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RPPR Final Report
as of 18-Nov-2019

Agency Code:

Proposal Number: 66664CH **Agreement Number: W911NF-16-1-0483**

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STEM Degrees: 2 **STEM Participants:** 3

Major Goals: The goal of this project is to better understand physics and chemistry of aerosol particles at the fundamental level (single aerosol particle). In particular, we investigate single aerosol properties and their changes over time under external chemical agitation, i.e. via surface chemical reactions, leading to new insights into aerosol particle reactivity and surface chemistry. Four research objectives are
(1) Trap chemical (e.g. solid oxide particles) and biological particles (pollen and spores) based on different trapping forces. Two major trapping schemes using a single laser beam or double beams will be employed to trap single particles in a closed reaction chamber or in a free space.
(2) Achieve chemical agitations on steadily trapped single aerosol particles in air through exposing gas phase molecules or droplets.
(3) Conduct physical characterization on the trapped particle's size, surface structure, and momentum transfer using real-time imaging.
(4) Investigate chemical variations of the trapped particles in air under agitations using OT-RS as the primary technique. Chemical compositions, thermodynamics, spectral fingerprints of some chemical function groups, hygroscopicity, and their viability will be investigated. Temporal behavior of the trapped particle's properties under chemical agitations will also be studied.

Accomplishments: A. Major activities
1. Introduced the new concept of the universal optical trap (UOT) which can trap particles of arbitrary physical and chemical properties in different environments such as in a solution and in air. UOT can trap particles stably for up to many hours for subsequent characterization, measurement, as well as even observation of chemical reactions.
2. Integrated the UOT with Raman spectroscopy (OT-RS) and the imaging technique to study single particle's physical and chemical properties as well as their time-dependent dynamics.
3. Integrated the UOT with cavity ringdown spectroscopy (OT-CRDS) and the imaging technique to measure single particle's extinction in air at different wavelengths in the UV spectral region.
4. Explore external chemical agitation to a trapped particle and observe the reaction dynamics. Observed the first chemical reaction on a single trapped particle in air under controlled environment.
5. Explore the position- and time-resolved Raman spectroscopy within a single trapped particle and study phase separation and dynamics of the single particle composed of different chemical components.
6. Train graduate and undergraduate students and disseminate research results in a broader research community.

B. Accomplishment highlights

RPPR Final Report
as of 18-Nov-2019

In this project, we have achieved the following highlighted accomplishments:
1. The project generated 16 peer-reviewed journal publications, of which 2 are highlighted in the front covers of Applied Physics Letters and Chemical Physics Letters.
2. The project delivered 20 conference presentations.
3. The project graduated 1 PhD student and trained 2 undergraduate students and trained 1 MS student.
4. The PI with his project mentor/collaborator in ARL co-organized the first special symposium on "single particle studies" in 2017 American Aerosol Association Research conference.
5. Developed and demonstrated the first-of-its-kind universal optical trap Raman spectroscopy (OT-RS) system that allows us to study time-dependent single particle (of arbitrary chemical and physics properties) properties and its reaction dynamics with or without an introduction of an external reactive agent (gas, vapor, radicals, etc). This system has become a new technology platform to study single particle (formation, composition, reaction, surface chemistry, phase separation, etc.).
6. Integrated the UOT with CRDS (OT-CRDS) for single particle extinction measurements in tunable UV spectral region.
7. Bioaerosol particle Raman is often notoriously interfered by strong fluorescence from the particle. This fact hinders the bioaerosol particle study using the Raman spectroscopy technique. We observed for the first time the effective and fast fluorescence quenching when a bioaerosol particle is trapped in the special universal optical trap. The part of the observations is published and highlighted in the front cover of Chemical Physics Letters.
8. We reported for the first time the high-resolution time- and position-resolved Raman spectra within a single optically trapped liquid droplet that consists of different organic components.
9. Demonstrated the first observation of chemical reaction in a single trapped airborne particle under controlled environment, which is published in Aerosol Science and Technology Letters (top 10 most cited article since its publication in July).

Training Opportunities: One PhD student has received training in this project. He graduated in May 2019. Now he is working as a senior research engineer in Wyatt Technology (California). One undergraduate student has received training in research experience in the fall semester of 2017 and the spring semester 2018. This student graduated with a B.S. degree in physics. Now he is working as a research associate in Naval Oceanographic Office. Another undergraduate student is trained in this project since the summer of 2019. Five other graduate students, who are working on other projects in the PI's groups, have benefited from the research discussion and exposure to the research work in the lab.

Training and Development:

Graduate student:
Mr. Zhiyong Gong is the PhD student who graduated from his PhD research in this project. He has committed 100% of his research time to this project. His research efforts are focused on the integration of the newly developed universal optical trap (UOT) with other measuring techniques such as imaging, scattering, cavity ringdown spectroscopy and Raman spectroscopy. He has been conducting measurements of trapped particles with introduction of moisture and ozone. He has been trained in particle trapping, measurements, writing a manuscript, etc. He plays an important role in the experimental work in this project. He has become an expert in optical trapping and spectroscopic measurements of single particles.

Undergraduate student:
Mr. Jeff Headley was a senior majoring in physics during the project report period. He is interested in gaining research experience in lasers, spectroscopy, optical trapping, sample handling, etc. He has been trained with basic knowledge of lasers and CRDS. He graduated with a B.S. degree in physics and he now joins the Naval Oceanographic Office as a research scientist.

Mr. Cameron Gaito: A senior physical major. He joined the PI's research group obtain research experience in laser and optical trapping, and spectroscopy. This research experience will be benefit him for his planned graduate study.

Results Dissemination: The project outcomes and research results are effectively and timely disseminated through peer-reviewed journal publications, conference presentations, and invited seminars, giving lab tours to visitors on and outside campus, project research site visit, project reporting, and the information posted in the PI's Research Group web site. Part of the activities is listed below:

- Via 16 peer-reviewed journal publications including two featured in the front cover of prestigious journals.
- Via 20 presentations in national and international conferences.
- Via the PI's research group web site, http://wang.physics.msstate.edu/index.php/en/

Honors and Awards: 1. The PI was elected as Chair of Physics Working Group in American Aerosol Associate Research (AAAR) for 2018-2019 period.

2. The PI with his project collaborator/mentor Dr. Yongle-Pan (ARL Fellow) recognized a special symposium on Single Particle Studies in 2017 AAAR conference. This honor is directly related the outstanding accomplishments from this project.

3. The PI has been recommended as a plenary speaker in 2020 AAAR conference with a tentative topic of New insight into single particles at the fundamental level.

4. Two publications directly generated from this project are highlighted in the from cover of journals.

5. One article directly from this project is "Top-10 most downloaded articles" in Aerosol Science and Technology since its publication in July 2019 to date (as of this report is written).

6. One invited review article is published in Journal of Quantitative Spectroscopy and Radiative Transfer (2018).

7. One invited topic review article is being written for Aerosol Science and technology.

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: Graduate Student (research assistant)
Participant: Zhiyong Gong
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Undergraduate Student
Participant: Jeff Headley
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Research Experience for Undergraduates (REU) Participant
Participant: Cameron Gaito
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:

International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co-Investigator
Participant: Yong-Le Pan
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co-Investigator
Participant: Gorden Videen
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Final Report on W911NF1610483

Annual Report Period: 9/01/2016 – 08/31/2019

Submitted on: 11/29/2019

Agreement Number: W911NF1610483

Principal Investigator: Chuji Wang

Organization: Mississippi State University

Program Manager: Dr. Robert Mantz

Submitted By: Chuji Wang - Principal Investigator

Title: Investigation of chemical agitations on single chemical and biological aerosol particles in air using optical trapping-Raman spectroscopy

What are the research goals of the project?

The goal of this project is to better understand physics and chemistry of aerosol particles at the fundamental level (single aerosol particle). In particular, we investigate single aerosol properties and their changes over time under external chemical agitation, i.e. via surface chemical reactions, leading to new insights into aerosol particle reactivity and surface chemistry. Four research objectives are

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What was accomplished under these goals?

(Period: 09/01/2016 – 08/31/2019)

A. Major activities

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5. Explore the position- and time-resolved Raman spectroscopy within a single trapped particle and study phase separation and dynamics of the single particle composed of different chemical components.

6. Train graduate and undergraduate students and disseminate research results in a broader research community.

B. Accomplishment highlights

In this project, we have achieved the following highlighted accomplishments:

1. The project generated **15** peer-reviewed journal publications, of which **2** are highlighted in the front covers of Applied Physics Letters and Chemical Physics Letters.

2. The project delivered **20** conference presentations.

3. The project graduated **1** PhD student and trained **2** undergraduate students and trained **1** MS student.

4. The PI with his project mentor/collaborator in ARL co-organized the first special symposium on "single particle studies" in 2017 American Aerosol Association Research conference.

5. Developed and demonstrated the first-of-its-kind universal optical trap Raman spectroscopy (OT-RS) system that allows us to study time-dependent single particle (of arbitrary chemical and physics properties) properties and its reaction dynamics with or without an introduction of an external reactive agent (gas, vapor, radicals, etc). This system has become a new technology platform to study single particle (formation, composition, reaction, surface chemistry, phase separation, etc.).

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9. Demonstrated the first observation of chemical reaction in a single trapped airborne particle under controlled environment, which is published in Aerosol Science and Technology Letters (top 10 most cited article since its publication in July).

