## Supplementary Information: Nanoscale Spectroscopic and Mechanical Characterization of Individual Aerosol Particles with Peak Force Infrared Microscopy

Le Wang,<sup>a</sup> Dandan Huang,<sup>b</sup> Chak K. Chan,<sup>b</sup> Yong Jie Li,<sup>c</sup> and Xiaoji G. Xu\*<sup>a</sup>

- a. Department of Chemistry, Lehigh University, 6 E Packer Ave. Bethlehem, PA, 18015. United States.
  b. School of Energy and Environment, City University of Hong Kong, Hong Kong, China
- c. Department of Civil and Environmental Engineering, Faculty of Science and Technology, University of Macau, Taipa, Macau, China

5 0 Signal (V) -5 Oscillation 10 Volume -15 change -20 0 2 6 8 10 12 4 14 Time ( 10-6 s) Pulsed IR laser Laser-induced response Focusing element Peak force tapping

Email: xgx214@lehigh.edu

Supplementary Figure S1. Scheme of the PFIR microscopy apparatus. The inset shows the laser-induced mechanical response due to the infrared resonances in the sample.

In brief, the peak force infrared (PFIR) microscopy is an action-based infrared microscopy that combines the peak force tapping mode of AFM<sup>1</sup> with infrared pulse excitation. As mentioned in the main text, the details of the PFIR are described in our article.<sup>2</sup> Supplementary Figure S1. conceptually shows the operational scheme of PFIR microscopy. The aerosol particles are supported on the flat substrate and are scanned under the feedback of peak force tapping of a Bruker Multimode 8 AFM. The infrared radiation from a quantum cascade laser (QCL, MIRcat, Daylight Photonics) is tunable in the mid-infrared frequency to match the vibrational resonances of the sample. The QCL emits laser pulse, and the timing of the pulse is synchronized with the intermittent contact of the AFM probe in the peak force tapping cycle. The infrared absorption of the sample leads to volume change (e.g., thermal expansion) and excitation of the contact resonance of the cantilever of the AFM probe. The position sensor of the cantilever deflection reads the thermal expansion through the detection of the baseline shift, and the excitation amplitude of the contact resonance oscillation. Because the amplitude of the laser-induced mechanical response is proportional to the absorption of the infrared laser, nanoscale imaging infrared absorption is achieved by registering the laser induced mechanical response while scanning the position of the AFM tip. At the same time, the mechanical information of the sample is simultaneously registered by the peak force tapping mode. A PFIR spectrum is obtained by registering the laser-induced mechanical response while sweeping the infrared frequency within the tuning range of the QCL, while the tip is held to a specific location in the peak force tapping feedback.

## Reference

- 1. B. Pittenger, N. Erina and C. Su, *Application Note Veeco Instruments Inc*, 2010.
- 2. L. Wang, H. Wang, M. Wagner, Y. Yan, D. Jakob and X. G. Xu, *Science Advances*, 2017, DOI: 10.1126/sciadv.1700255, Accepted.