A Review of Measuring Coupled Brownian Motion and Rotation

Liu Haoyang

February 10, 2025

1 Introduction

Since Einstein proposed the physical explanation for Brownian motion in 1905, it has sparked extensive research across various fields. His work not only inspired the Langevin equation, paving the way for non-equilibrium statistical mechanics, but also laid the foundation for applications in diverse disciplines such as biology and finance. Through Einstein's diffusion theory, we understand that by measuring key quantities such as the velocity autocorrelation function (VACF) and mean-squared displacement (MSD), we can calculate the diffusion coefficient of a Brownian particle. This provides valuable insights into the system's viscosity and other important physical properties, thereby enhancing our understanding of fluid dynamics and molecular behavior in various contexts [1]. This review aims to introduce recent methods and achievements in measuring coupled Brownian motion and rotation.

2 Two-Dimensional System

The vast majority of studies focus on observing two-dimensional or near-surface dynamic Brownian motion, often supplemented by computational simulations. The preference for a two-dimensional system is evident: the correlation matrix is reduced to 3×3 instead of 6×6 . In this case, only two physical quantities are required to describe orientation, one to characterize rotation, and three to capture the coupling between translation

and rotation. This simplification not only makes data analysis more tractable but also allows researchers to explore a broader range of particle shapes.

2.1 The Center of Hydrodynamic Stress

This section is about the center of hydrodynamic stress (CoH), at which all coupled diffusion coefficients vanish. Ayan Chakrabarty implemented microfabricated boomerang particles with unequal arm lengths as a model for non-symmetry particles and study their Brownian motion in a quasi-two dimensional geometry by using an electron-multiplying charge coupled device (EMCCD, Andor Technology) [2]. They obtained videos of individual isolated asymmetric boomerangs. Using a high precision tracking algorithm, they determined the position and orientation of asymmetric boomerang particles. To measure all elements of the diffusion coefficient tensor, they converted the displacements in the lab frame into those in the body frame through a rotation transformation. Using this data, diffusion coefficients and coupled diffusion coefficients were calculated. And they also noticed all translation-rotation and translation-translation coupling disappear when the point fixed to the particle at CoH is used for motion tracking. So their experimental results present the first experimental evidence that in 2D the CoH exists even for Brownian particles with no symmetry.

In other paper, using same method and material, **Ayan Chakrabarty** shown that the mean displacements were biased towards CoH, and that the mean-square displacements exhibited a crossover from short time faster to long time slower diffusion with the short-time diffusion coefficients dependent on the points used for tracking [3].

2.2 Optical Trap

Faegheh Hajizadeh elucidated the Brownian dynamics of gold nanorods that are trapped and rotated at high frequency in a 2D optical trap [4]. They used quadrant photodiode (QPD) to probe real-time nanoparticle lateral displacement. The rotational motion of the nanorod were independ ently analyzed through the fiber-coupled avalanche photodiode (APD)/autocorrelator setup. They showed that, for typical settings, the effective rotational and translational Brownian temperatures are drastically different, being closer to the nanorod surface temperature and ambient temperature, respectively.

O.M. Marago study the Brownian motion of optically trapped graphene flakes [5]. Positional and angular displacements of a flake in the optical trap are detected by back focal plane (BFP) interferometry using the forward scattered light from the trapped particle. Using this data, they evaluated the temporal correlations between the particle tracking signals and explaint the flake dynamics, measure force and torque constants and derive a full electromagnetic theory of optical trapping.

There are two modes of microparticle rotation controlled by the interplay between magnetic and viscous torques. In magnetic fields rotating at low frequencies, the microparticles rotate synchronously with a rotating magnetic field. Once the frequency of the magnetic field rotation exceeds some critical value ω , the microparticle motion becomes asynchronous. *Maria N. Romodina* studied single magnetic microparticle rotation using optical tweezers combined with four electromagnets [6]. Charge-coupled device (CCD) camera and QPD were used to collect signal. Finally they found that the transition between the synchronous and asynchronous modes of particle rotation exhibits a smooth shape without any fracture because of the presence of Brownian torque.

