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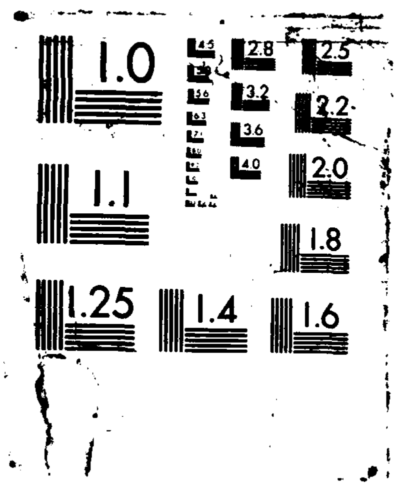
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This report describes work done under the cited ARO research grant. Experimental and theoretical results are described, papers and conference presentations resulting from this work are enumerated, and a list of personnel supported under this grant is provided.

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ATMOSPHERIC PROPAGATION OF LASER BEAMS

FINAL REPORT

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I. State of Problem Studied

The interactions of intense laser beams with small particles is an increasingly important field of study. In particular, atmospheric aerosols interact with propagating laser beams to produce a variety of significant effects. At low irradiance, aerosol scattering and linear absorption are the dominant interactions. As the irradiance increases, absorbing aerosols exhibit vaporization, ablation, shock-wave initiation and, ultimately, plasma formation. Transparent aerosols show little heating or vaporization, but internal field effects result in a variety of nonlinear optical phenomena. At sufficiently high intensity, transparent aerosols also initiate a plasma; however the mechanism of plasma formation is different than that for absorbing aerosols.

In work supported by this contract, experimental and theoretical investigations of aerosol interactions with intense laser beams have been carried out. Experimental studies of elastic and stimulated Raman scattering, plasma spectroscopy and photo-thermal spectroscopy were conducted on nominally transparent droplets. Theoretical modeling of laser-aerosol interactions emphasized aerosol heating, vaporization, hydrodynamic effects and plasma initiation. In collaboration with colleagues at Los alamos National Laboratory, an atmospheric propagation code has also been developed incorporating many of these interactions.

A number of important experimental and theoretical findings have resulted from work performed under this contract. In addition, more than 30 publications and conference papers have reported this work. Finally, contract funds have provided support for research personnel. These are discussed in the following paragraphs.

II. Summary of Most Important Results

II. A Experimental Results

The experimental phase of this work has led to several important findings in the area of aerosol properties and aerosol-beam interactions. These experimental results are conveniently grouped under four major headings which are discussed in the following paragraphs.

II. A. 1. Plasma Studies

An extensive investigation of plasmas initiated by a high-irradiance laser beam on aerosols was conducted. Both individual, levitated liquid and solid droplets, and streams of quasi-monodispersed liquid droplets from a vibrating orifice generator were used in these experiments. Plasmas were initiated at the aerosols when they were irradiated with a Q-switched (7 ns FWHM) Nd:YAG laser operating at either the fundamental frequency of 1.06μm or in one of the harmonic frequencies of .532, .353, or .266 μm. Plasma spectra were obtained using a 0.5-m scanning monochromator configured as an optical multichannel analyzer (OMA) by the addition of a photodiode array detection system, and a transient waveform digitizer was employed to obtain temporal information of the developing plasma.

Breakdown threshold measurements of droplet-initiated plasmas were obtained as a function of several droplet-beam parameters, notably droplet size and composition, and laser wavelength. These experiments revealed the importance of multiphoton ionization in initiating plasmas at lower laser wavelengths. In particular, the measured wavelength dependence of the breakdown threshold laser irradiance $I_{BD} \sim \lambda^{0.5-1.0}$, characteristic of multiphoton initiation may be compared with the wavelength dependence $I_{BD} \sim \lambda^{-2}$ obtained by other workers at longer (10.6μm) wavelengths where cascade ionization is the dominant mechanism.