C. Significant results

1. Characterization of single airborne particle extinction using the tunable optical trap-cavity ringdown spectroscopy (OT-CRDS) in the UV

We integrated a rigid optical trap into a tunable pulsed cavity ringdown spectroscopy (OT-CRDS) system to characterize the extinction of single airborne particles in the UV spectral region (306-315 nm). Single solid particles from multi-walled carbon nanotube (MWCNT), Bermuda grass smut spore, carbon microsphere, or blackened polyethylene microsphere were trapped in air based on the photophoretic force. The improved OT-CRDS system was highly sensitive and able to resolve extinctions of single particles from different materials and sizes at a given wavelength. Further, we successfully manipulated the number of particles, e.g., 1, 2 or more particles, in the trap and measured their distinguishable extinctions using the OT-CRDS. We also show that the particle size and extinction have a good linear correlation from the measurements of 24 single MWCNT particles. Material- and wavelength-dependent extinctions of the four types of airborne particles were also characterized. Results reveal that single airborne particles regardless of their differences in material and size, due to their heterogeneous morphology, have *individual-particle* dependent extinctions and that dependence can be resolved and characterized using the OT-CRDS technique.

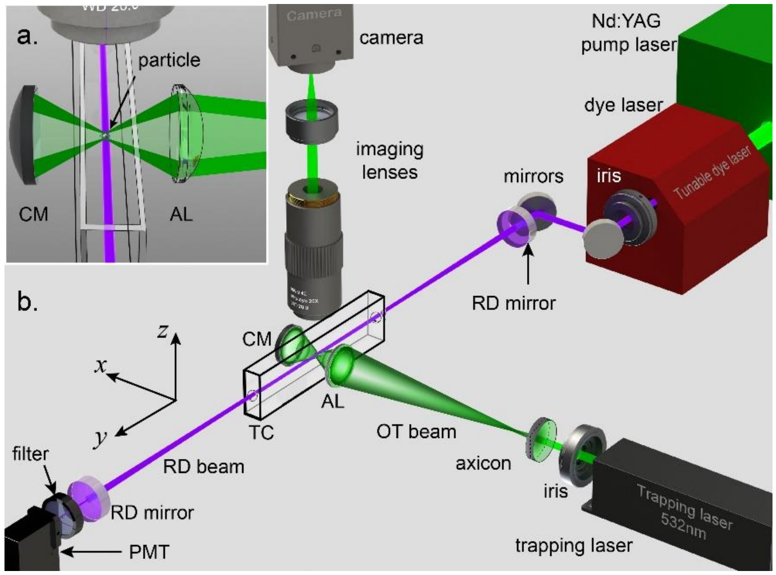

Fig. 1. Experimental scheme of the improved optical trap-cavity ringdown spectroscopy (OT-CRDS) system. CM: concave mirror, AL: aspheric lens, RD mirror: ringdown mirror, RD beam: ringdown beam, OT beam: optical trapping beam, TC: trapping cell, PMT: photomultiplier tube. The inset figure illustrates the detailed optical field near the trapping zone.

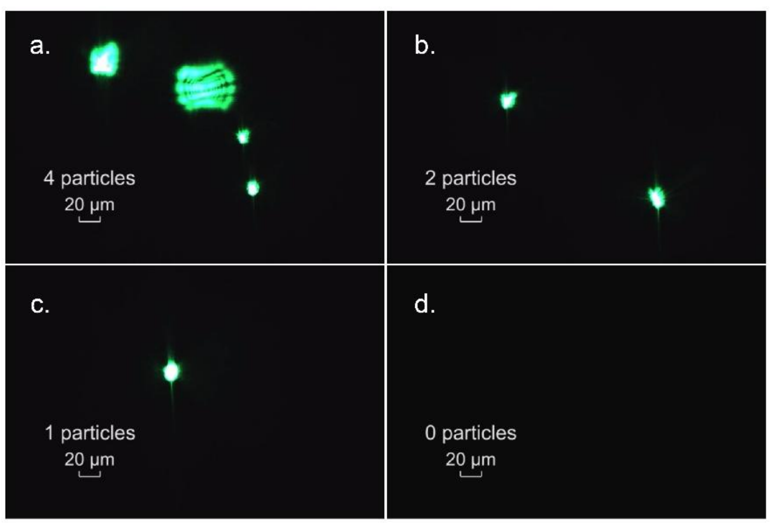

Fig. 2. Manipulation of the particles number in the optical trap. (a) Four, (b) two, (c) one, and (d) zero Bermuda grass smut spores were trapped inside the optical trap Fig. 2. A few examples for the trapping schemes and some images of the apparatus.

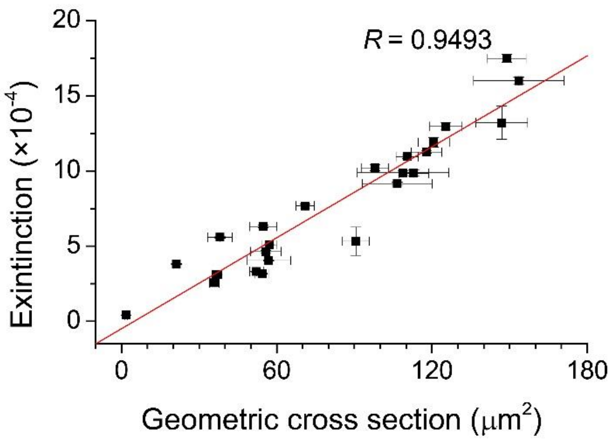

Fig. 3. The linear relation between the extinction at 308 nm and geometric cross section of 24 single MWCNT particles.

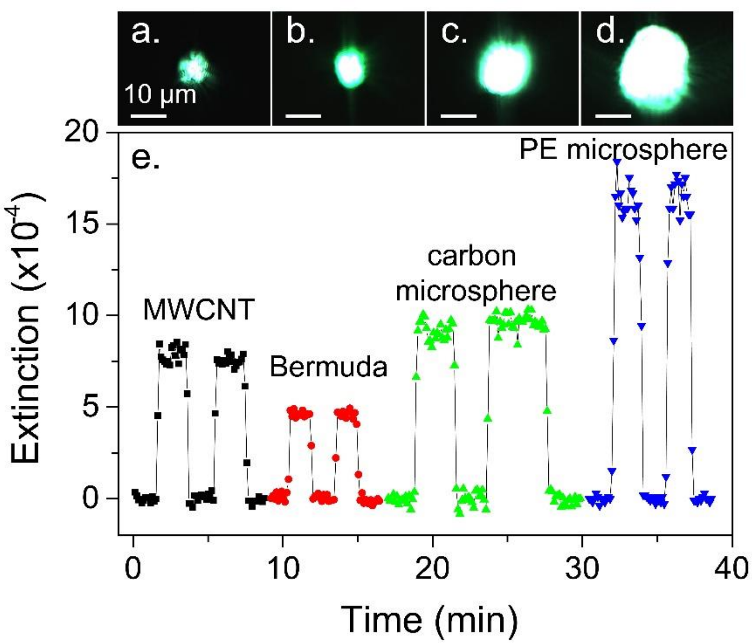

Fig. 4. Extinction of four different types of particles at 308 nm. Top row shows the images of single trapped (a) MWCNT, (b) Bermuda, (c) carbon microsphere, and (d) PE microsphere. And their corresponding extinction measurement results are shown in (e). The measurements are repeated at least twice for each type of particles.

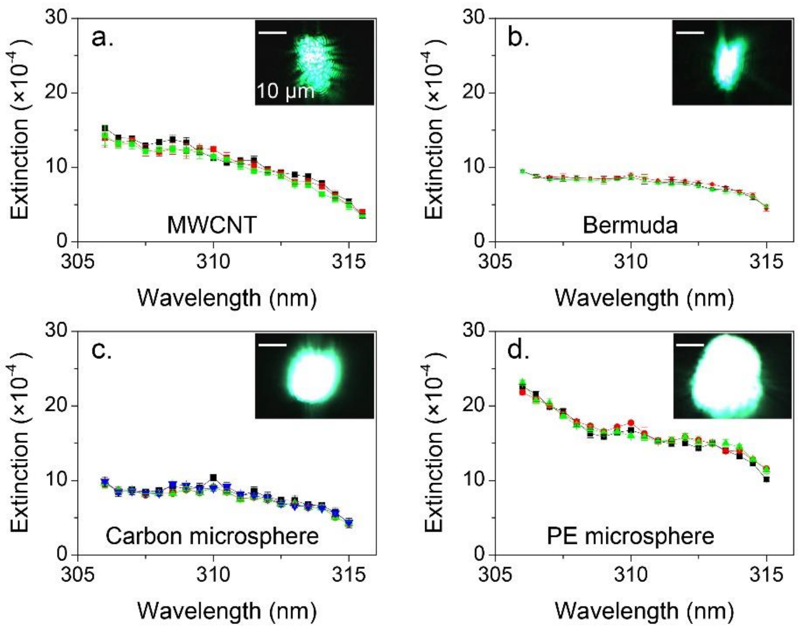

Fig. 5. Wavelength-dependent extinction datasets of a single (a) MWCNT particle, (b) Bermuda grass smut spore, (c) carbon microsphere, and (d) PE microsphere.

