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6. AUTHOR(S)
Richard K. Chang

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Richard K. Chang
Department of Applied Physics
Yale University
P.O. Box 208284
New Haven, CT 06520

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13. ABSTRACT (Maximum 200 words)

Liquid microdroplets play an important role in combustion as well as atmospheric chemistry. Characterizing droplets in terms of size, shape, chemical composition, temperature, etc., is essential for research in these two fields. Nonlinear optics has proven to be a useful diagnostic tool for determining these important characteristics. The research funded by this contract has resulted in the development of three new diagnostic techniques for characterizing microdroplets with mode-locked laser pulses: (1) The temporal beating of adjacent, degeneracy-split droplet cavity modes has been used to determine both the droplet distortion amplitude and the linewidths of the cavity resonances. (2) The localized, laser-induced electrostrictive distortion induced by a train of mode-locked laser pulses was shown to increase the input coupling to droplet cavity resonances for subsequent laser pulses and reduce the threshold for stimulated Raman scattering by almost two orders of magnitude. (3) Optical second harmonic generation has been used to both detect surfactant molecules adsorbed on droplet surfaces, as well as to determine the relative concentrations and molecular orientations of the surfactants.

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Temperature, Trace Speices, and Phase conjugation in Droplets and Sprays

Final Technical Report: August 31, 1997

(AASERT Contract No. F49620-94-1-0360)

Principal Investigator: Richard K. Chang

Yale University
Department of Applied Physics and Center for Laser Diagnostics
New Haven, Connecticut 06520-8284

OVERVIEW

Liquid microdroplets play an important role in combustion as well as atmospheric chemistry. Characterizing droplets in terms of size, shape, chemical composition, temperature, etc., is essential for research in these two fields. Nonlinear optics has proven to be a useful diagnostic tool for determining these important characteristics. The research funded by this contract has resulted in the development of three new diagnostic techniques for characterizing microdroplets with mode-locked laser pulses: (1) The temporal beating of adjacent, degeneracy-split droplet cavity modes has been used to determine both the droplet distortion amplitude and the linewidths of the cavity resonances. (2) The localized, laser-induced electrostrictive distortion induced by a train of mode-locked laser pulses was shown to increase the input coupling to droplet cavity resonances for subsequent laser pulses and reduce the threshold for stimulated Raman scattering by almost two orders of magnitude. (3) Optical second harmonic generation has been used to both detect surfactant molecules adsorbed on droplet surfaces, as well as to determine the relative concentrations and molecular orientations of the surfactants.

TECHNICAL DISCUSSION

The highly spherical shape of liquid microdroplets, combined with the abrupt index of refraction mismatch at the liquid-air interface, gives rise to high-Q optical modes known as morphology dependent resonances (MDR's). High-Q modes result when light circumnavigating the rim of the droplet, trapped by total internal reflection, closes upon itself in phase. When the pump wavelength and the gain bandwidth for a nonlinear process [such as stimulated Raman scattering (SRS), stimulated Brillouin scattering, or lasing] both overlap spectrally with MDR's, the resonances provide feedback for the nonlinear interaction, and the threshold for nonlinear processes is reduced. This condition is known as double-resonance. These MDR's, in combination with nonlinear optical processes, have been used to characterize many important aspects of droplet morphology.

For many applications involving dielectric spheres, both the spectral linewidths and the degeneracy splitting of the droplet azimuthal modes must be measured. The extremely high-Q ($Q > 10^{10}$), narrow linewidth modes of silica spheres are approaching the limits of resolution for spectral measurements. For droplets which deviate only slightly from sphericity, the splitting of the azimuthal modes can be too fine to be resolved spectrally. Therefore, a technique which is not limited by the spectral resolution of the instrumentation would be necessary for characterizing the high-Q modes of highly spherical droplets.

A new technique for simultaneously measuring both the MDR spectral linewidths and the degeneracy splitting of the droplet azimuthal modes has been developed. The decay of SRS from droplet cavity modes has been used to determine both of these droplet characteristics temporally, rather than spectrally. The exponential decay time of the SRS, $\tau_{MDR} = 1/\Delta\omega_{MDR} = Q/\omega$, was used to determine both the Q's and the resonance linewidths of the MDR's. When the SRS was detected from a narrow region near the droplet equator, oscillations were superimposed on this smooth exponential decay. These oscillations arose from the beating of degeneracy split azimuthal modes in slightly distorted droplets. The period of these oscillations was used to find the spectral spacing between adjacent, degeneracy split azimuthal modes. This spectral spacing was then used to determine the droplet distortion amplitude. Measurements of the MDR spectral linewidths and the droplet distortion amplitude can be made temporally even when these characteristics cannot be determined spectrally.

