locations very well, and gives less accurate, but still useful, approximations of the decrease in the Q, if the Q decrease in the Q is less than about 50 percent. Preliminary findings were presented by Hill at the SPIE meeting at Dallas, May, 1991, and by Chowdhury at the CRDEC meeting in June, 1991.

2.1.8 Scattered and Internal Intensity of a Spherical Object Illuminated with a Gaussian Beam

The method we used to compute the fields of a droplet illuminated by a Gaussian beam is as follows. A polarized Gaussian beam is first expanded in an angular spectrum of plane

waves by doing a spatial Fourier transform. These plane waves are then expanded in vector spherical harmonics. Consequently the beam is a vector field which satisfies Maxwell's equations. The beam can be focused as tightly as can real fields, and the method can be used to model any field which can be represented as a summation of homogeneous plane waves. Calculated results for beams having waists much smaller than the radius of the sphere help in visualizing how a beam reflects and refracts at the surface of the sphere. This work was presented by Khaled at the Applied Computational Electromagnetics Society Meeting in March, 1992.

2.1.9 Third-Order Sum-Frequency Generation

New third-order sum-frequency generation (TSFG) spectra of droplets have been measured. Some of these spectra have a very high resolution. The results and the required phase matching were compared and contrasted with the case of sum frequency generation in optical fibers. A detailed model of time-harmonic TSFG has also been developed and the numerical results have been compared qualitatively with the experimental spectra. The spatial and frequency overlap between the generating and output modes are included in the analysis. This work was discussed at the CRDEC meeting in June 1992 and presented at the OSA meeting in Albuquerque in Sept. 1992. This work will be discussed at the SPIE meeting in January, 1993.

2.1.10 Effective Average Gain and Loss Coefficients in Inhomogeneous Spheres: Application to Two-Photon Absorption

The effective gain or loss coefficient for a laser mode typically is computed by taking the average of the gain or loss coefficient weighted by the intensity of that mode at each point. In droplets the evanescent fields outside the droplet overlap with the energy radiating away from the droplet. Consequently, computing the effective average gain or loss coefficient is not as straightforward as in the diode laser case.

A key to overcoming the problem of the overlap of the evanescent fields and the radiated fields outside the droplet, is to realize that in the analysis of a linear laser cavity, the energy that has left the cavity is not considered as part of the mode in determining the effective gain or loss. The same must be true in a droplet: only the evanescent fields outside the droplet need be considered in computing the average gain or loss coefficient for a mode. The energy that is radiating away is irrelevant.

Using these ideas we have computed the effective average loss coefficient for various MDRs in homogeneous and layered spheres, have used these values to estimate the Q 's, and have compared these Q 's to the exact Q 's computed using separation of variables. The Q 's of inhomogeneous absorbing spheres were also computed using the first order time-independent perturbation method [14] and the results were very good. (Note that only first-order perturbation theory is required when there is gain or loss, but that second order perturbation theory [15] is required for the change in Q caused by perturbations in the real part of the refractive index.) The effective average gain or loss for each mode can be computed even far from a resonance.

Professor Richard Chang and his students have been measuring the scattering and SRS from droplets containing materials which absorb two-photons. The loss coefficient at any

point is proportional to the intensity at that point. With the method of determining the average loss coefficient mentioned above included two-photon absorption effects in our modeling. We have noted that in the case of two-photon absorption at a single frequency at least, bistability should not occur. This work was presented by Chowdhury at the CRDEC meeting in June 1992 and at the OSA meeting in Albuquerque in Sept., 1992.

2.2 Papers Near Completion — Not Yet Submitted

2.2.1 Linear Scattering of Gaussian Beams, and Time-Dependent Gaussian Beams by Absorbing Droplets

Richard Chang from Yale raised two questions about our findings on low order modes being excited by off-axis Gaussian beams: 1) Absorption is known to affect high-Q (low-order) MDRs more than low-Q MDRs. Are the internal fields of an absorbing spheres illuminated with a Gaussian beam still dominated by the low-order modes? 2) Most of the experimental work on nonlinear optics in droplets is done with pulses which may be as short as 100 ps or as long as many ns, or is done with trains of very short pulses. It takes time for energy to get into the high-Q MDRs.

We studied the time dependence of the internal and scattered fields of lossless and absorbing droplets excited with pulses focused on or outside the droplet. The incident fields were plane waves and Gaussian beams. With long pulses the low-order modes can continue to dominate the internal intensity even with absorption. The intensity of the mode many linewidths from the resonance is not so affected by the absorption many linewidths from the resonance frequency. This work was presented by Khaled at the CRDEC meeting, June, 1992, and at the OSA meeting in Albuquerque, in Sept., 1992. The time-dependent Gaussian beam work has been submitted to the Applied Computational Electromagnetics Society Meeting in March, 1993.

2.2.2 Optical Bistability

The shift in the resonance location of an MDR is linearly proportional to the maximum amplitude of the change in the refractive index (Δn) of an inhomogeneous sphere if Δn is real and small. The proportionality constant can be computed using the time-independent perturbation method. We have found that if the sphere is illuminated near an MDR so that one MDR dominates the field, then a simple expression for optical bistability can be written.

3 LIST OF PUBLICATIONS

3.1 Journal Articles Published or Submitted

1. G. Chen, W. P. Acker, R. K. Chang, and S. C. Hill, "Angular fine structure in the stimulated Raman scattering from single droplets," *Opt. Lett.*, 16, 117-119 (1991).