2. The temporal evolution process from fluorescence bleaching to clean Raman spectra of single solid particles optically trapped in air

We observe the entire temporal evolution process of fluorescence and Raman spectra of single solid particles optically trapped in air. The spectra initially contain strong fluorescence with weak Raman peaks, then the fluorescence was bleached within seconds, and finally only the clean Raman peaks remain. We construct an optical trap using two counter-propagating hollow beams, which is able to stably trap both absorbing and non-absorbing particles in air, for observing such temporal processes. This technique offers a new method to study dynamic changes in the fluorescence and Raman spectra from a single optically trapped particle in air.

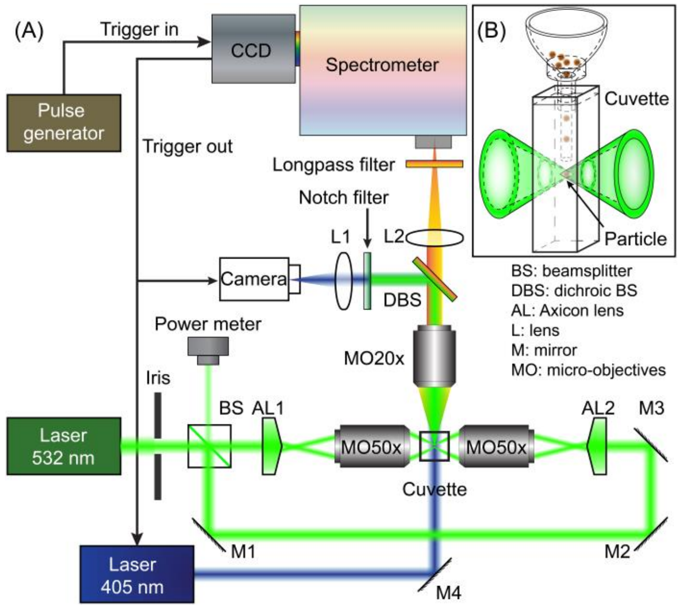

Fig. 6. The experimental setup of the Optical Trapping-Raman Spectroscopy (OT-RS) system. (A) The OT-RS system based on two counter-propagating hollow beams for trapping and characterizing both absorbing and transparent particles. (B) The detailed particle introduction setup and the hollow optical trap.

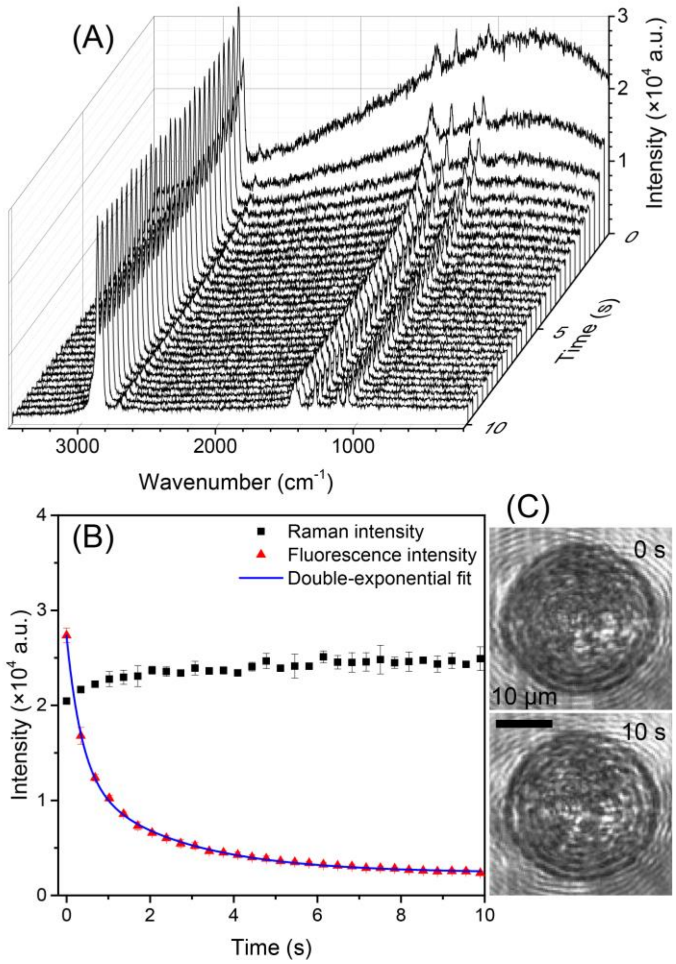

Fig. 7. The evolution process from fluorescence to Raman spectra of a single strongly absorbing rhodamine B doped PE microsphere in the optical trap. (A) The time-resolved spectra in the first 10 s. (B) The fluorescence intensity (red triangles) was fitted to a double-exponential decay (blue solid line). The intensity of the Raman peak at ~2890 cm^{-1} (black squares) remained relatively constant. (C) Images of the trapped particle taken at 0 s and 10 s.

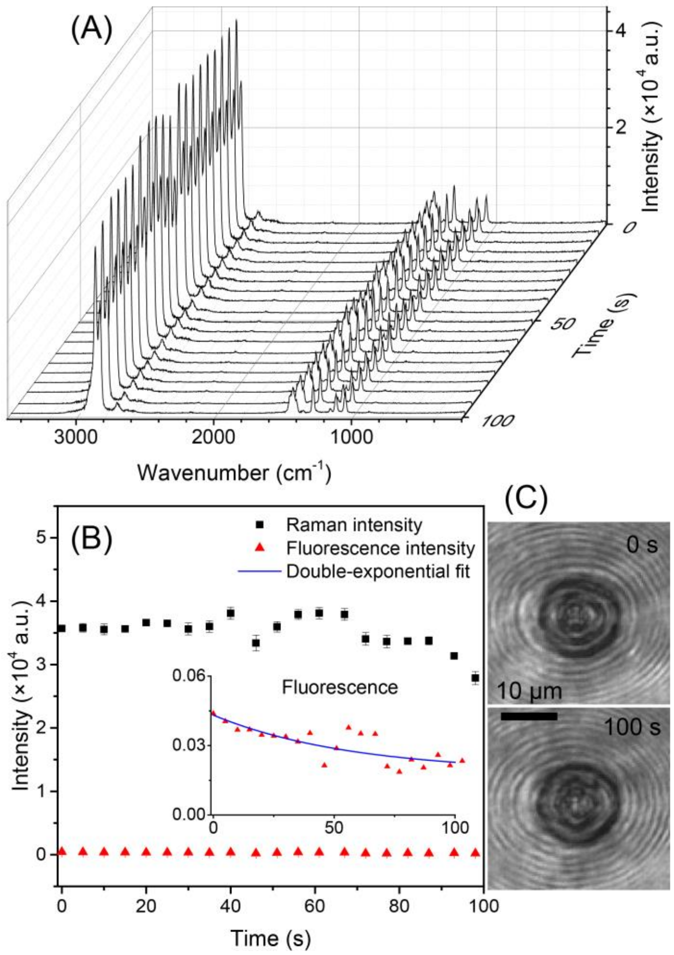

Fig. 8. The evolution process from fluorescence to Raman spectra of a single weakly absorbing fluorescent green PE microsphere in the optical trap. (A) The time-resolved spectra in the first 100 s. (B) The fluorescence intensity (red triangles) was fitted to a double-exponential decay (blue solid line). The Raman peak at ~2890 cm^{-1} (black squares) remained relatively constant. (C) The images of the trapped particle taken at 0 s and 100 s.

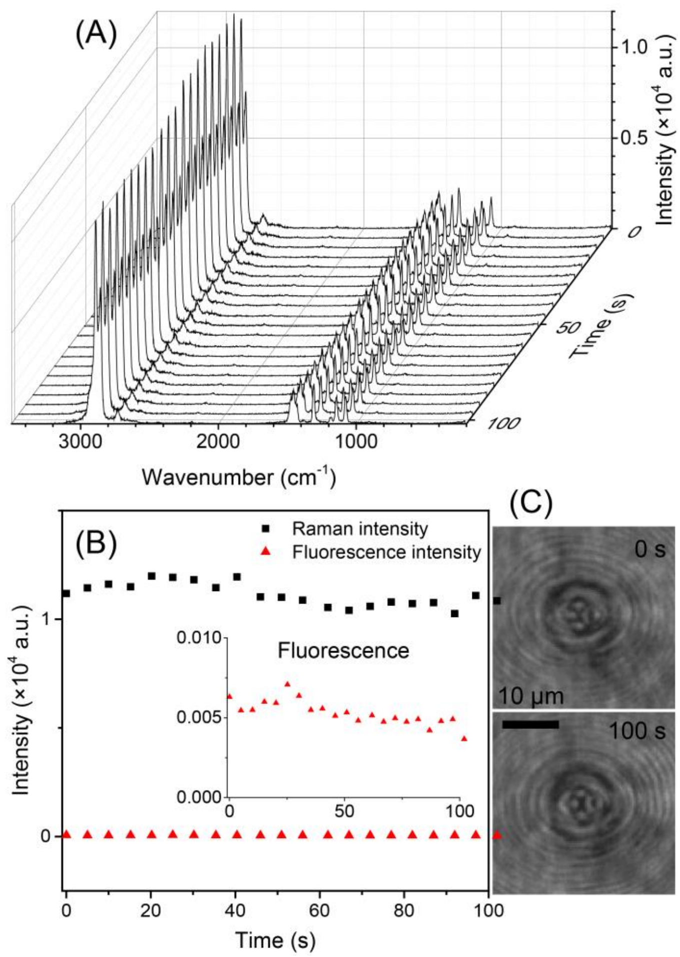

Fig. 9. The evolution process from fluorescence to Raman spectra of a single non-absorbing PE microsphere in the optical trap. (A) The time-resolved spectra in the first 100 s. (B) The fluorescence intensity (red triangles) and the Raman peak at ~2890 cm^{-1} (black squares) remained relatively constant. (C) The images of the trapped particle taken at 0 s and 100 s.

