

Anomalous Dynamics of Colloids and Their Cluster Formation in Active Liquids

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Abstract. We examine the dynamics of individual colloidal particles in active media. Unlike Brownian particles, which display a Gaussian distribution in displacement, the colloids in active media show deviations from this form. These deviations depend on the size of the colloidal particles. We have characterized these deviations and interpreted the results using the concept of superstatistics. In addition, we also analyse the dynamic clustering of colloids at intermediated densities in the steady state using aggregation and fragmentation models, by constructing a transition matrix and applying a monomer approximation.

1 Introduction

Colloidal particles undergoing Brownian motion exhibit two key characteristics: their mean squared displacement (MSD) increases linearly with time—known as Fickian diffusion—and their displacement distribution follows Gaussian statistics. However, when colloids are suspended in an active medium, such as a bacterial suspension, their MSD shows superdiffusive behavior at short time scales and transitions to normal diffusion at longer times [?]. At short times, the displacement distribution deviates from a Gaussian form, displaying a Gaussian core with exponential tails. Over longer timescales, it gradually approaches a Gaussian distribution.

Interestingly, for larger colloids, where thermal fluctuations are negligible and motion is governed solely by the activity of the surrounding medium [1], the displacement distribution remains non-Gaussian even at long times. However, even in this diffusive regime, deviations from Gaussian statistics may persist [2]. Instances of non-Gaussian displacement distributions with linear MSD—referred to as “Fickian yet non-Gaussian” diffusion—have been observed [3]. Such deviations may arise from spatial or energetic disorder, viscoelastic effects, environmental heterogeneity, or fluctuating instantaneous diffusivities. To explain these phenomena, superstatistical framework [4] and diffusing diffusivity models [5] have been developed. These approaches treat diffusion as an ensemble of local environments, each characterized by different diffusivities sampled from statistical distributions (e.g., Gaussian, Gamma, or Laplace), thereby capturing the heterogeneous and fluctuating nature of diffusion in complex media.

Additionally, assemblies of these large particles at intermediate densities exhibit dynamic clustering behavior,

where clusters continuously form and break apart. Analysis of cluster statistics at steady state reveals a clear trend toward the monomer limit. The corresponding transition matrix for aggregation and fragmentation supports this, and the dynamics can be described by a solvable kinetic model under the condition of detailed balance.

In this study, we investigate the non-Gaussian displacement distributions of passive colloids in an active bacterial bath, focusing on the regime where MSD becomes linear at long times. We also examine the kinetics of colloidal cluster formation and fragmentation to better understand the dynamic steady-state behavior of such systems.

2 Results on displacement PDF of isolated colloids suspended in active liquid

To investigate the PDF of displacement of a single particle suspended in the active liquid, we considered a suspension of *E. coli* and polystyrene colloids. The experimental procedure is detailed in SI. We used colloids of two sizes, $7\mu\text{m}$ and $31\mu\text{m}$, with a fixed density of bacteria $c_b = 5b_0$ where $b_0 = 6 \times 10^9$ cells/ml. The area fraction of colloids was maintained at $\phi \sim 0.005$ to ensure they remained isolated, minimizing interactions between them. Figure 1(a)-(e) shows the MSD and PDF for $7\mu\text{m}$ particles at various lag-times, which are well beyond the crossover time from superdiffusive to linear in MSD, indicated by dashed lines in the colors as corresponding PDF, shown in the Fig 1(a). Figure 2(a)-(e) presents the MSD and PDF for $31\mu\text{m}$ particles. The dashed black curve represent the Gaussian fitting, while the solid black curves show the Gaussian + exponential fitting as described in the Eq. 1.

$$P(dx) = \frac{a}{b} \exp\left(-\frac{dx^2}{2b^2}\right) + \frac{c}{d} \exp\left(-\frac{|dx|}{d}\right) \quad (1)$$

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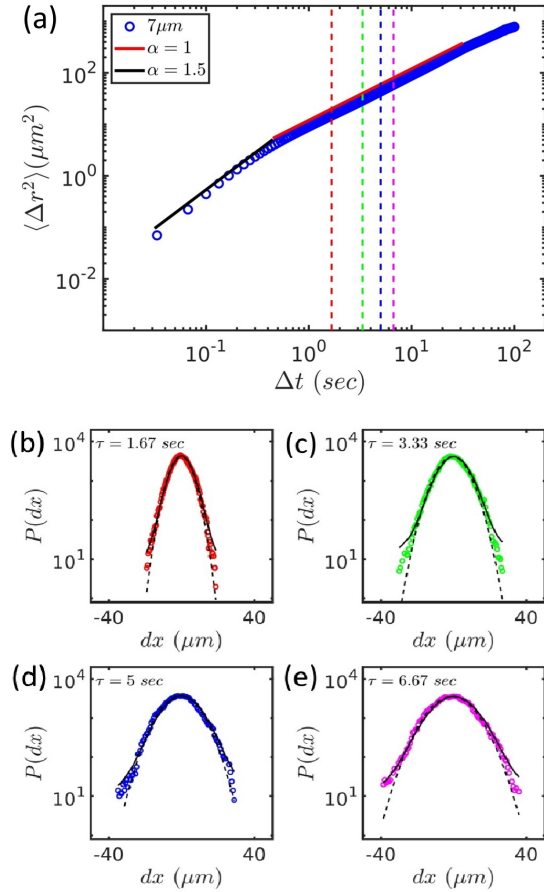


Figure 1. Mean square displacement (MSD) and Probability distribution function (PDF) of displacement for $7\mu\text{m}$ particles at bacteria density $c_b = 5b_0$, (a) black and red solid lines are showing the slope of 1.5 and 1 respectively to indicate the crossover from super-diffusive to normal diffusive regime, vertical dashed lines are showing the various lag-times for PDF: (b) for $\tau = 1.67 \text{ sec}$, (c) for $\tau = 3.33 \text{ sec}$, (d) for $\tau = 5 \text{ sec}$, (e) for $\tau = 6.67 \text{ sec}$, the dashed black curve is Gaussian fitting and black solid curve represents the Gaussian +exponential fitting.