For generating nonlinear processes in droplets, high-Q cavity modes are often preferred for both the pump and nonlinearly generated wavelengths. High-Q MDR's have longer photon lifetimes within the cavity and, therefore, enhanced feedback compared with low-Q modes. The coupling of mode-locked laser pulses to high-Q droplet cavity resonances is inefficient, however. Mode-locked laser pulse have large Fourier transform limited spectral linewidths, whereas high-Q MDR's have narrow spectral profiles. This mismatch between the laser and MDR spectral profiles results in inefficient coupling of the mode-locked pulse to the droplet cavity modes.

A new mechanism to improve the input coupling of mode-locked laser pulses to high-Q droplet cavity modes has been investigated. The microdroplets were illuminated at the edge with a train of Q-switched, mode-locked laser pulses. The electrostrictive impulse resulting from high-intensity, mode-locked pulses locally distorted the droplet shape in the region of illumination. Since the shape distortion was retained for much longer than the 13.2 ns interpulse separation, the train of pulses had a cumulative effect. The localized shape distortion generated by previous mode-locked pulses and retained through the 13.2 ns interpulse separation was shown to increase the input coupling of subsequent mode-locked pulses to the droplet cavity resonances. Each successive pulse augmented the distortions caused by previous pulses and further increased the coupling to input MDR's for subsequent pulses. The increase in input coupling resulted in a higher internal intensity pumping the SRS for each subsequent pulse. The result was a decrease in the input intensity required to generate SRS for successive pulses in the mode-locked train.

Because the shape distortion was localized, the leakage rate of light from the cavity modes was only locally affected. The overall output MDR leakage rate was determined from a spatial average of the leakage rates from around the droplet rim. Provided the localized perturbed leakage rate was not too high, the overall Q's of those output MDR's which supported the SRS were not significantly degraded. When the cumulative distortion from a train of high-intensity pulses was too large, however, the output MDR's were degraded to such a degree that there was not sufficient feedback to support the SRS. Thus, the input intensity is not the only determining factor in generating nonlinear processes in droplets as it is in generating nonlinear interactions in optical cells. The history of the previous pulses and their impact on the cavity Q's has been shown to affect the generation of nonlinear processes in double-resonance. The results demonstrated that a gradual distortion to the droplet shape caused by a train of lower intensity mode-locked pulses is ideal for increasing the input coupling, while still allowing high-Q ($Q > 10^7$) output MDR's with to remain and support the nonlinearly generated radiation.

This technique could be applied to other nonlinear optical processes in the double-resonance condition such as stimulated Brillouin scattering and third-order sum frequency generation. Additionally, nonlinear effects which are pumped by SRS will be enhanced by the reduced SRS threshold. The reduced input intensity threshold for SRS was also shown to improve the detectability of minority species in multi-component droplets. The scattering of mode-locked pulses from microdroplets is used in light detecting and ranging (LIDAR) in the atmosphere. For these LIDAR studies, it is important to understand how the history of the mode-locked pulses affects the scattering of light from microdroplets. These studies indicate that the scattering of mode-locked pulses from droplets is not always linear for a train of high-intensity pulses.

Third-order nonlinear effects have proven useful for characterizing the bulk properties of microdroplets. To date, however, second-order effects [second harmonic generation (SHG), sum frequency generation, etc.] have not been investigated in microdroplets. While second-order effects are dipole forbidden in media with inversion symmetry, they are dipole allowed at the interface between two homogeneous media and are ideal for characterizing surfaces and interfaces. Many important processes in atmospheric and combustion chemistry occur at the surfaces of droplets. Detection of adsorbates on droplets could enable chemical reactions occurring on droplet surfaces in the environment to be characterized. Because of their surface sensitivity, second-order nonlinear effects could prove to be uniquely useful for such studies.

SHG from droplet surfaces coated with surfactant molecules was investigated. While the second harmonic intensity from pure water droplets was too weak to be detected, the second harmonic from droplets coated with surfactant molecules was easily detected. The surfactant molecules adsorbed on the surface of a hanging water droplet were observed to radiate second harmonic light in both the transmitted and specularly reflected directions when illuminated with mode-locked laser pules.

The hanging droplet generator was used to decrease the size of the droplet and, thereby, increase the density of the surfactant molecules, similar to the techniques used in the formation of Langmuir-Blodgett films. The number of surfactant molecules on the surface remained constant while the droplet surface area decreased. The increase in surfactant density resulted in an increase in the intensity of the SHG from the droplet surface. The SHG intensity was observed to vary as the inverse fourth power of the droplet radius. This functional form was consistent with the square dependence of the SHG intensity on the surfactant surface density and the inverse square dependence of the surfactant density on the droplet radius.