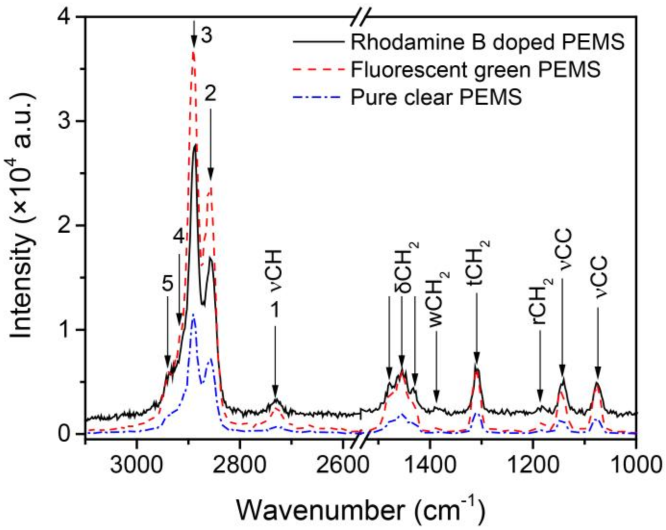

Fig. 10. Assignments of the Raman spectra from the three types of single PE microspheres (PEMS) trapped in air. (Notation: ν: stretch; r: rock; t: twist; w: wag; δ: deformation.)

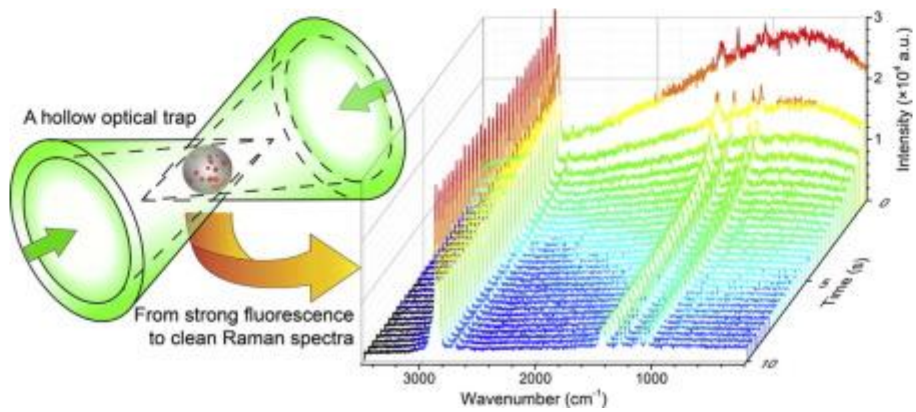

Fig. 11. From strong fluorescence to clean Raman of a bioaerosol particle optically trapped in the universal optical trap.

3. The novel universal optical trap (UOT)

Current optical traps are either for light absorbing particles only using primarily the photophoretic force or for nonabsorbing particles only using the radiation pressure force. In each of the two categories, there are various optical configurations used to trap single particles in air; however none of the optical traps are suitable for both absorbing and nonabsorbing particles, except for one recent work which demonstrates the capability of trapping both absorbing and nonabsorbing particles using a single setup. We have developed a universal optical trap (UOT) that can trap particles of arbitrary chemical and physical properties in the micron-sized range. The UOT uses two identical specially formed counter-propagating hollow beams to form a hollow optical trapping region. Single particles or multi-particles are trapped in the region for a long period of time (hours or longer) without directly contact of the light. Furthermore, we have demonstrated for the first time the integration of the UOT into different measuring techniques such as optical trapping-cavity ringdown spectroscopy (OT-CRDS) and optical trapping-Raman spectroscopy (OT-RS). We have successfully trapped absorbing and nonabsorbing particles in air using the UOT, followed by OT-CRDS and OT-RS measurements.

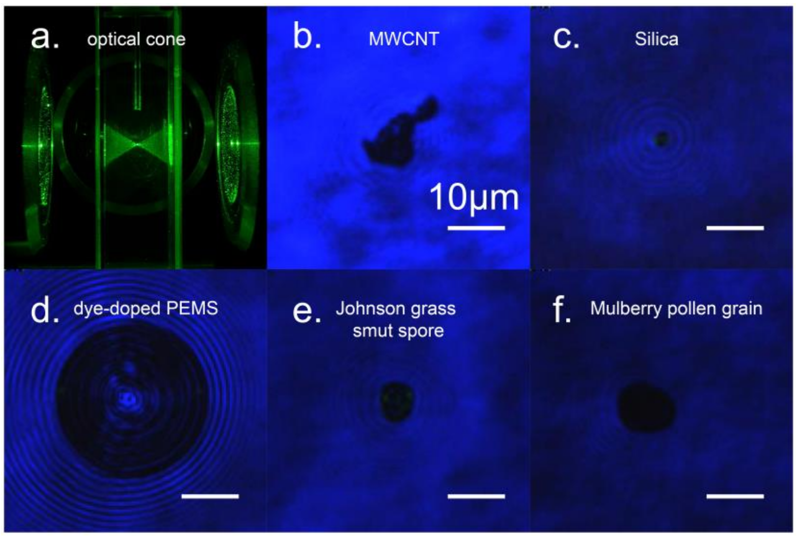

Fig. 12. (a) The novel universal optical trap for trapping both absorbing and nonabsorbing particles in air. (b-f) The images of the single optically trapped particles: MWCNTs, silica microsphere, rhodamine B doped polyethylene microsphere (RhB-PEMS), Johnson grass smut spore, and paper mulberry pollen grain.

4. Optical trapping-Raman spectroscopy (OT-RS) with embedded microscopy imaging for concurrent characterization and monitoring of physical and chemical properties of single particles

Highlights

- •A universal optical trap for trapping a wide variety of particles in air.
- •Integrated OT-RS system for concurrent measurements of physical and chemical properties.
- •Three test cases to demonstrate the analytical merits of the integrated OT-RS system.
- •A new method to study the physical and chemical evolutions of single particles in air.

The study of physical and chemical properties of a microscopic object, such as a single particle, is made possible using optical trapping (OT) technology combined with other measuring techniques. Here we show a universal optical trap combined with Raman spectroscopy (RS) and microscopy imaging for single-particle studies. The universal optical trap is constructed using two counter-propagating hollow beams and is able to stably levitate single particles of a wide range of properties, such as transparent or absorbing materials, organic (polymers, bioaerosols, etc.) or inorganic constituents (carbon, silica, glass, etc.), and spherical or irregularly shaped morphologies. Both physical and chemical properties and their temporal evolution of the trapped particle can be characterized simultaneously using the integrated OT-RS and imaging system. We created three sample cases to demonstrate the analytical merits of the system: (I) a single particle with no change, (II) partially degraded over the measuring period, and (III) one part from the fragmentized single particle. The particles' chemical compositions, crystalline states, etc. are inferred from their Raman spectra, while their physical properties (sizes, shapes, morphologies, etc.) are revealed by images. This integrated OT-RS system provides a new approach to concurrently characterize and monitor physical and chemical properties of single micrometer-sized objects optically trapped in air.

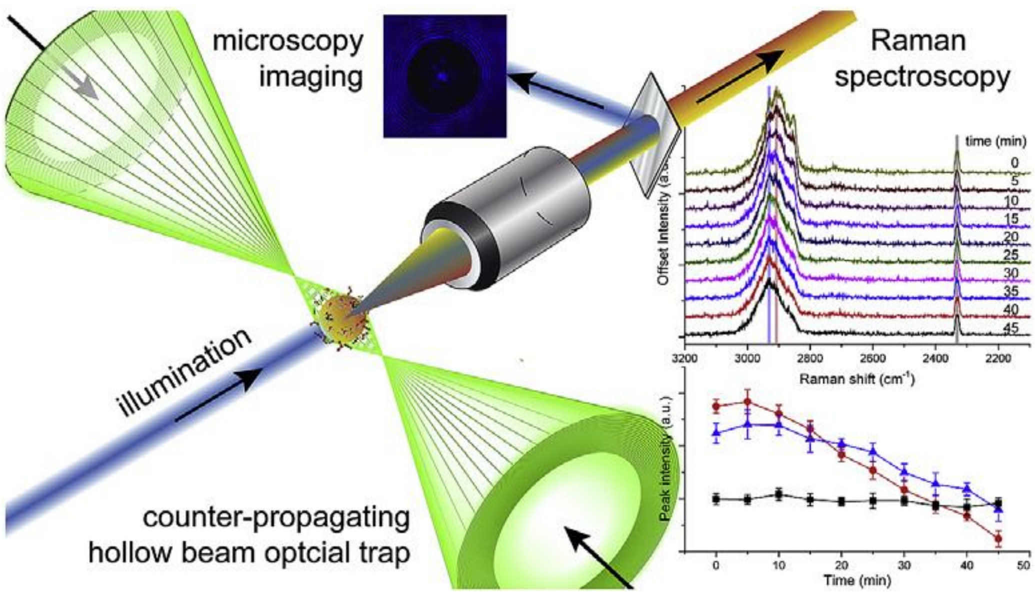

Fig. 13. The Optical trapping-Raman spectroscopy (OT-RS) with embedded microscopy imaging system.

