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Chemical Analysis of Aerosol Particle Surfaces

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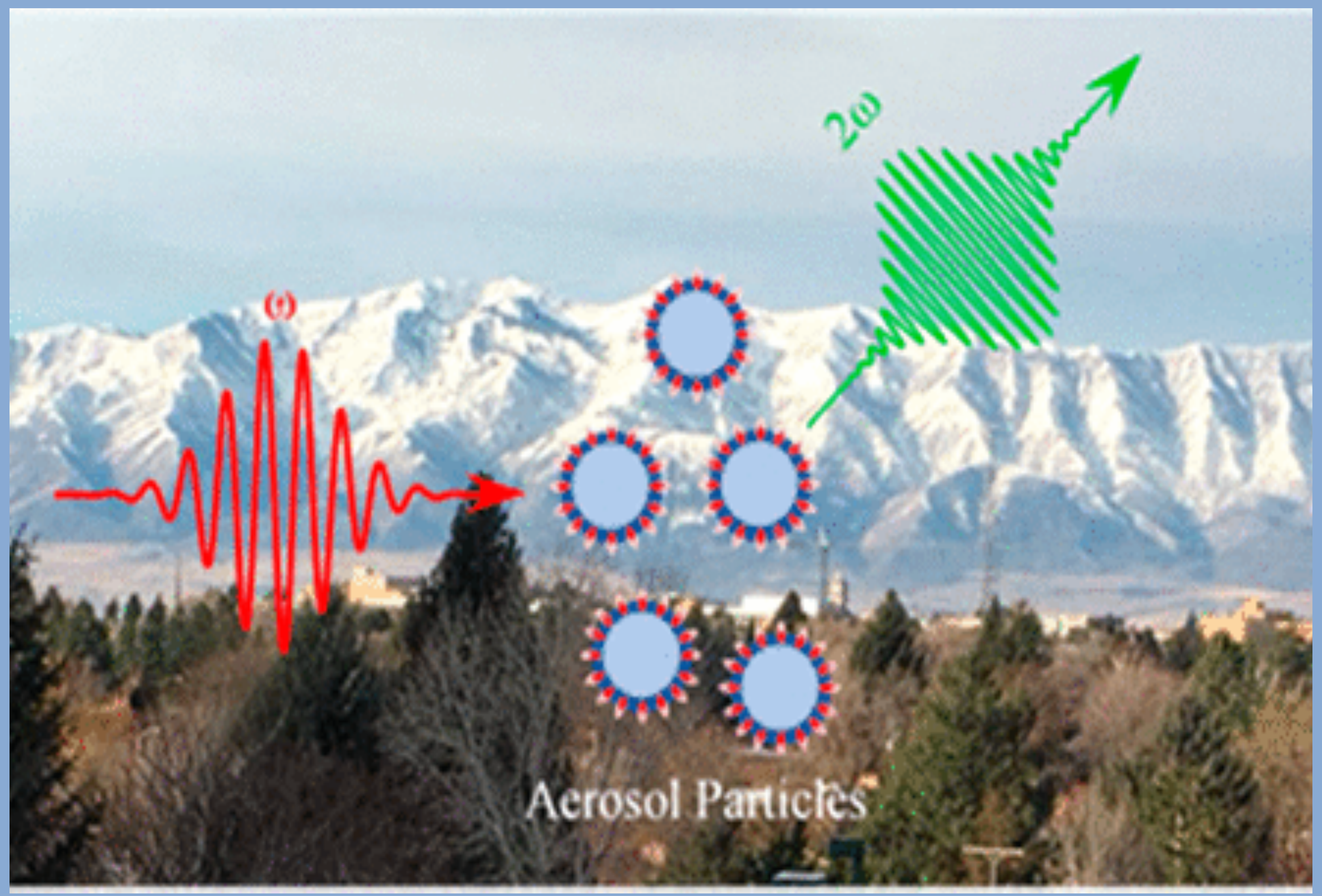
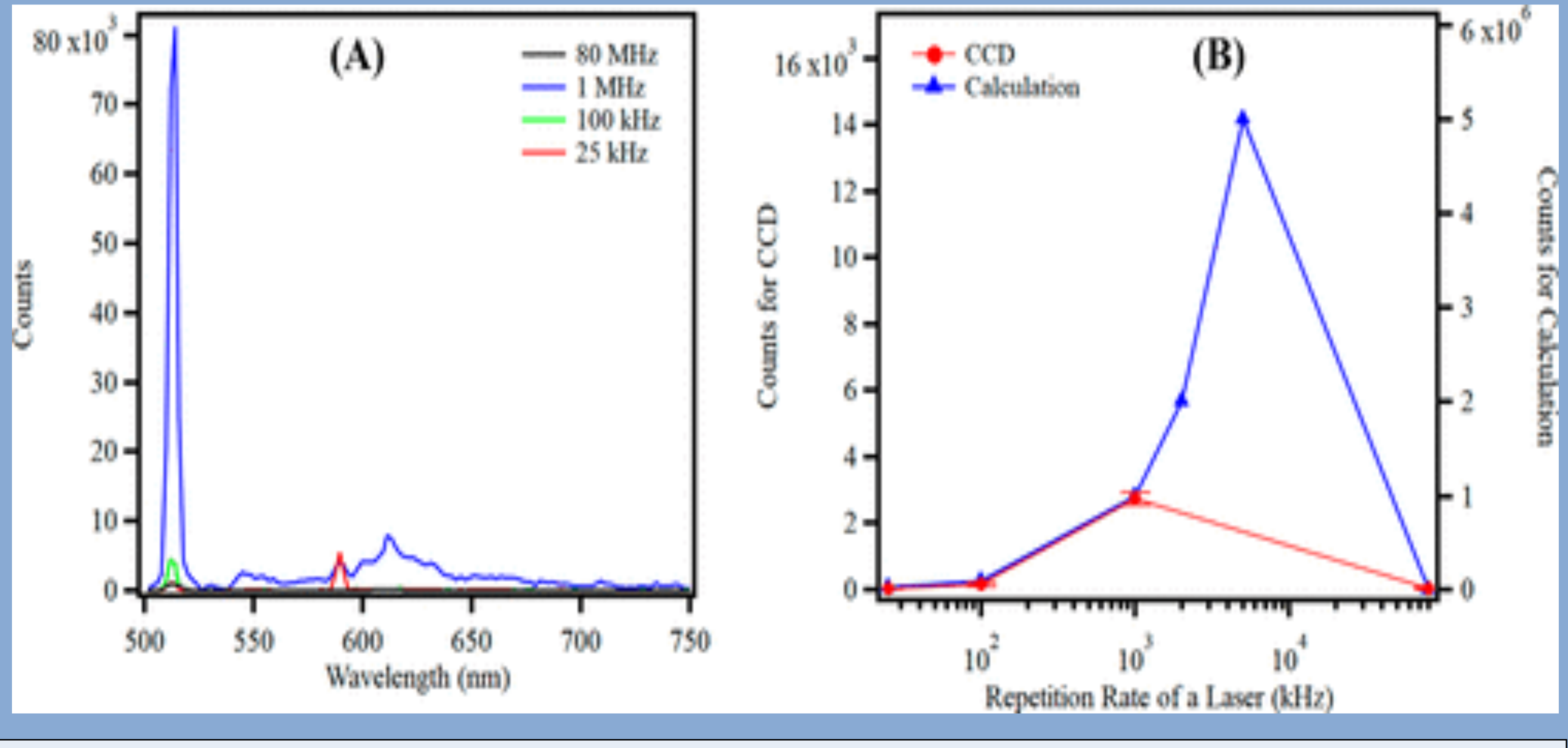
Abstract