The location of the initial breakdown site in transparent droplets is a topic of some controversy. Our measurements reveal that the initial breakdown site occurs outside the droplet when the surrounding medium has a low ionization potential and inside the droplet when the surrounding medium has a high ionization potential. In each case, the initial breakdown site is observed at one of the well-known "hot spots" of enhanced field strength that occur in transparent droplets irradiated by intense laser radiation.

Time-resolved plasma spectra of irradiated droplets revealed the time history of the plasma as well as providing a measure of important plasma properties such as the temperature and electron density. Our measurements revealed that droplet plasmas were significantly hotter than corresponding plasmas initiated on absorbing surfaces. The measurements also provided for the first time a detailed time history of ionic and neutral species emission for droplet-initiated plasmas.

II.A.2. Stimulated Raman Scattering

The presence of intense nonlinear optical effects in micron-sized, transparent liquid droplets has been known for several years. In particular, multi-order stimulated Raman scattering (SRS) has been observed in small droplets indicating the presence of high-order nonlinear susceptibilities. In contrast to similar effects occurring in bulk media, there is a significant amount of evidence indicating that SRS emission in small droplets occurs at morphology-dependent resonance (MDR) frequencies. Our measurements of SRS spectra in droplets concentrated on the temporal development of the SRS emission and on the spectral positions of the sharp, SRS peaks. We showed for the first time that the time-dependence of the SRS emission is not coincident with the temporal pulse shape of the exciting laser radiation. There is a delay ~ 5-7 ns between the peak of the laser light and the corresponding peak of the SRS emission. We attribute this delay to the time necessary to build up the MDR resonance modes responsible for the observed SRS emission. Spectral studies of the SRS emission, obtained with the aid of the OMA, revealed conclusively for the first time that SRS emission occurs at MDR frequencies associated with internal droplet fields. The wavelength separation between SRS peaks were fit directly (with no adjustable parameters) to a theoretical expression for the separation between MDR resonances. Excellent agreement between our experimental data and the theoretical model was obtained. Current experimental work on this problem is concerned with the angular dependence of the SRS emission. Some indication of a resonance structure has been seen in preliminary measurements.

II.A.3 Photothermal Diagnostics

A photothermal diagnostic system was constructed in order to probe optical perturbations in the neighborhood of intensely irradiated aerosol droplets. Either individual, levitated droplets or a stream of quasi-monodisperse droplets from a droplet generator were perturbed by a YAG laser. A HeNe laser monitored photothermal perturbations. To date, only breakdown - related perturbations have been studied. Sub-breakdown, thermodynamic and hydrodynamic interactions will be probed at a later date.

Following plasma initiation on liquid droplets, we have observed a variety of optical perturbations of the HeNe beam. These perturbations occur over time scales ranging from 10^{-9} sec (the shortest time scale we can measure) to 10^{-3} sec. The earliest-occurring phenomena are associated with either hydrodynamic shock waves or super detonation waves generated by the plasma emission. These are followed by a relaxation process during which the shock-heated air returns to near-ambient conditions. Thermal diffusion processes occur on much longer (10^{-3} sec) time scales. We have been able to partially model the optical perturbations using Gaussian optics techniques. We continue to work on this problem.

II.A.4. Droplet Evaporation

Following construction of the optical levitation apparatus, individual droplets of glycerine or a water-glycerine mixture were successfully levitated for long periods of time (up to 36 hours). The argon-ion levitating laser also provided an intense source for observing Mie scattering from the levitated droplets. We observed a train of MDR's in the scattered light as the size parameter changed due to droplet evaporation. This technique provides an extremely high resolution probe of droplet evaporation rates. It will also be useful in the investigation of laser evaporation of droplets in the diffusive evaporation regime where hydrodynamic effects do not perturb the droplet evaporation process.

II.B Theoretical Results

Theoretical work performed under this contract has resulted in a number of important results in the area of aerosol-beam interactions. These results, which are conveniently grouped under three major headings, are discussed in the following sections.

