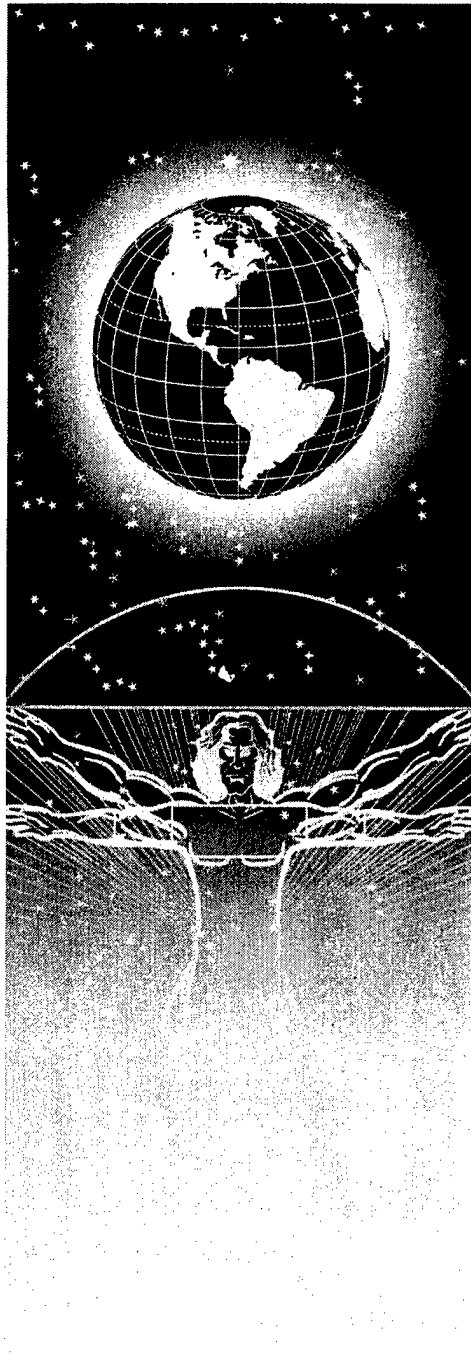


UNITED STATES AIR FORCE
RESEARCH LABORATORY



Measurement of Quantitative IR
Properties of Single Aerosol
Particles with Emphasis
On Biological and Chemical
Stimulants

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September 2004
Interim Report - June 2003 – June 2004

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This technical report has been reviewed and is approved for publication.

FOR THE DIRECTOR

//signed//

STEPHEN R. CHANNEL, DR-IV
Director, AF CBD Tech Base Programs
Air Force Research Laboratory

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13. ABSTRACT (Maximum 200 words)
A simplified experiment in the visible range using 2 wavelengths (one in the absorption band and the other outside the absorption band) was performed initially. We then shifted to the mid-IR (3.41 μm) using a mixture of H₂O and D₂O, with the latter being the absorber. Again the peak-to-valley ratio increased upon the addition of D₂O into H₂O. The angular distribution was collected on the backward scattering direction using an ellipsoidal mirror. The scattered light angle spanned from 0° ≤ φ ≤ 360° and 90° ≤ θ ≤ 163°.

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Measurement of Quantitative IR Properties of Single Aerosol Particles with Emphasis on Biological and Chemical Stimulants

June 2003 to June 2004

Research Participants:

From Yale – Kevin Aptowicz, Yong-Le Pan, and Richard K. Chang

From ARL – Ron Pinnick, Steve Hill, Kris Gurton, and Richard Tober

From Lincoln Labs – Anish Goyal and Tom Jeys

From AFRL – Burt Bronk

Elastic light scattering is being investigated as a means to extract single particle absorption. In particular, the technique labeled TAOS (Two-dimensional Angular Optical Scattering) detects the angularly resolved elastically scattered light from a particle. These TAOS patterns are sensitive to a particle's morphology (size, shape, structure, and complex refractive index) and therefore can be used to extract information on particle absorption.

Previous work has been done in the visible (532 nm), where the TAOS patterns were successfully shown to vary considerable when the absorptivity of a droplet varied. This matched expectations drawn from Mie theory. The next step was to move the excitation wavelength into the mid-IR where strong absorption bands exist due to molecular vibrational modes.