5. An invited review article on
Optical trapping and manipulation of single particles in air: Principles, technical details, and applications

Single particle studies have gained increasing interest in recent years; researchers in the community welcome a timely update on the new optical trapping and manipulation technologies. We were invited to contribute a review article with this regard. The review highlights:

- Optical trapping technique for diverse single particles in air.
- Technical details of optical trapping for single particles in air.
- Recent applications of single optically trapped airborne particles.

The content list of the article:

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2. *Principle of optical trapping ············4*
   - *2.1. Radiation pressure force ············4*
   - *2.2. Photophoretic force ············5*
3. *Experimental configurations for optical trapping and manipulation in air ············7*
   - *3.1. Trapping schemes for transparent particles ············7*
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   - *4.3. Cavity ringdown spectroscopy ············30*

Trapping a single aerosol particle allows detailed investigation of its fundamental properties over extended time periods without external interferences. Optical trapping has developed into a powerful tool to perform such single-particle studies. However, trapping and manipulating a single particle in air, especially an irregularly shaped, absorbing particle, is much more challenging than that of a particle in a liquid solution. Even though the underlying mechanisms are not fully understood, recent experimental developments advanced the technique for trapping single particles in air, making it possible to manipulate and characterize a wide range of single particles. In this paper, we review recently demonstrated optical configurations for trapping and manipulating single airborne particles. Based on different trapping principles, we tentatively categorize them into radiation-pressure traps, photophoretic traps, and universal optical traps (UOTs). Radiation-pressure traps are based on the radiation pressure force resulting from photon momentum transfer; they include the early optical levitation configurations and the well-known optical tweezers. Photophoretic traps are based on the complex photophoretic forces that occur in absorbing particles; they are classified by the optical arrangements and include single-beam, dual-beam, and confocal-beam traps. UOTs can trap a variety of different types of particles, including transparent or absorbing, spherical or irregularly shaped, and liquid or solid particles. In order to evaluate each optical trapping scheme, four key aspects, i.e., simplicity, robustness, flexibility, and efficiency, of an optical trapping configuration are discussed. In addition to the stable optical trapping, optical manipulations from one dimension to three dimensions allow studying various single particles with great flexibility. With the single particle stably trapped and flexibly manipulated in air, other analytical techniques can be used to characterize these particles. Recent updates on optical methods for characterizing and monitoring single particles in air are discussed, such as light scattering, Raman spectroscopy, and cavity ringdown spectroscopy (CRDS).

6. Study of single airborne particle using laser-trapped submicron position-resolved temporal Raman spectroscopy

Highlights:

- •Evaporation of mixed droplet composed of diethyl phthalate and glycerol.
- •Submicron position-resolved and time-resolved Raman spectroscopy.
- •Distributions of molecules in airborne droplet.
- •Laser-trapped single airborne particle.

Study of various molecules located in different positions and of their temporal reactions within a micron sized particle in its natural phase is essential to deeper understanding the functions of these molecules. Here, we measured the temporal Raman spectra in different submicron positions of a laser-trapped droplet composed of diethyl phthalate and glycerol. Results demonstrated the micro-cavity enhanced effects, the morphology change, dynamics of phase-separation, and evaporation of the mixed droplet. This method offers a powerful tool to monitor various chemical reactions in different positions within a single cell, spore, or particle in life science and atmospheric science.

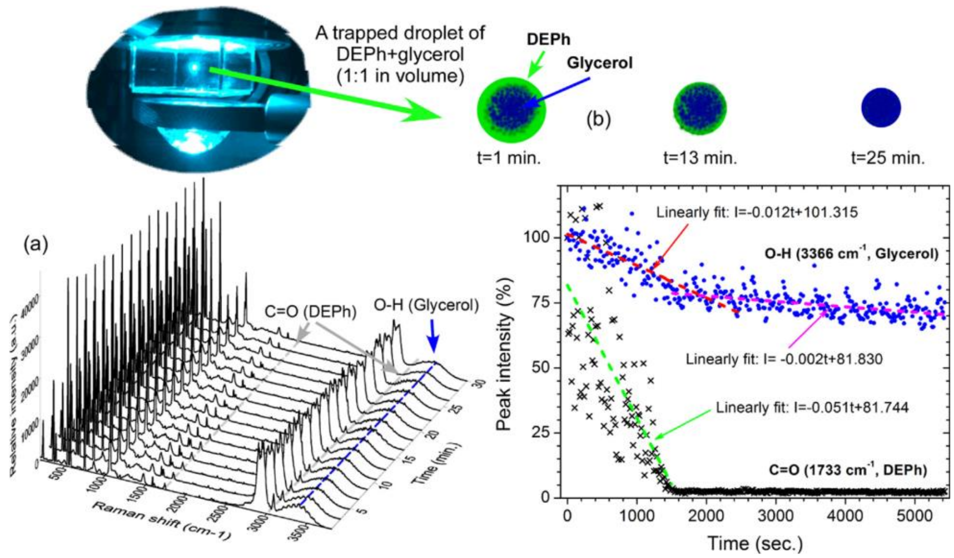

Fig. 14. (a). The time-resolved Raman spectra from a laser-trapped single-droplet composed of DEPh and glycerol in a 1:1 initial volume ratio, the broad band at 3366 cm^{-1}(marked with dashed blue line) is indicative of glycerol; and the small sharp peaks at 1733 cm^{-1} and 3080 cm^{-1} (marked with dashed grey line) are indicative of DEPh. (b) The time-dependent intensity decay and their linear fittings of the two indication peaks from DEPh and glycerol, as they were separating and evaporating. In the top-left shows an actual picture of a laser-trapped droplet, and the three cartoons in the top-right illustrate the mixed droplet at different times. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.).

7. Study of chemical reactions on a single trapped particle with chemical agitation under controlled environment

Highlights:

- Single particle chemical reactions
- Time-resolved single particle OT-RS
- Reactions of ozone with single pollen fragments

Single mulberry pollen grain fragments were trapped in a universal optical trap, in which laser trapping beam also serves as Raman excitation laser beam yet without touching the particle directly. Time-resolved OT-RS were recorded with and without introduction of reactant ozone to observed differences in the Raman spectra in terms of changes in spectral intensity, Raman band structures, and particle scattering over the observation period ranging from 0 to hrs. We used the time-resolved OT-RS of the single pollen fragment as a baseline. We conclude:

- We demonstrate the OT-RS system's capability of monitoring chemical reactions in a single pollen fragment in a controlled environment.

- Rapid and significant chemical reactions between single pollen fragments and ozone were observed directly from their time-resolved Raman spectra.
- At the beginning, the Raman spectra of the pollen fragments trapped in ozone are similar to those trapped in room air, but all the peaks in the ozone dropped dramatically within a few minutes and then stayed at a similar changing rate as the baseline in room air. The peaks decreased heterogeneously for pollen in ozone, leading to a significant band shape change, indicating different reaction rates for different chemical compositions.
- The OT-RS technique can be applied to further investigate and monitor the chemical reactions of different single bioaerosols in their aerosol state and with a broad range of atmospheric reagents.

Some of the results are highlighted below.

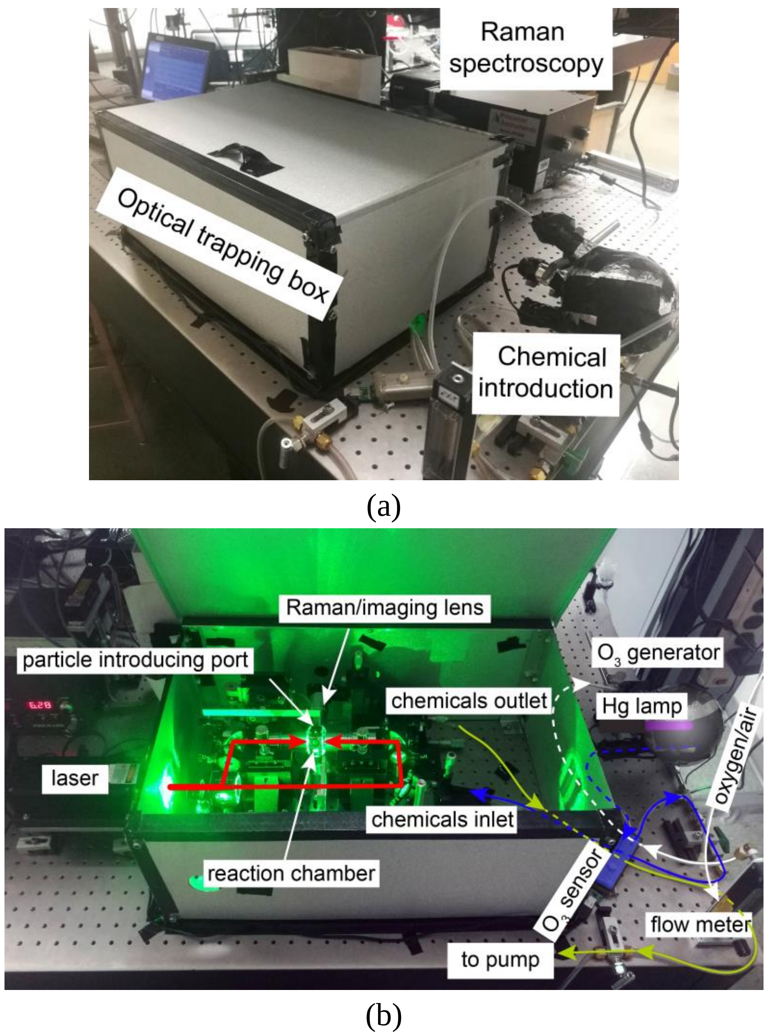

Fig. 15. The single particle OT-RS reactor system. (a) The integrated instrument system. (b) The detailed instrument components of the system.