II.B.1. Hydrodynamics

The interactions of an aerosol particle with an intense laser beam include a variety of hydrodynamic effects. We have developed a general hydrodynamic model that is capable of describing spherically symmetric laser heating of aerosol droplets. Diffusion was included in this model so the model's capability extended from the low-irradiance regime where diffusive vaporization and thermal conduction are the dominant mass and energy transport processes, respectively, to the high-irradiance regime where internal ablation and plasma formation become important. For example, for water droplets at 10.6 μm, the regime of our model's applicability extends to approximately 10^8 W/cm^2. We are currently extending our model to include plasma formation.

II.B.2 Photothermal Spectroscopy

In our photothermal measurements, the medium surrounding aerosol droplets irradiated by an intense pulse from a YAG laser was probed by a HeNe beam. Perturbations of the medium alter the refractive index and, hence, modulate the transmission of the HeNe probe beam. Using the well-known formalism of Gaussian optics, we have obtained expressions for the transmitted probe beam intensity as a function of the refractive index of the medium. The remaining problem is to relate the thermal perturbations of the medium to refractive index changes. We have developed a simple hydrodynamic model that accomplishes this, and we are currently in the process of refining this model.

II.B.3. Irradiated Droplet Field Calculations

Internal and external fields have been calculated for irradiated water droplets. We have found enhancement factors of the electromagnetic energy density of the order of 10^6 when the fields correspond to one of the MDR's of the droplet. These results will be extremely useful in developing a theory of SRS (SRS experimental results have been discussed in Section IIA.2).

III. List of Publications and Papers

A number of publications and conference papers resulted from work supported by this contract. These are given in the following list.

III. A. Publications

R.L. Armstrong and A. Zardecki, "Vaporization of Irradiated Droplets," Phys. Fluids, 29, 3573 (1986).

P. Chylek, J.D. Pendleton, and R.G. Pinnick, "Internal and Near-surface Scattered Field of a Spherical Particle at Resonant Conditions", Appl. Opt. 24, 3940 (1985).

P. Chylek, M. Jarzembski, N. Chou, R. Pinnick, "Effect of Size and Material of Liquid Spherical Particles on Laser-Induced Breakdown," Appl. Phys. Lett 49, 1475 (1986).

A. Biswas, H. Latifi, P. Shah, L.J. Radziemski, and R.L. Armstrong, "Time-Resolved Spectroscopy of Plasmas Initiated on Single, Levitated Aerosol Droplets, "Opt. Lett. 12, 313 (1987).

R.L. Armstrong, "Laser-Induced Droplet Heating," Chapter for book on Laser-Droplet Physics edited by P.W. Barber and R.R. Chang, World Scientific Publicing Company, 1988.

R. L. Armstrong and A. Zardecki, "Diffusive and Convective Evaporation of Irradiated Droplets," J. Appl. Phys. 62, 4571 (1987).

P. Chylek, M. Jarzembski, V. Srivastava, R. Pinnick, D. Pendleton, and J. Cuncleton, "Effect of Spherical Particles on Laser-Induced Breakdown of Gases," Appl. Opt. 26, 760 (1987).

R.G. Pinnick, A. Biswas, P. Chylek, R.L. Armstrong, H. Latifi, E. Creegan, V. Srivastava, M. Jarzembski, and G. Fernandez, "Stimulated Raman Scattering from Micron-Sized Droplets: Time Resolved Measurements, Optics Letters (in press)

A. Biswas, H. Latifi, L.J. Radziemski, R.L. Armstrong, "Irradiance and Laser Wavelength Dependence of Plasma Spectra from Single Levitated Aerosol Droplets, Applied Optics, (in press).

III. B. Conference Presentations

■ Papers given at 1985 CRDC Conference, June, 1985.

"On the Propagation of Intense Optical Pulses through Vaporizing Aerosols", R.L. Armstrong, A. Zardecki and S. A. W. Gerstl.

"Surface Waves, Structural Resonances, and Surface Heating of Spherical Particles", P. Chylek.