Two separate experiments were conducted. The first utilizes a lens in the Abbé sine condition to detect the TAOS pattern of single droplets. These TAOS patterns are then compared to Mie theory predictions to deduce the imaginary index from the best-fit. A second experiment was conducted where TAOS patterns at two different wavelengths were simultaneously detected from a single aerosol particle. This dual TAOS patterns were collected over a large solid angle encompassing almost the entire back hemisphere of scattered light. Both systems detect the scattered light in-situ and in real-time.

A simplified schematic of the first experimental arrangement to extract the imaginary part of the refractive index of a spherical particle is shown in figure 1. A liquid dispenser system based on piezo technology generated droplets from an exit orifice (41 μm diameter) at a rate of ~ 500 Hz. The droplet diameter varied from 54 μm to 57 μm . The droplets were composed of three different solutions: distilled water (H_2O), deuterium oxide (D_2O), and a 50%-50% mixture of H_2O and D_2O .

The laser source was a type-II InAs/GaInSb interband cascade laser operating in CW mode to generate light at a wavelength of 3.41 μm with optical powers approaching 40 milliwatts. An $f/2$ CaF_2 plano-convex lens collimated the scattered light onto the detecting focal plane array. The angular range of collected light spanned 23° from $\theta = 23.5^\circ$ to $\theta = 46.5^\circ$. However, because of severe spherical aberration in the outer limits of the angular range the practical collection range was 26° to 42° . These effects were

mitigated by performing an exact ray-trace on the system calculated in Matlab®. The detector, an InSb 320x256 focal-plane-array, was run at a frame rate of 63.13 Hz with an integration window of 217 μs so that only a single scattering event would be detected. Three TAOS patterns from the three different solutions are shown in Figure 2. The effects of spherical aberration can be noted by the increase in intensity around the perimeter of the TAOS pattern. Also, the TAOS patterns have different overall signal intensities due to both the effects of particle absorption as well as fluctuations in laser intensity and detected background noise. Thus the data does not lend itself to making an absolute comparison of intensity between data sets.

A cross-section of the TAOS pattern is plotted to the right of each image. The experimental data shows a good match with numerical data based upon Mie theory, although again the effects of spherical aberration can be seen at the edges of the plotted angular range. The input parameters to calculate the Mie theory curve were both experimental measured, as was the case for the droplet diameter, and taken from literature, as was the case of for the complex refractive index. The table below summarizes the input parameters for the different solutions. To calculate the refractive index of H₂O-D₂O an average was taken of the H₂O data and D₂O data. As one final note, due to variability in particle location, a precise value for the collection angle can not be known. Therefore, overall angle range was slightly shifted ($\sim 1^\circ$) from the measured values to fit the data to theory.

Solution	Complex Refractive Index (at 3.41 μm)	Droplet Diameter (μm)
D ₂ O	1.279 + i 0.0021	55.176
H ₂ O + D ₂ O	1.342 + i 0.0098	54.254
H ₂ O	1.405 + i 0.0176	57.401

Table 1: Mie theory input parameters

A computer program was written in hopes of extracting the particle's absorptive properties from the raw data. However, due to the noise level, spherical aberration distortions, as well as uncertainty in the angle, the computer analysis was unable to find a strong match to the data set. On the other hand, it is believed that with increase in signal-to-noise ratio as well as a better calibration routine for defining the scattering angles, the complex refractive index, and hence the absorption, can be inferred.

The scheme to collect TAOS images at two wavelengths simultaneously is shown in figure 3. The two laser sources were optically pumped GaSb-based semiconductor lasers with type-II InAs/InGaSb quantum well gain regions emitting at 3.9 μm and 5.1 μm with a peak power of ~ 0.4 watts and pulse duration of 100 μs . Both wavelengths are relatively transparent in H₂O, but D₂O has relatively high absorption at 3.9 μm while being transparent at 5.1 μm . Beam shaping optics (spherical and cylindrical lenses) as well as an F/3.75 CaF₂ focusing lens were utilized to achieve a desired spot size of 50 μm x 500 μm where the major axis is perpendicular to the propagation direction of the droplet. An ellipsoidal mirror collected the backward hemisphere of scattered light ($0^\circ \leq \phi \leq 360^\circ$, $90^\circ \leq \theta \leq 163^\circ$) and focused it through a spatial filter located at the ellipsoid's second