The density increase resulted in a reorientation of the surfactant molecules. The polarization dependence of the SHG intensity can be used to determine the approximate orientations of the surfactants. Before decreasing the droplet size, the surfactants were observed to be oriented with their axes nearly parallel to the droplet surface. After decreasing the droplet size, the surfactant molecules were observed to reorient towards the surface normal. This reorientational motion was consistent with the well established Langmuir-Blodgett film behavior. As the surface density increases in the formation of a Langmuir-Blodgett film, the molecules begin to interact until they form a close packed structure in which all of the molecules stand with their axes normal to the liquid surface. The limited range of droplet sizes available did not allow this close packed arrangement to be reached. A smaller degree of molecular reorientation was observed, however.

This work could ultimately be extended to investigate other second-order effects in droplets such as sum-frequency generation. Sum-frequency studies in which one of the frequencies is resonant with a vibrational mode of the surface molecules could ultimately be used to both detect and identify molecular species on the surfaces of droplets. Such studies, even in a laboratory environment, could allow chemical reactions occurring on droplet surfaces in the environment to be characterized and would advance our understanding of atmospheric chemistry.

AUGMENTATION AWARDS FOR SCIENCE & ENGINEERING RESEARCH TRAINING (AASERT) REPORTING FORM

The Department of Defense (DoD) requires certain information to evaluate the effectiveness of the AASERT Program. By accepting this Grant which bestows the AASERT funds, the Grantee agrees to provide 1) a brief (not to exceed one page) narrative technical report of the research training activities of the AASERT-funded student(s) and 2) the information requested below. This information should be provided to the Government's technical point of contact by each annual anniversary of the AASERT award date.

1. Grantee identification data: (R&T and Grant numbers found on Page 1 of Grant)

a. Yale University
University Name

b. F49620-94-1-0360
Grant Number

c. ____________
R&T Number

d. Richard K. Chang
P.I. Name

e. From: 15/7/96 To: 14/7/97
AASERT Reporting Period

NOTE: Grant to which AASERT award is attached is referred to hereafter as "Parent Agreement".

2. Total funding of the Parent Agreement and the number of full-time equivalent graduate students (FTEGS) supported by the Parent Agreement during the 12-month period prior to the AASERT award date.

a. Funding: $ 107,161

b. Number FTEGS: 2

3. Total funding of the Parent Agreement and the number of FTEGS supported by the Parent Agreement during the current 12-month reporting period.

a. Funding: $ 99,874

b. Number FTEGS: 1

4. Total AASERT funding and the number of FTEGS and undergraduate students (UGS) supported by AASERT funds during the current 12-month reporting period.

a. Funding: $ 36,371

b. Number FTEGS: 1

c. Number UGS: 0

VERIFICATION STATEMENT: I hereby verify that all students supported by the AASERT award are U.S. citizens.

Richard K. Chang
Principal Investigator

August 1, 1997
Date

Principal Investigator Annual Data Collection (PIADC) Survey Form

NOTE: If there is insufficient space on this survey to meet your data submissions, please submit additional data in the same format as identified below.

PI DATA

Name (Last, First, MI): Chang, Richard K.

Institution: Yale University

Contract/Grant No: F49620-94-1-0360 AASERT

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PUBLICATIONS RELATED TO AFOREMENTIONED CONTRACT/GRANT

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Include: Articles in peer reviewed publications, journals, book chapters, and editorships of books.

Do Not Include: Unreviewed proceedings and reports, abstracts, "Scientific American" type articles, or articles that are not primary reports of new data, and articles submitted or accepted for publication, but with a publication date outside the stated time frame

Name of Journal, Book, etc: Submitted to JOSA B

Title of Article: "Laser Induced Distortion for Increased Input Coupling of Light to Droplet-Cavity Modes".

Author(s): Justin Hartings, Xiaoyun Pu, Janice Cheung, Richard K. Chang

Publisher (if applicable): Optical Society of America

Volume: ____ **Page(s):** ____ **Month Published:** ____ **Year Published:** ____

Name of Journal, Book, etc: Submitted to Chem. Phys. Lett.

Title of Article: "Second Harmonic Generation and Fluorescence Images from Surfactants on Hanging Droplets"

Author(s): Justin Hartings, Andrew Poon, Xiaoyun Pu, and Richard K. Chang

Publisher (if applicable): Elsevier Scientific

Volume ____ Page(s) ____ Month Published ____ Year Published: ____

Name of Journal, Book, etc: Submitted to Applied Optics

Title of Article "Temporal beating; a technique to measure degeneracy-splitting of Azimuthal modes in nonspherical microstructures"

Authors: Justing M. Hartings, Janice L. Cheung and Richard K. Chang

Publisher (if applicable) Optical Society of America

Volume: ________ **Page(s):** ________ **Month Published:** ________ **Year Published:** ________

Name of Journal, Book, etc ________

Title of Article ________

Author(s) ________

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HONORS/AWARDS RECEIVED DURING CONTRACT/GRANT LIFETIME

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