Aerosol particles are thought to impact climate properties. The ability to probe aerosols to understand their chemical properties has been difficult due to a lack of appropriate analytical techniques. Here we present interface-specific SHS techniques to allow for in situ analysis of aerosol particles. Transportation of aerosols from their collection site to the laboratory disturbs their chemical and physical properties thus making it difficult to take accurate measurements of aerosol particles. We show that using a CCD detector is a more effective option for producing more sensitive results and can greatly reduce sampling time and could possibly detect measurements from a single aerosol particle. Using these techniques we can gain a greater understanding of aerosol composition and of how aerosol properties affect atmospheric conditions. SHS can quantitatively determine the density of organic molecules at the aerosol particle's surface. In combination with the in-situ techniques developed we can gain better understanding of aerosol properties.



Experimental Results or Data

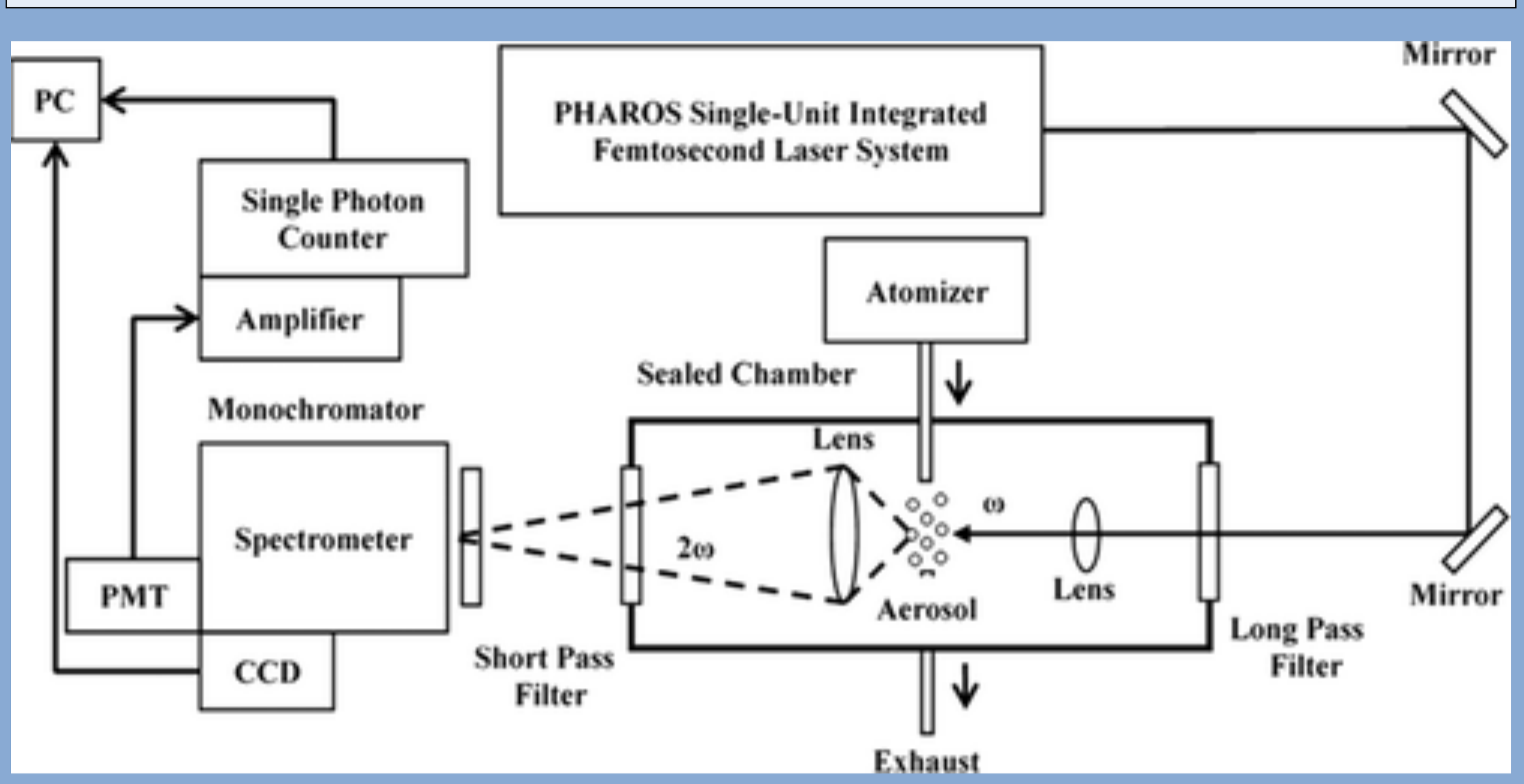


(A) Two-photon emission spectra of aerosol particles generated from 1.0 M NaCl solution mixed with 20 μM CV under four different laser repetition rates of 25 kHz, 100 kHz, 1 MHz, and 80 MHz, respectively. The pulse energies for the 25 kHz, 100 kHz, and 1 MHz lasers were kept at 1 μJ. The pulse energy for the 80 MHz laser was kept at 10 nJ. The spectra were taken with an integration time of 30 s. The interfacial responses of the aerosol particles show the largest SHS signal under a laser of 1 MHz. (B) Comparison of the experimental SHS signals from (A) with the calculated values. All of the calculated values were normalized with respect to the experimental value at the 1 MHz.

The experimental setup includes three units, including a generation unit for SHS, a generation and handling unit for aerosol particles, and a collection and detection unit for the SHS signal. A detailed description of the experimental setup is seen in the Supporting Information. Briefly, A 10 W single-unit integrated femtosecond laser system (PHAROS, Light Conversion) with a seed oscillator was used in our experiments. The polarization of the incident light was set to be perpendicular to the optical table. No analyzer was placed in the measurements. Aerosol particles were generated by compressing nitrogen gas at a pressure of 40 psi with a constant output atomizer (TSI 3076). A spectrometer (Acton 300i, Princeton Instruments) equipped with a charge coupled device (LN/CCD-1340/00, Princeton Instruments) or a photomultiplier tube (PMT H11901–20, Hamamatsu) constituted the SHS detection system.

Discussion of Results or Data

SHS signal for single particle is readily measured in one second with our laser and detection system. An experiment could be designed to monitor kinetic behaviors of interfaces at a single aerosol particle. Both a CCD detector and a single-photon counter have been proven to detect the interfacial second harmonic scattering signal. The CCD detector is a more effective option for the detection of second harmonic scattering and could greatly reduce sampling time of the interfacial responses. Combination of the optimal laser system with the CCD detector has greatly improved the detection sensitivity of interfacial molecules by more than 2 orders of magnitude and could potentially detect interfacial SHS from a single aerosol particle. We believe that this is beneficial to the study of aerosol chemistry.



Schematic experimental setup for second harmonic scattering measurements. The experimental setup includes three units. (1) Generation unit for SHS. The PHAROS femtosecond laser system centered at 1030 nm was used for the SHS experiments. The femtosecond laser system was focused on aerosol flows with a lens (a diameter of 1 in., a focal length of 10 cm). (2) Generation and handling unit for aerosol particles. This unit consists of an atomizer, a do-it-yourself sealed chamber, and an exhaust pump. (3) Collection and detection unit for SHS signal. A lens (a diameter of 2 in., a focal length of 2 cm) was used to collect SHS signal. A spectrometer with a liquid-nitrogen-cooled CCD and a PMT was used for dispersing light wavelength. The output from the PMT was followed by an amplifier and a single-photon counter for signal detection.

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