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Optimization of Aggregation Kinetics of SERS-active Nanoparticles in Evaporating Sessile Droplets MEYSAM BARMI, CHRYSAFIS ANDREOU, MEHRAN HOONEJANI, MARTIN MOSKOVITS, CARL MEINHART, University of California, Santa Barbara — We studied evaporating sessile droplets containing silver nanoparticles as a platform for chemical detection using Surface-Enhanced Raman Spectroscopy (SERS), yielding molecule-specific Raman spectra. Controlling the degree of aggregation of nanoparticles is the key to the signal enhancement. Therefore, the aggregation kinetics of nanoparticles in droplets was investigated both experimentally and numerically to determine the evaporation and aggregation parameters leading to optimal detection. The signal depends on the degree of aggregation, which is affected by the initial concentration of nanoparticles (cNP), the dimerization rate (k), and the droplet evaporation time (τevap). We introduced the aggregation parameter Γa $\equiv$ τevap /τa, which is the ratio of the evaporation to the aggregation time scales. We found the different aggregation regimes based on aggregation parameter and stirring level within the droplet. For a well-stirred droplet, the optimal condition for SERS detection was found to be Γa,opt = k cNP τevap $\approx$ 0.3. The intensity of the SERS signal is near maximal in a wide range of aggregation parameters between 0.05 and 1.25 defining the time window during which trace analytes can be measured.

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