

Studying Diffusion of Colloidal Nanoparticles in Solution Using Liquid Phase TEM and Machine Learning

Vida Jamali^{1,*}, A. Paul Alivisatos^{1,*}

¹. Department of Chemistry, University of California – Berkeley, Berkeley, CA, United States.

* Corresponding author: vidaj@berkeley.edu, paul.alivisatos@berkeley.edu

Motion and dynamics of nanoscale particles and macromolecules in bulk solutions and at interfaces are of fundamental importance in many areas of science and engineering. Liquid phase transmission electron microscopy (LPTEM) is a promising technique that allows us to probe such dynamics with nanometer scale resolution and in real time [1]. Yet, a challenge in the field has been in understanding and controlling the effect of the electron beam of the microscope on the dynamics of the system under study. Previous work has shown that nanoparticles diffuse very slowly inside the liquid cell chamber of LPTEM [2,3,4]. However, the underlying mechanism of diffusion and how that is affected by the electron beam dose rate of the microscope is not well understood. A key challenge in studying this effect is that we often do not have access to a large ensemble of spatiotemporal trajectories from LPTEM experiments. This makes canonical statistical tests including mean squared displacement insufficient for extracting information about the underlying mechanism of diffusion and the physical parameters such as anomaly diffusion exponent from the trajectory data.

Here, we present a supervised machine learning-based workflow, dubbed MoNet, that classifies the mechanism of diffusion manifested by the nanoparticles moving near the silicon nitride membrane of the liquid cell using only few hundred frame-long single trajectories [5,6]. MoNet is a dilated convolutional deep neural network that has an architecture inspired by the canonical p-variation test [7] and is trained on tens of thousands of simulated trajectories from three classes of diffusion with ground truth behavior [6]. The dilated architecture of the neural network allows us to capture the multiresolution correlations that may exist along the frames of the trajectory. To study the mechanism of diffusion of LPTEM trajectory data, we collected a large dataset from the diffusive motion of a model system of gold nanorods dispersed in water and moving near the silicon nitride membrane of the liquid cell, and over a broad range of electron beam dose rates (2 to 50 e⁻/Å²s) (**Figure 1**). The results from MoNet shows that gold nanorods move in an anomalous way inside the liquid cell chamber. By increasing the electron beam dose rate, the mechanism of diffusion crosses over from a viscoelasticity-mediated fractional Brownian motion (FBM), to an energy landscape-mediated continuous time random walk (CTRW) (**Figure 2**). This change in the mechanism of diffusion is indicative of how the material properties of the environment is affected by the electron beam of the microscope. We explain this crossover by the presence of silanol molecular groups on the surface of the silicon nitride membrane of the liquid cell and the changes in the electrochemistry of the solution environment with increasing the electron beam dose rate of the microscope [8].

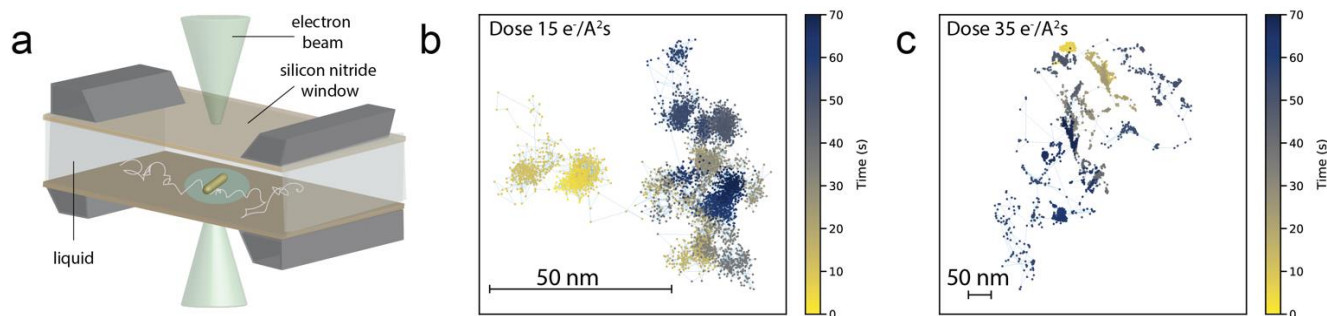


Figure 1. Schematic of a liquid cell TEM chamber (a) and example trajectories collected from gold nanorods diffusing in water and near the SiN_x membrane at dose rates of 15 (b) and 35 (c) e⁻/Å²s, respectively.

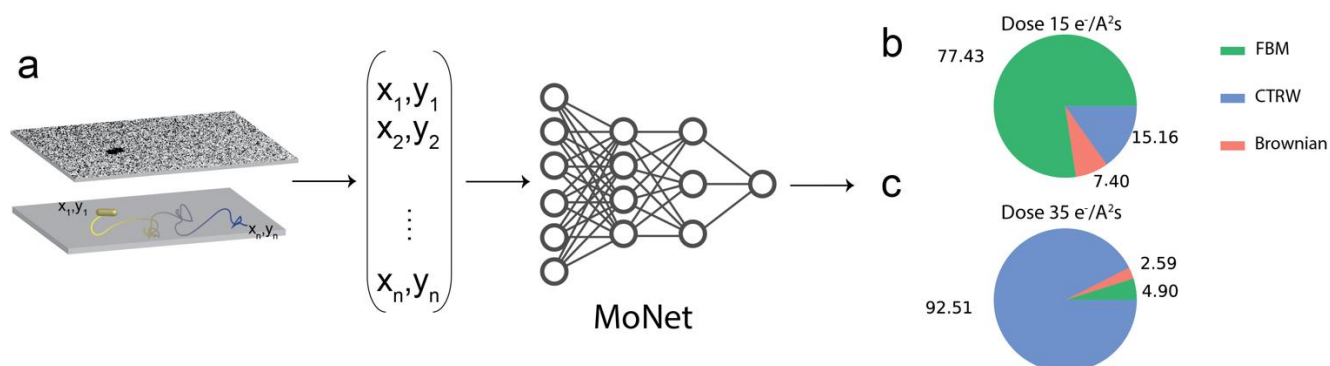


Figure 2. a) Schematic of the MoNet workflow for analyzing spatiotemporal trajectories from LPTEM experiments. (b-c) Probability values of the three classes of diffusion (FBM, Brownian, and CTRW) as outputted by MoNet for the trajectories shown in Figure 2 (b-c).

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