focal point. To reduce aberrations effects, an F/1 ZnSe aspheric lens collimated the scattered light, after which the two wavelengths were separated via a dichroic mirror. Finally, a bi-convex F/1 CaF₂ lens coupled the scattered light onto the InSb detectors. Droplets from three different mixtures of H₂O-D₂O were analyzed: 100%-0%, 75%-25%, and 50%-50%. The experimentally collected data, as well as numerical simulations based on Mie theory, are shown in figure 4. Each column from left to right indicates a different H₂O-D₂O droplet composition, as labeled. TAOS patterns (row I) collected at 3.9 μm show the effects of increasing the concentration of D₂O that leads to an increase in absorption. These patterns qualitatively match the numerical simulations based on Mie theory (row II). TAOS patterns (row III) are collected simultaneously at 5.1 μm from the same single droplets. These patterns also agree with predictions from Mie theory in which there was a slight increase in scattering intensity of the central region. By comparing the data in row I with row III, it is clear that one could distinguish between droplets of pure H₂O and droplets that contain a considerable amount of D₂O by comparing the angular scattering patterns.

As was done in the visible, TAOS patterns of dry aggregate particles were also captured as is shown in figure 5. Due to the low pulse energies of the mid-IR laser, it was difficult to analyze the data since the scattered signal was just above the noise level. However, certain features can be noticed such as the quasi-ring shaped scattering patterns for the Tryptophan and Bovine Albumin particles. This contrasts the scattering patterns seen in the visible which are primarily made up of island like features. The reason for these ring-like patterns is the shift in wavelength to a size comparable to the particle diameter. Thus the laser light is no longer sensitive surface roughness of the particle, rather just its overall shape.