■ Paper given at 1986 CRDEC Conference, June 1986

"Hydrodynamic Effects in Evaporating Droplets," R.L. Armstrong and A. Zardecki

"The Effect of Size and Size and Refractive Index of Liquid Spherical Particles on Laser-Induced Breakdown," P. Chylek, M. A. Jarzembski, and N. Y. Chou.

■ Invited paper presented at the International Conference on Millimeter Wave Propagation and Scattering in the Atmosphere, Florence, Italy, 1986.

"Propagation of Intense Light Beams in the Presence of Hydrodynamic Aerosol-Beam Interactions," R. L. Armstrong

■ Paper given at International Conference on Lasers, '85, Las Vegas, Nevada, December, 1985.

"Hydrodynamics of Evaporating Aerosols Irradiated by Intense Laser Beams," R. L. Armstrong, A. Zardecki, and S.A.W. Gerstl.

■ Papers presented at Workshop on the Interactions of Electromagnetic and Particle Beams with the Atmosphere," Las Cruces, NM, January, 1986.

"Spectroscopy of Laser Induced Plasmas," L. Radziemski

"Laser-Induced Evaporation of Liquid Droplets Containing Small Carbon Particles," H. Latifi, P. Chylek, R. L. Armstrong, Srivastava, J. Jernigan and R. G. Pinnick.

"Hydrodynamics of Intensely Irradiated Aerosols," R. L. Armstrong and A. Zardecki.

"On the Prediction of Contrast Transmission in Aerosols from the Angular Distribution or Radiation Received from a Point Source," G. Goedecke.

"Scattering of Electromagnetic Radiation by Inhomogeneous Targets," V. Srivastava, P. Chylek, and R. G. Pinnick.

"Effect of Aerosols on Laser-Induced Plasmas," N. Chou, P. Chylek and R. G. Pinnick.

- Papers presented at Fourth Workshop on the Physics of Directed Energy Propagation in the Atmosphere, Las Cruces, NM, January, 1987.

"Detection of Photothermal Signals Caused by Laser Induced Plasmas," L. Radziemski and A. Biswas.

- Paper presented at CLEOS in Baltimore, MD, April 1987.

"Time Resolved Spectra of Laser Induced Plasma on Single Aerosols," A. Biswas, H. Latifi, R. L. Armstrong, and L. J. Radziemski.

- Papers presented at 1987 CRDEC Conference, June, 1987.

"Experimental Effects Following Immediately After the Formation of Laser-Induced Breakdown Plasma," L. J. Radziemski and A. Biswas.

"Time Resolved Spectra of Plasmas Initiated by Single Aerosols: An Update," L. J. Radziemski, R. L. Armstrong, A. Biswas, and H. Latifi.

"Energy Balance in Laser Irradiated Vaporizing Droplets," A. Zardecki and R. L. Armstrong.

"Pressure Dependence of Laser Induced Breakdown of Aerosols and Gases," P. Chylek, M. Jarzembski, V. Srivastava, and R. G. Pinnick.

"Aerosol-Induced Breakdown: Wavelength Dependence," R. G. Pinnick, P. Chylek, M. Jarzembski, E. Creegan, V. Srivastava, and J. Cruncleton.

"Pressure Dependence of Laser Induced Breakdown of Aerosols and Gases," P. Chylek, M. Jarzembski, V. Srivastava, and R. G. Pinnick.

"Poynting Flux Calculations for Small Droplets," D. Pendelton, R. G. Pinnick, and P. Chylek.

■ Invited paper presented at NATO Advisory Group for Aerosol Research and Development, Rome, Italy, May 1987.

"Non-linear Effects on the Atmospheric Transmission of High Flux Electromagnetic Beams," R. L. Armstrong.

IV. Personnel Supported

The people who received support from this contract were:

■ Principal Investigators:

R. L. Armstrong, P. Chylek, L. J. Radziemski

■ Post-Doctoral Fellows:

A. Biswas, N. Y. Chou

■ Graduate Students:

G. Garwood, M. Jarzembski, H. Latifi, P. Shah,
V. Srivastava, R. S. Wong

■ Research Associate:

J. Jernigan

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