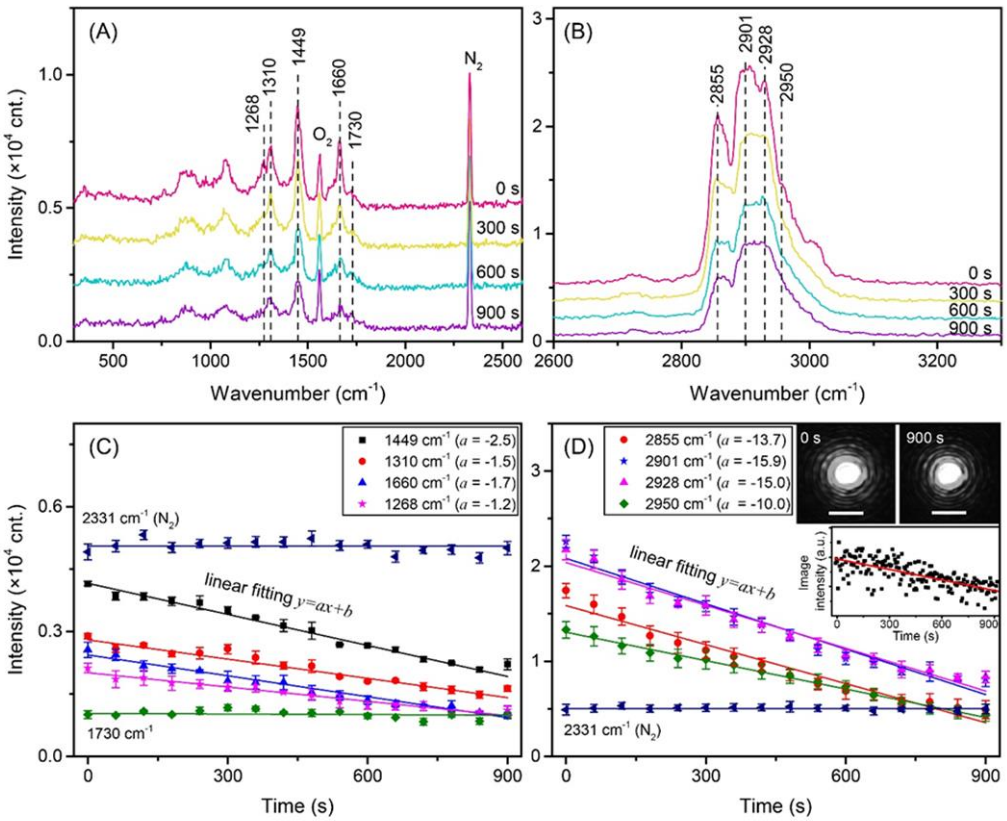

Fig. 16. Time-resolved Raman spectra were acquired from a single trapped pollen fragment in room air. A number of Raman peaks are marked in (A) the lower wavenumber region and (B) the ν(CH) band. Corresponding peak intensity analyses are shown in (C) and (D). The intensities of those marked peaks slowly decreased over time. The intensity change rate was indicated by the slope of linear fittings. The inset figure of (D) shows the decreasing intensity of the scattering images.

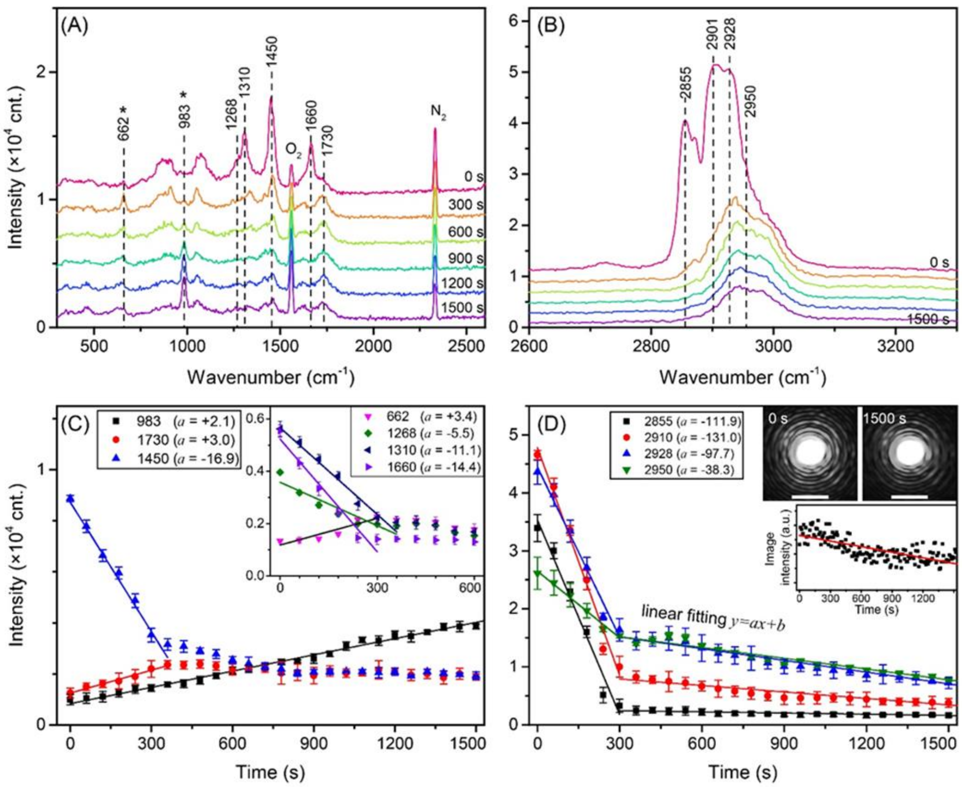

Fig. 17. Raman spectra of a single pollen fragment reacting with ozone. A number of peaks indicating chemical reactions are marked in (A) the lower wavenumbers and (B) the ν(CH) band. Compared with a single pollen fragment in room air, new peaks at 662 and 983 cm-1 are marked by stars. Intensities of each marked peak were linearly fitted in (C) and (D). The insets in (D) show slightly decreased intensities of scattering images.

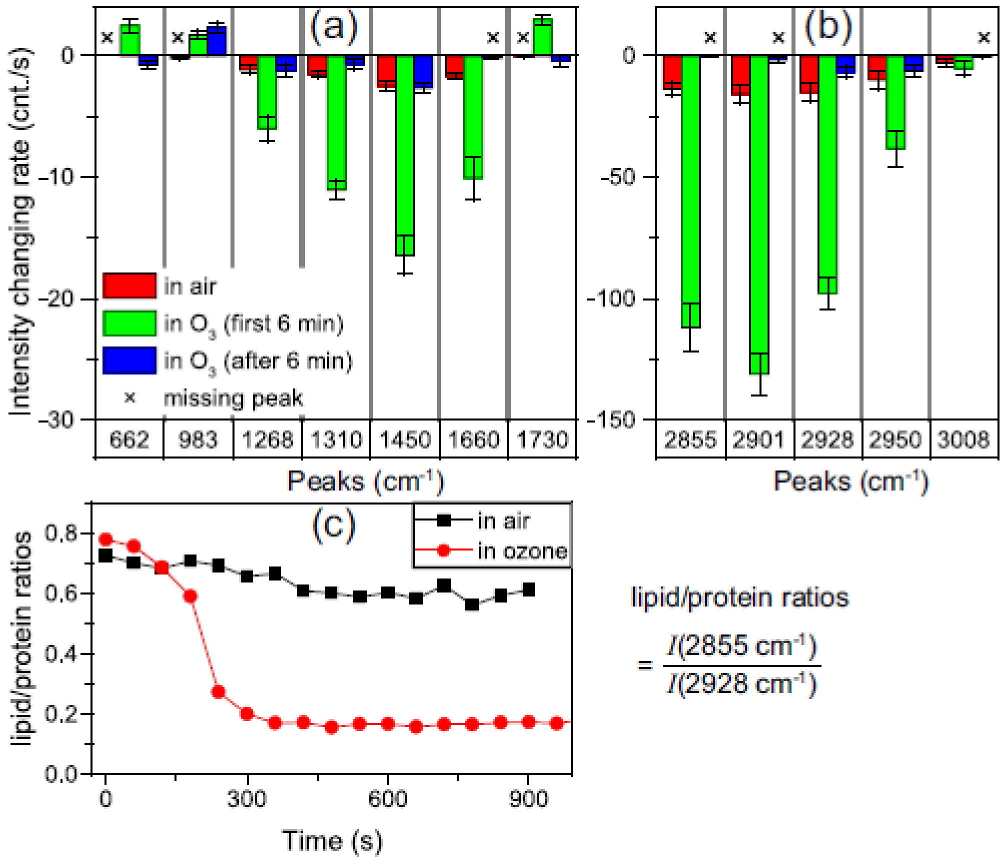

Fig. 18. Quantitative analysis of reaction rate. Intensity of changing rate of selected Raman peaks at (a) 500-1800 cm^{-1} and (b) the CH band. Three categories of changing rates were grouped: pollen in room air (red bars), the first 6 min in ozone (green bars), and after 6 min in ozone (blue bars). (c) Peak intensity ratios of a lipid peak (2855 cm^{-1}) and a protein peak (2928 cm^{-1}) under different environments.