2.3 Carbon Nanofibers and Carbon Nanotubes

Adrian Neild used an Olympus BX51 optical microscope to record isolated carbon nanofibers near plane surface (contact with a wall or in very close proximity) [7]. Then using same method as [2], they found that both the rotational and translational diffusion coefficients are strongly affected by hydrodynamic interactions with the walls and the tethered rods demonstrate a coupling between translational and rotational diffusive effects.

Nikta Fakhri studied the thermal diffusion of individual single-walled carbon nanotubes (SWNTs) confined in porous agarose networks using near-infrared video microscopy [8]. From video microscopy, they got many images, which was used to calculated the mean-square angular displacement (MSAD) and MSD. They found that even a small bending flexibility of SWNTs strongly enhances their motion: The rotational diffusion constant is proportional to the filament-bending compliance and is independent of the network pore size.

2.4 Others

Daniel B. Mayer studied the 2D motion of colloidal dimers by single-particle tracking and compare the experimental observations obtained by bright-field microscopy to theoretical predictions for anisotropic diffusion [9]. Similarly, they used two frame: laboratory frame and particle frame to do analysis of translational-rotational coupling. But additionally, they suggested self-intermediate scattering functions (ISF) to study spatiotemporal behavior.

Guanglai Li examined swimming trajectories of the singly flagellated bacterium near a glass surface with dark-field and total internal reflection fluorescence (TIRF) microscopy [10]. TIRF microscopy uses the evanescent wave of total reflection to illuminate fluorescently-labeled cells. The intensity of evanescent light decreases exponentially as the distance from the glass surface increases. Only cells very close to the surface can be imaged, and the distance of the cell to the surface can be calculated from the intensity of its fluorescence image. They found that when a bacterium swims near a surface, it experiences a larger drag than when in bulk fluid.

Ruben W. Verweij studied the Brownian motion of flexible trimers and found features that are unique to flexible objects. They found a hydrodynamic coupling between conformational changes and translations perpendicular to the particles long axis, which they call the Brownian quasiscallop mode because of its resemblance to scallop propulsion at high Reynolds numbers [11].

3 Three-Dimensional System

Daniela J. Kraft synthesized colloidal particles with various anisotropic shapes and tracked their orientationally resolved Brownian trajectories using confocal microscopy [12]. In their paper, the results of three different colloidal particles (regular trimer, regular tetramer, and irregular trimer) were given. With those data, they calculated short-time correlation functions and hydrodynamic friction tensor \mathcal{H} , which includes off-diagonal terms coupling the three translational and three rotational degrees of freedom.

Kazem V. Edmond used high-speed confocal microscopy to directly visualize the 3D translational and rotational motion of tetrahedral clusters in a dense amorphous suspension of colloidal spheres, which serve as tracers in colloidal supercooled fluids [13].

With those data, they obtained the translational and rotational trajectories for clusters and calculated MSD and MSAD. Finally they found that wide variety of systems can exhibit decoupling and the fundamental nature of diffusion is changed near the glass transition.

4 Summary of Methodology

- **S. M. Anthony and Y. Yu** wrote a review about recent advances in single-particle rotational tracking [14]. Also they summarised some of the methods, including:
 - Creating optical anisotropy using surface-tethered fluorescent nanoprobes
 - Tracking rotation of rods and ellipsoids
 - Tracking rotation of colloidal clusters

These three methods are very common, and they were used in most of the papers I mentioned earlier. But they also motioned another methods:

- Tracking particle rotation with digital holographic microscopy (DHM)
- Tracking rotation of modulated optical nanoprobes (MOONs)

The principle of first method is that DHM could generate a 2- dimensional hologram recording the light wave-front information. The resulting 2D holographic image encodes the 3D details of the object. Simultaneous measurements of all six modes of translational, rotational and vibrational dynamics are achieved by fitting the holograms with appropriate scattering models. But it's a computationally expensive process. MOON particles are an example of spherical Janus particles. Janus particles are particles that have distinct chemistries on opposite sides. Just as one detects the rotation of the moon by observing changes in its phases, people detect the rotation of Janus particles by changes in fluorescence or reflected light.