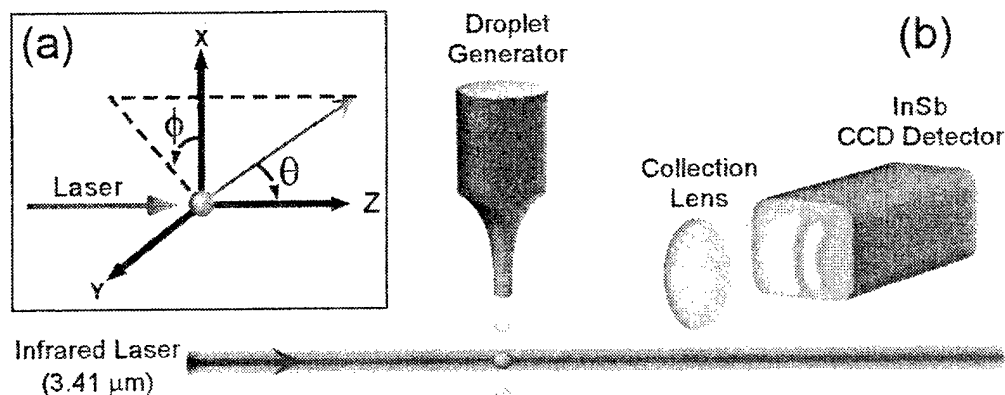


Figure 1: Experimental Set-up

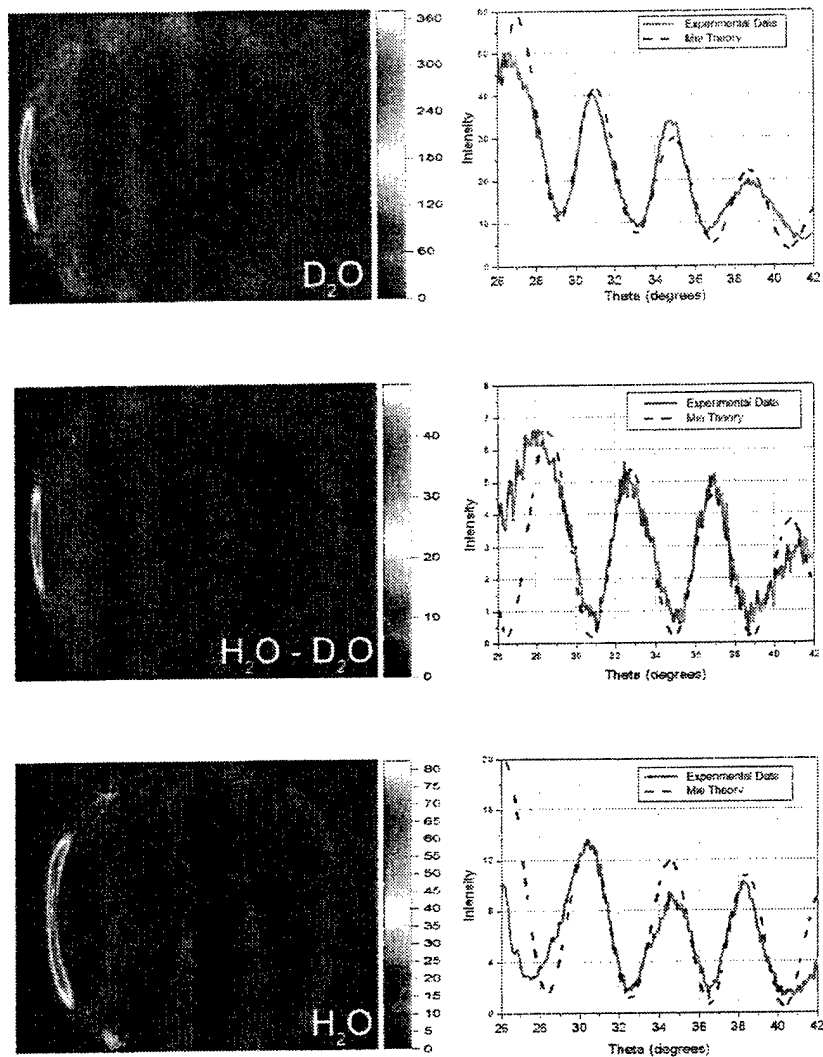


Figure 2: Left, TAOS patterns collected from droplets of D₂O, 50/50 H₂O-D₂O mixture, and H₂O. Right, horizontal cross-sections of the TAOS patterns are compared with Mie theory results.

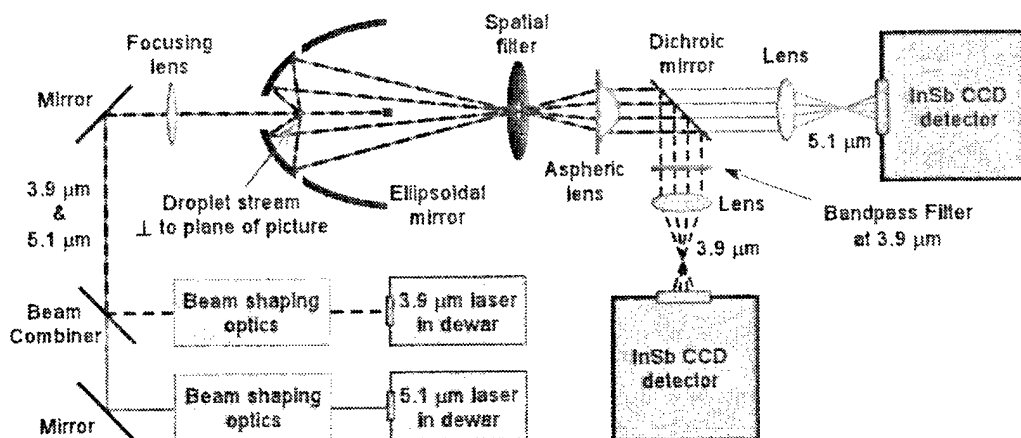


Figure 3: Experimental set-up to collect simultaneous TAOS patterns at two wavelengths.