<u>What opportunities for training and professional development has the project provided?</u>

One PhD student has received training in this project. He graduated in May 2019. Now he is working as a senior research engineer in Wyatt Technology (California). One undergraduate student has received training in research experience in the fall semester of 2017 and the spring semester 2018. This student graduated with a B.S. degree in physics. Now he is working as a research associate in Naval Oceanographic Office. Another undergraduate student is trained in this project since the summer of 2019. Five other graduate students, who are working on other projects in the PI's groups, have benefited from the research discussion and exposure to the research work in the lab.

Training and Development:

Graduate student:

Mr. Zhiyong Gong is the PhD student who graduated from his PhD research in this project. He has committed 100% of his research time to this project. His research efforts are focused on the integration of the newly developed universal optical trap (UOT) with other measuring techniques such as imaging, scattering, cavity ringdown spectroscopy and Raman spectroscopy. He has been conducting measurements of trapped particles with introduction of moisture and ozone. He has been trained in particle trapping, measurements, writing a manuscript, etc. He plays an important role in the experimental work in this project. He has become an expert in optical trapping and spectroscopic measurements of single particles.

Undergraduate student:

Mr. Jeff Headley was a senior majoring in physics during the project report period. He is interested in gaining research experience in lasers, spectroscopy, optical trapping, sample handling, etc. He has been trained with basic knowledge of lasers and CRDS. He graduated with a B.S. degree in physics and he now joins the Naval Oceanographic Office as a research scientist.

Mr. Cameron Gaito: A senior physical major**.** He joined the PI's research group obtain research experience in laser and optical trapping, and spectroscopy. This research experience will be benefit him for his planned graduate study.

<u>How have the results been disseminated to communities of interest?</u>

The project outcomes and research results are effectively and timely disseminated through peer-reviewed journal publications, conference presentations, and invited seminars, giving lab tours to visitors on and outside campus, project research site visit, project reporting, and the information posted in the PI's Research Group web site. Part of the activities is listed below:

- Via **15** peer-reviewed journal publications including two featured in the front cover of prestigious journals.
- Via **20** presentations in national and international conferences.
- Via the PI's research group web site, http://wang.physics.msstate.edu/index.php/en/

<u>What do you plan to do during the next reporting period to accomplish the goals?</u>

N/A

<u>Products</u>

<u>A. Journal Publications (15)</u>

1. Zhiyong Gong, Yong-Le Pan, Gorden Videen & Chuji Wang, Chemical reactions of single optically trapped bioaerosols in a controlled environment, Aerosol Science and Technology, 53, 853 (**2019**).

2. Zhiyong Gong, Yong-Le Pan, Gorden Videen & Chuji Wang, Online Characterization of Single Airborne Carbon Nanotube Particles Using Optical Trapping Raman Spectroscopy, Applied Spectroscopy, 73, 910 (**2019**).

3. Yong-Le Pan, Aimable Kalume, Isaac C. D. Lenton, Timo A. Nieminen, Alex B. Stilgoe, Halina Rubinsztein-Dunlop, Leonid A. Beresnev, Chuji Wang, and Joshua L. Santarpia, Optical-trapping of particles in air using parabolic reflectors and a hollow laser beam, Optics Express, 27, 33061 (**2019**)

4. Zhiyong Gong, Yong-Le Pan, GordenVideen, and Chuji Wang, Optical trapping-Raman spectroscopy (OT-RS) with embedded microscopy imaging for concurrent

characterization and monitoring of physical and chemical properties of single particles, Analytica Chimica Acta, 1020, 86 (**2018**).

5. Aimable Kalume, Chuji Wang, Joshua Santarpia, and Yong-Le Pan, Liquid–liquid phase separation and evaporation of a laser-trapped organic–organic airborne droplet using temporal spatial-resolved Raman spectroscopy, Physical Chemistry Chemical Physics, 20, 19151 (**2018**).

6. Zhiyong Gong, Yong-Le Pan, GordenVideen, and Chuji Wang, Optical trapping and manipulation of single particles in air: Principles, technical details, and applications, Journal of Quantitative Spectroscopy and Radiative Transfer, 214, 94 (**2018**).

7. Aimable Kalume, Chuji Wang, Joshua L. Santarpia, and Yong-Le Pan, Study of single airborne particle using laser-trapped submicron position-resolved temporal Raman spectroscopy, Chemical Physics Letters, 706, 255 (**2018**).

8. Yong-Le Pan, Aimable Kalume,Chuji Wang, Joshua L. Santarpia, Opto-aerodynamic focusing of aerosol particles, Aerosol Science and Technology, 52, 13 (**2018**).

9. Zhiyong Gong, Yong-Le Pan, GordenVideen, and Chuji Wang, The temporal evolution process from fluorescence bleaching to clean Raman spectra of single solid particles optically trapped in air, Chemical Physics Letters, 689, 100 (**2017**).

10. Aimable Kalume, Eric Zhu, Chuji Wang, Joshua Santarpia, and Yong-Le Pan, Position-resolved Raman spectra from a laser-trapped single airborne chemical droplet, Optics Letters, 42, 5113 (**2017**).

11. Zhiyong Gong, Yong-Le Pan, and Chuji Wang, Characterization of single airborne particle extinction using the tunable optical trap-cavity ringdown spectroscopy (OT-CRDS) in the UV, Optics Express 25, 6732 (**2017**).

12. Yong-Le Pan, Chuji Wang, Leonid A. Beresnev, Alex J. Yuffa, Gorden Videen, David Ligon, and Joshua L. Santarpia, Measurement of back-scattering patterns from single laser trapped aerosol particles in air, Applied Optics, 56, pp. B1-B4 **(2017)**

13. Richard Fu, Chuji Wang, Olga Muñoz, Gorden Videen, Joshua L. Santarpia, and Yong-Le Pan, Elastic back-scattering patterns via particle surface roughness and orientation from single trapped airborne aerosol particles, Journal of Quantitative Spectroscopy and Radiative Transfer 187, 224 (**2017**).

14. Zhiyong Gong, Yong-Le Pan, and Chuji Wang, Optical configurations for photophoretic trap of single particles in air, Review of Scientific Instruments, 87, 103104 (**2016**).

15. Chuji Wang, Zhiyong Gong, Yong-Le Pan, and Gorden Videen, Laser pushing or pulling of absorbing airborne particles, Applied Physics Letters, 109, 011905 (**2016**).

B. Books or Other One-time Publications

None

C. Conference Proceedings, Presentations, and Invited Talks (20)

1. Chuji Wang, Zhiyong Gong, Yong-Le Pan, Gorden Videen, Chemical reactions on single particles under controlled environment, AAAR, Portland OR, Oct. 13-18, 2019.
2. Aimable Kalume, Chuji Wang, Joshua Santarpia, Yong-Le Pan, Anatomy of single airborne aerosol particle using laser-trapped submicron position-resolved temporal Raman spectra, 10th International Aerosol Conference, St. Louis, MI, Sept. 2-7, 2018.
3. Aimable Kalume, Chuji Wang, Joshua Santarpia, Patricio Piedra, Yong-Le Pan, Dynamics of liquid-liquid phase-separation using spatial-resolved Raman spectroscopy of a laser-trapped mixed organic-organic aerosol droplet. 10th International Aerosol Conference, St. Louis, MI, Sept. 2-7, 2018.
4. Zhiyong Gong, Yong-Le Pan, Gorden Videen, Chuji Wang, Optical trapping-Raman spectroscopy (OT-RS) for concurrent characterization and monitoring of physical and chemical properties of single airborne particles, 10th International Aerosol Conference, St. Louis, MI, Sept. 2-7, 2018.
5. Chuji Wang, Zhiyong Gong, Gorden Videen, Yong-Le Pan, Single airborne particle studies using optical trapping and manipulations: What we have and what we have not, 10th International Aerosol Conference, St. Louis, MI, Sept. 2-7, 2018.
6. Aimable Kalume, Zhiyong Gong, Chuji Wang, Joshua Santarpia, and Yong-Le Pan, Detection and characterization of chemical and biological aerosol using laser trapping single particle Raman spectroscopy, 26th International Conference on Modelling, Monitoring and Management of Air Pollution, Naples, Italy, 19-21, Jun. 2018.
7. Zhiyong gong, Yong-Le Pan, Gorden Videen, and Chuji Wang, Optical trapping-Raman spectroscopic (OT-RS) study of single biological particles in air, Seventeenth Conference on Electromagnetic & Light Scattering Texas A&M University, College Station, TX, USA 4–9 Mar. 2018.
8. Aimable Kalume, Chuji Wang, Joshua Santarpia, and Yong-Le Pan, Submicron position-resolved Raman spectra for characterizing laser-trapped single airborne particles, Microscopy Histopathology and Analytics 2018, Hollywood, Florida United States, 3–6 April 2018, ISBN: 978-1-943580-41-5.
9. Chuji Wang, Zhiyong Gong, Yong-Le Pan, and Gorden Videen, Single airborne aerosol particle raman and cavity ringdown spectroscopy, AAAR, Raleigh, NC, Oct. 16-20, 2017.