5 Conclusion

In this review, I have summarized recent methods and advancements in detecting the motion and rotation of individual particles. However, most of the work still focuses on two-dimensional systems. Researchers analyze video images and calculate quantities such as MSD, MSAD, and the coupling between translational and rotational motion by tracking the changes in lab frame and body frame. While the particles being observed are more diverse and the systems applied vary, the methods used remain quite similar to those outlined in the 2006 paper. Of course, two studies also used confocal microscopy to track the three-dimensional case, which may be similar to ODMR tracking translational motion, since ODMR also uses confocal microscopy. Surprisingly, there are also articles that use light to record information about the translation and rotation of particles. I think this should be a better way, because analysing directly from video images will inevitably produce errors, but using light may avoid such errors.

References

- [1] Xin Bian, Changho Kim, and George Em Karniadakis. 111 years of brownian motion. 12(30):6331–6346.
- [2] Ayan Chakrabarty, Andrew Konya, Feng Wang, Jonathan V. Selinger, Kai Sun, and Qi-Huo Wei. Brownian motion of arbitrarily shaped particles in two dimensions. 30(46):13844–13853.
- [3] Ayan Chakrabarty, Andrew Konya, Feng Wang, Jonathan V. Selinger, Kai Sun, and Qi-Huo Wei. Brownian motion of boomerang colloidal particles. 111(16):160603.
- [4] Faegheh Hajizadeh, Lei Shao, Daniel Andrn, Peter Johansson, Halina Rubinsztein-Dunlop, and Mikael Kll. Brownian fluctuations of an optically rotated nanorod. 4(7):746.
- [5] O. M. Marago, F. Bonaccorso, R. Saija, G. Privitera, P. G. Gucciardi, M. A. Iati, G. Calogero, P. H. Jones, F. Borghese, P. Denti, V. Nicolosi, and A. C. Ferrari. Brownian motion of graphene.
- [6] Maria N. Romodina, Evgeny V. Lyubin, and Andrey A. Fedyanin. Detection of brownian torque in a magnetically-driven rotating microsystem. 6(1):21212.

- [7] Adrian Neild, Johan T. Padding, Lu Yu, Basanta Bhaduri, Wim J. Briels, and Tuck Wah Ng. Translational and rotational coupling in brownian rods near a solid surface. 82(4):041126.
- [8] Nikta Fakhri, Frederick C. MacKintosh, Brahim Lounis, Laurent Cognet, and Matteo Pasquali. Brownian motion of stiff filaments in a crowded environment. 330(6012):1804–1807.
- [9] Daniel B. Mayer, Erick Sarmiento-Gmez, Manuel A. Escobedo-Snchez, Juan Pablo Segovia-Gutirrez, Christina Kurzthaler, Stefan U. Egelhaaf, and Thomas Franosch. Two-dimensional brownian motion of anisotropic dimers. 104(1):014605.
- [10] Guanglai Li, Lick-Kong Tam, and Jay X. Tang. Amplified effect of brownian motion in bacterial near-surface swimming. 105(47):18355–18359.
- [11] Ruben W. Verweij, Pepijn G. Moerman, Nathalie E. G. Ligthart, Loes P. P. Huijnen, Jan Groenewold, Willem K. Kegel, Alfons Van Blaaderen, and Daniela J. Kraft. Flexibility-induced effects in the brownian motion of colloidal trimers. 2(3):033136.
- [12] Daniela J. Kraft, Raphael Wittkowski, Borge ten Hagen, Kazem V. Edmond, David J. Pine, and Hartmut Lwen. Brownian motion and the hydrodynamic friction tensor for colloidal particles of complex shape. 88(5):050301.
- [13] Kazem V. Edmond, Mark T. Elsesser, Gary L. Hunter, David J. Pine, and Eric R. Weeks. Decoupling of rotational and translational diffusion in supercooled colloidal fluids. 109(44):17891–17896.
- [14] S. M. Anthony and Y. Yu. Tracking single particle rotation: probing dynamics in four dimensions. 7(17):7020–7028.