

***In-situ* optical spectroscopy to capture the birth of morphology**T. Adachi<sup>1</sup><sup>1</sup>Department of Physical Chemistry, University of Geneva

Self-organization of individual atoms/molecules into ordered structures results in the emergence of various physical properties. A fascinating aspect of the self-assembly is that atoms and molecules can choose slightly different packing structure that severely changes the resulting property. In a wide range of scientific to industrial researches, it is critical to control the structure of the assembly so that a desired property can be obtained. The microscopic description of how the “birth of morphology” occurs is yet to be established, which prevents one from rationally design the property of matters in solid phase.

The biggest challenge to experimentally probe the birth of morphology (i.e. nucleation process) is the stochastic and heterogenous nature of the nucleation occurring at nanometer scale. This has been particularly detrimental for the application of optical spectroscopy in this field. In our group, we have been developing *in-situ* optical spectroscopy tool to address this problem to establish microscopic understanding of the early stage of nucleation and growth of matters.

The first technique I will highlight is called Single Crystal Nucleation Spectroscopy (SCNS). SCNS is based on the combination of optical trapping and Raman microspectroscopy, which confines one crystal nucleation event under a Raman probe light. This technique allows us to follow the Raman spectroscopic feature of crystal nucleation at the tens of ms time resolution. We applied this technique on the crystallization of glycine in pure water. Through the spectral analysis, we could identify the formation of prenucleation aggregates as well as polymorph formation pathway.

The second technique I will highlight is called time-resolved dynamic light scattering microscopy (time-resolved micro-DLS). DLS is a commercially available tool to characterize the particle size in solution at the range of nm to  $\mu\text{m}$ . While it is quick and easy to use, it is applicable only for a static system because the measurement typically takes tens of seconds to minutes. To follow the formation of nucleus and growth in solution, it is desirable to improve the time resolution of this technique to ms time scale. We have developed a system and software that allows one to extract the particle size distribution of solution from light scattering data as quick as 40 ms acquisition. I will show the results obtained on model nanoparticle systems to highlight its capability of characterizing monomodal and multimodal size distributions (static system, not the size evolution yet) at tens of ms time resolution. This technique will widely broaden the application of DLS towards the non-static system in which the particle size distribution evolves over time.