10. Yong-Le Pan, Aimable Kalume, Chuji Wang, Joshua L. Santarpia, Opto-aerodynamic manipulating and focusing of aerosol particles, AAAR, Raleigh, NC, Oct. 16-20, 2017.
11. Chuji Wang, Investigation of chemical agitations on single chemical and biological aerosol particles in air using optical trapping-Raman spectroscopy, US Army Research Office Annual Project Review Meeting, Raleigh, NC, Aug. 7-11, 2017.
12. Chuji Wang, Zhiyong Gong, Yong-Le Pan, and Gorden Videen, Single Aerosol Particle Studies Using Optical Trapping Raman And Cavity Ringdown Spectroscopy, AGU, New Orleans, 11-15, Dec. 2017.
13. Zhiyong Gong, Chuji Wang, Yong-Le Pan, and Gorden Videen, Characterizing physical properties and heterogeneous chemistry of single particles in air using optical trapping-Raman spectroscopy, AGU, New Orleans, 11-15, Dec, 2017.
14. Chuji Wang, Optical trapping- cavity ringdown spectroscopy and optical tapping Raman spectroscopy, Jackson State University, Invited Seminar, Feb. 2017.
15. Chuji Wang, Zhiyong Gong, Yong-Le pan, and Gorden Videen, Single airborne aerosol particle Raman and cavity ringdown spectroscopy, Isalsars 2017, Hefei, China, June 19-23, 2017.
16. Yong-Le Pan, Richard Fu, Gorden Videen, Chuji Wang, Joshua L. Santarpia, Simultaneous measurement of elastic back-scattering patterns and images from laser-trapped single airborne particles, Isalsars 2017, Hefei, China, June 19-23, 2017.
17. Zhiyong Gong, Chuji Wang and Yong-Le pan, Optical trapping and manipulation configurations for measuring light extinctions of single particles, The 16th Electromagnetic and Light Scattering Conference (ELS-XVI), College park, Maryland, Mar. 19-25, 2017.
18. Zhiyong Gong, Yong-Le Pan, and Chuji Wang, Optical configurations for photophoretic trap of single particles in air, American Aerosol Association Research, 35th Annual Conference, Portland, Oregon, Oct. 17-22, 2016.
19. Chuji Wang, Zhiyong Gong, Yong-Le Pan, Gorden Videen, Laser pushing or pulling of absorbing airborne particles, American Aerosol Association Research, 35th Annual Conference, Portland, Oregon, Oct. 17-22, 2016.
20. Yong-Le Pan, Brandon Redding, Chuji Wang, Steven C. Hill, Joshua L. Santarpia, Optical trap for both transparent and absorbing particles in air using a single shaped laser beam for measuring Raman spectra, American Aerosol Association Research, 35th Annual Conference, Portland, Oregon, Oct. 17-22, 2016.

Project Participants

Senior Personnel

Name: Chuji Wang

Worked for more than 160 Hours/year: Yes

Contribution to Project:

Conducted experiments, wrote papers, and advised students

Project collaborators

Name: Dr. Yong-Le Pan and Dr. Gorden Videen

Contribution to Project:

Research results discussion, data sharing, instrument/equipment sharing, co-authoring articles and conference presentations

Graduate Student

Name: Zhiyong Gong (PhD candidate)

Worked for more than 160 Hours/year: Yes

Contribution to Project:

Conducted experiments; collected experimental data; analyzed experimental data.

Undergraduate Student

Name: Jeff Headley (Physics major, graduated in 2018), Cameron Gaito (Physics major, senior in 2019)

Worked for more than 160 Hours/year: Yes

Contribution to Project:

Research Experience:

Gained research experiences in lasers and cavity ringdown technologies; understood basics of optical trapping.

Impact

A. Impact within Discipline

One of the most significant changes in chemistry history is the introduction of the molecular beam (or called supersonic beam), which allows us to study chemical reactions and dynamics at the molecular level for the first time. This change was credited to Y T Lee who won the Nobel

Prize in chemistry in 1986. Now we may ask can we study aerosol particle reactions and dynamics at the fundamental level (the single-particle level)? The answer is YES. This should be credited to the novel technique—optical trapping, which was first introduced by A. Ashikin who received the Nobel Prize in Physics in 2018 for his contribution to the optical tweezers. From the original concept of optical tweezers to our most recently defined universal optical trap (UOT), there have been a lot of developments in optical trapping and its applications. One of them is the research activities conducted in this project.

An optically levitated particle in air provides a unique vehicle to study the surface chemistry/heterogeneous reactions, in which the sample is free of electrical charge and has no interferences from other sources (bulk samples and substrates), yet it has high flexibility to interact with selected chemical reagents. Our UOT integrated with the single particle OT-RS is a platform technology that is applicable to study the surface chemistry processes under a controlled chemical environment (e.g. in air, with selected reactive reagents). This technology provides a unique channel to better understand heterogeneous reactions and dynamics under various environmental conditions. As seen in the demonstrated results (see section *"What was accomplished under these goals?"* in this report)), we expect the research activities will generate significant impact in the field of environmental and atmospheric chemistry and processing, particularly, in heterogeneous surface reactions between a solid (or liquid) surface and selected reactive reagents. They key point lies in the unique capability of observing the surface reactions with no interferences from other sources such as those from bulk samples and/or substrates as in the conventional approaches.

The research activities have generated several technological breakthroughs:

- The universal optical trap (UOT) that taps particles of arbitrary physical and chemical properties in air or in a solution.
- UOT for the study of trapped single particles with externally introduced reactive agents.
- The integrated OT-CRDS for single particle extinction measurement.
- The integrated OT-RS with a fast imaging system for concurrent characterization of physical and chemical properties of single particles in air.

- The high-resolution time- and position-resolved OT-RS within a single particle.

B. Impact on Human Resource Development

One graduate student and two undergraduate students have received training in this project. Five other graduate students, who are working on other projects in the PI's groups, have benefited from the research discussion and exposure to the research work in the lab. The graduated student, Dr. Gong joined Wyatt Technology, a leading analytical instrumentation company in the US.
The trained undergraduate student has been employed in Naval Oceanographic Office as a research scientist.

C. Impact in a larger community

Single particle studies are related to atmospheric sciences, environmental processes, material fabrication, etc. Single particles, especially single bioaerosol particles, are strongly related to missions in the US defense and national security defense. Optical trapping, transport, manipulation of single particles in air provides an unprecedented channel to study particle (solid and liquid) surface chemical reactions without interferences from substrates and bulk samples. Project results are widely disseminated in national and international research communities via publications and presentations in prestigious conferences. The developed technologies, such as the newly demonstrated universal optical trap (UOT), OT-CRDS, and OT-RS, single particle OT-RS reactor, are transformative and universally applicable to studies of single particles in various forms, particle formation and degradation, heterogeneous and multiphase reactions, single biomolecule sensors and sensing, etc. Research activities conducted in this project are in strong collaboration with several scientists in ARL (Drs and ARL Fellows Yong-Le Pan and Gorden Videen).

Changes and Problems

Special reporting requirements: None

Change in Objectives or Scope: None

Reprints of the 15 articles listed below are uploaded in the "Product" category of the project report website.

1. Zhiyong Gong, Yong-Le Pan, Gorden Videen & Chuji Wang, Chemical reactions of single optically trapped bioaerosols in a controlled environment, Aerosol Science and Technology, 53, 853 (**2019**).

2. Zhiyong Gong, Yong-Le Pan, Gorden Videen & Chuji Wang, Online Characterization of Single Airborne Carbon Nanotube Particles Using Optical Trapping Raman Spectroscopy, Applied Spectroscopy, 73, 910 (**2019**).

3. Yong-Le Pan, Aimable Kalume, Isaac C. D. Lenton, Timo A. Nieminen, Alex B. Stilgoe, Halina Rubinsztein-Dunlop, Leonid A. Beresnev, Chuji Wang, and Joshua L. Santarpia, Optical-trapping of particles in air using parabolic reflectors and a hollow laser beam, Optics Express, 27, 33061 (**2019**)

4. Zhiyong Gong, Yong-Le Pan, GordenVideen, and Chuji Wang, Optical trapping-Raman spectroscopy (OT-RS) with embedded microscopy imaging for concurrent characterization and monitoring of physical and chemical properties of single particles, Analytica Chimica Acta, 1020, 86 (**2018**).

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13. Richard Fu, Chuji Wang, Olga Muñoz, Gorden Videen, Joshua L. Santarpia, and Yong-Le Pan, Elastic back-scattering patterns via particle surface roughness and orientation from single trapped airborne aerosol particles, Journal of Quantitative Spectroscopy and Radiative Transfer 187, 224 (**2017**).

14. Zhiyong Gong, Yong-Le Pan, and Chuji Wang, Optical configurations for photophoretic trap of single particles in air, Review of Scientific Instruments, 87, 103104 (**2016**).

15. Chuji Wang, Zhiyong Gong, Yong-Le Pan, and Gorden Videen, Laser pushing or pulling of absorbing airborne particles, Applied Physics Letters, 109, 011905 (**2016**).

The End