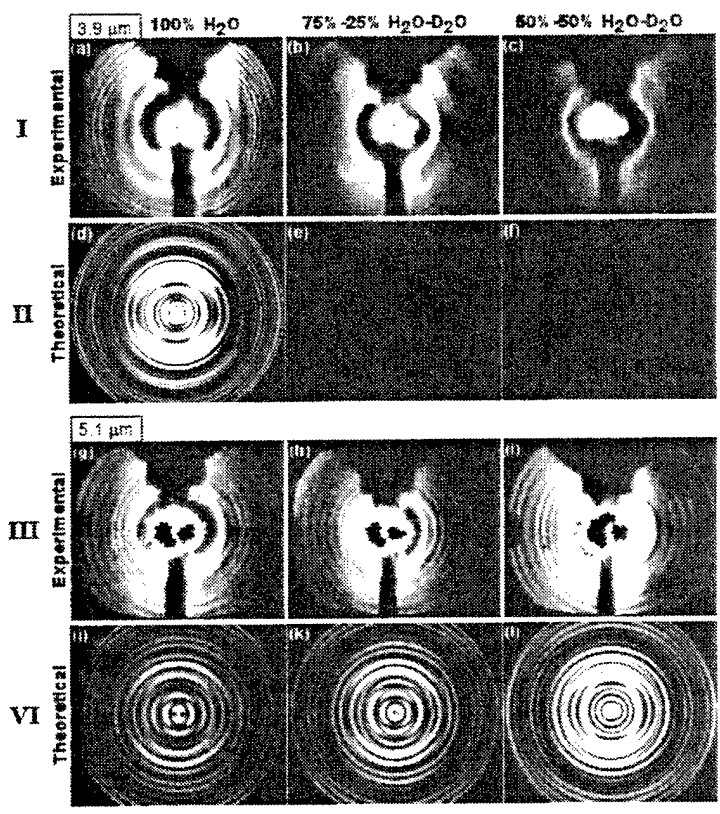


Figure 4: (Row I) TAOS pattern at $\lambda = 3.9 \mu\text{m}$ detected from a single droplet (diameter $\sim 55 \mu\text{m}$) composed of (a) H_2O , (b) 75%-25% $\text{H}_2\text{O}-\text{D}_2\text{O}$, and (c) 50%-50% $\text{H}_2\text{O}-\text{D}_2\text{O}$. (Row II) Corresponding numerical simulations of row I based upon Mie theory. (Row III) TAOS pattern collected simultaneously at $5.1 \mu\text{m}$ from the same single droplets as row I. (Row IV) Corresponding numerical simulations of row III based upon Mie theory.

**Simultaneously Captured
Dual TAOS patterns of 8 μm aerosols**

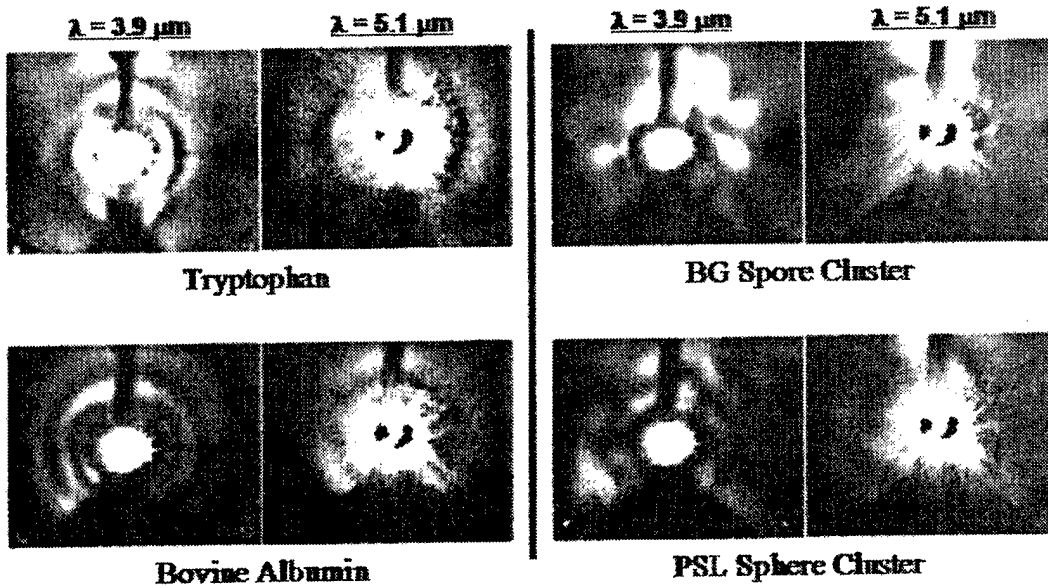


Figure 5: TAOS patterns captured simultaneously of dry aggregate particles.

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Two-dimensional angular optical scattering patterns in the mid-infrared of microdroplets: on and off absorption, K.B. Aptowicz, Y.L. Pan, and R.K. Chang, R.G. Pinnick, S.C. Hill, R.L. Tober, B.V. Bronk, to be published in September issue of Optics Letters.

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Fudan University, May 11, 2004, Shanghai, China

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