2. G. Chen, R. K. Chang, S. C. Hill, and P. W. Barber "Frequency splitting of degenerate spherical cavity modes: stimulated Raman scattering spectrum of deformed droplets," *Opt.*

Lett., 16, 1269-1271 (1991).

3. D. Q. Chowdhury, S. C. Hill, and P. W. Barber, "Morphology-dependent resonances in radially-inhomogeneous spheres," J. Opt. Soc. Am. A, 8, 1702-1705 (1991).

4. D. Q. Chowdhury, P. W. Barber, and S. C. Hill, "Energy-density distribution inside large nonabsorbing spheres via Mie theory and geometrical optics," Appl. Opt., pp. 3518-3523 (1992).

5. E. E. M. Khaled, S. C. Hill, P. W. Barber, and D. Q. Chowdhury, "Near-resonance excitation of dielectric spheres with plane waves and off-axis Gaussian beams," Appl. Opt., pp. 1166-1169 (1992).

6. D. Q. Chowdhury, S. C. Hill, and P. W. Barber, "Time dependence of internal intensity of a dielectric sphere on and near resonance," J. Opt. Soc. Am. A., pp. 1364-1373 (1992).

7. M. M. Mazumder, S. C. Hill, and P. W. Barber, "Morphology-dependent resonances in inhomogeneous spheres: comparison of the layered T-matrix and time-independent perturbation method," J. Opt. Soc. Am. A, 9, pp. 1844-1853 (1992).

8. E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Scattered and internal intensity of a sphere illuminated with a Gaussian beam," accepted in IEEE Trans. Antennas Propagat.

9. D. H. Leach, R. K. Chang, W. P. Acker, and S. C. Hill, "Third Order Sum Frequency Generation in Droplets: Experimental Results," J. Opt. Soc. Am. B., scheduled for January 1993.

10. S. C. Hill, D. H. Leach, and R. K. Chang, "Third Order Sum Frequency Generation in Droplets: Model with Numerical Results for Third-Harmonic Generation," J. Opt. Soc. Am. B., scheduled for January 1993.

11. D.Q. Chowdhury, M.M. Mazumder, and S.C. Hill, "Quality Factors and Effective Absorption and Gain Coefficients in Inhomogeneous Spheres: Application to Two-Photon Absorption," submitted to IEEE J. Quantum Electronics.

3.2 SPIE Proceedings Articles

S. C. Hill, P. W. Barber, D. Q. Chowdhury, E. E. M. Khaled, and M. Mazumder, "Resonances and internal energy density in droplets," in Nonlinear Optics and Materials, SPIE Proceedings Vol. 1497 (1991).

R. K. Chang, G. Chen, S. C. Hill, and P. W. Barber, "Nonlinear optical processes in droplets with single-mode laser excitation," in Nonlinear Optics and Materials, SPIE Proceedings Vol. 1497 (1991).

S. C. Hill, D. H. Leach, R. K. Chang, "Model of coherent nonlinear optical interactions in droplets: third-order sum-frequency generation," submitted to SPIE conference on Laser Applications in Combustion and Combustion Diagnostics (Los Angeles, January, 1993).

3.3 Journal Articles Nearly Completed

E. E. M. Khaled, D. Q. Chowdhury, S. C. Hill, and P. W. Barber, "Internal and scattered time-dependent intensity of a dielectric sphere illuminated with a Gaussian or a plane wave pulse," will be submitted to IEEE Trans. Antennas and Propagat.

E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Internal electric energy in a spherical particle illuminated with plane waves or off-axis Gaussian beams," will be submitted.

E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Light scattering by a coated sphere illuminated with a Gaussian beam," will be submitted, probably to Appl. Opt.

M.M. Mazumder, S.C. Hill, and D.Q. Chowdhury, "Dispersive bistability in a dielectric sphere," will be submitted to Phys. Rev. A.

4 PARTICIPATING SCIENTIFIC PERSONNEL

Steven C. Hill and Professor Peter W. Barber (co-PI's)

Dipakbin Q. Chowdhury — PhD, August 1991, Clarkson University. Now he is a research associate (Yale University and New Mexico State University) continuing to work on these problems.

E. Esam M. Khaled — PhD, November 1992, Clarkson University. He plans to return to Assiut University as an Assistant Professor of Electrical Engineering.

M. Mohiuddin Mazumder — MS, May 1992, New Mexico State University. Now he is a graduate student at Yale University with Professor Richard K. Chang where he continues to work on nonlinear optics in droplets.

Professor Richard K. Chang (Yale University) and his students Paul Chen, David H. Leach, and William Acker have collaborated extensively in this work. They, Professor Chang's other students (Ali Serpenguzel, Alfred Kwok, Christian J. Swindal, and Janic Cheng), and their experimental results have provided a great impetus for the work accomplished and the ideas generated.

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9. W. P. Acker, D. H. Leach, and R. K. Chang, "Third-order optical sum frequency generation in micrometer-size liquid droplets," submitted to *Opt. Lett.*

10. H.-M. Tzeng, K. F. Wall, M. B. Long, and R. K. Chang, "Laser emission from individual droplets at wavelengths corresponding to morphology-dependent resonances," *Opt. Lett.*, 9, 499 (1984).

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