The $31\mu\text{m}$ particles are large enough to have negligible Brownian motion compared to the $7\mu\text{m}$ particles. As shown in Fig. 1(a), the smaller particles fit well with the Gaussian distribution, whereas the larger particles fit the Gaussian and exponential distribution, as shown in Fig. 2(a). The non-Gaussian exponential core observed in the larger particles as presented in Fig. 2(b)-(e) may be attributed to their widely varying diffusivities over different time intervals along their trajectory. These aspects requires further investigations that are in progress. This work is ongoing, and these findings are the initial results.

3 Aggregation and fragmentation kinetic model

The ability of active matter to self-organize and exhibit various emergent behaviors is a well-established phenomenon. However, understanding how passive and thermal systems organize in the presence of active particles

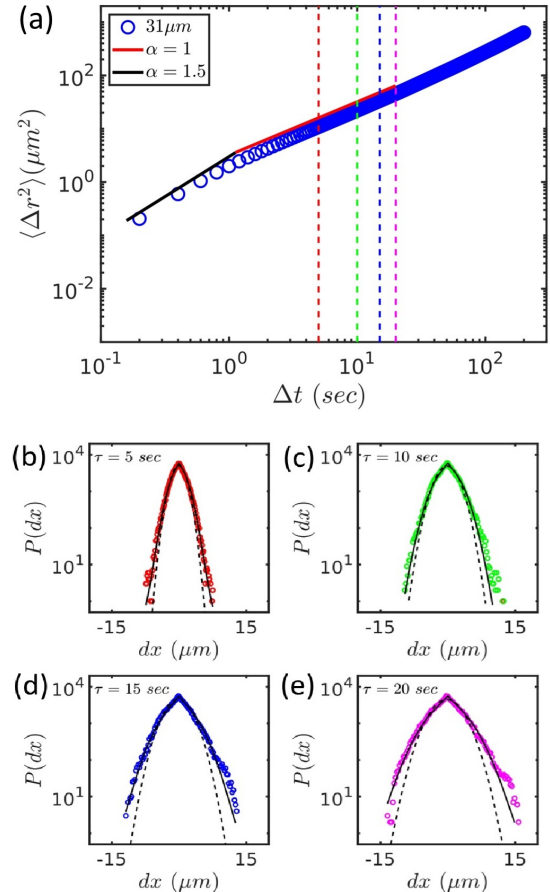


Figure 2. Mean square displacement (MSD) and Probability distribution function (PDF) of displacement for $31\mu\text{m}$ particles at bacteria density $c_b = 5b_0$, (a) black and red solid lines are showing the slope of 1.5 and 1 respectively to indicate the crossover from super-diffusive to normal diffusive regime, vertical dashed lines are showing the various lag-times for PDF: (b) for $\tau = 5 \text{ sec}$, (c) for $\tau = 10 \text{ sec}$, (d) for $\tau = 15 \text{ sec}$, (e) for $\tau = 20 \text{ sec}$, the dashed black curve is Gaussian fitting and black solid curve represents the Gaussian + exponential fitting.

remains an fascinating area of study. In all living matter, there is a mix of active and passive components. While the self-organization of passive components in passive environments is well understood and follows equilibrium statistics, their behavior in active environments is less explored. In this study, we present an experimental analysis of the cluster dynamics of non-Brownian passive colloids in an active liquid, revealing their continuous formation and disintegration by kinetic theory.

F. Ginot et al. [6] investigated the cluster phase of Janus active particles by analyzing the size-dependent fragmentation and aggregation rates. Their study rationalized the cluster size distribution and lifetimes using a simple kinetic approach with a monomer approximation, meaning that only single colloids take part in the aggregation and fragmentation events, as most transitions occur along the diagonal. In addition, it uses perimeter approximation, indicating that particles associate and dissociate primarily around the perimeter of clusters.

The aggregation-fragmentation kinetic model is a fundamental concept in the study of systems where various particles or clusters combine to form larger aggregates and break apart into smaller fragments. The elementary binary event can be represented as:



where, a cluster of size i and size j merges to form a cluster of size $i + j$ with aggregation rate $A_{ij}C_iC_j$. And the reverse process occurs with the fragmentation rate $F_{ij}C_{i+j}$. The general form of the time evolution of the cluster size distribution, $\dot{C}_n(t)$, is governed by the master equation:

$$\dot{C}_n(t) = \frac{1}{2} \sum_{i+j=n} A_{ij}C_iC_j - C_n \sum_{j \geq 1} A_{nj}C_j + \sum_{j \geq 1} F_{nj}C_{j+n} - \frac{1}{2} C_n \sum_{i+j=n} F_{i+j} \quad (3)$$

The kernels are time-independent, meaning the rates depend only on cluster concentrations and time-independent geometrical factors. This is a mean field theory, implying no spatial correlations between clusters. Therefore, the system must be sufficiently dilute for clusters to aggregate by diffusion.

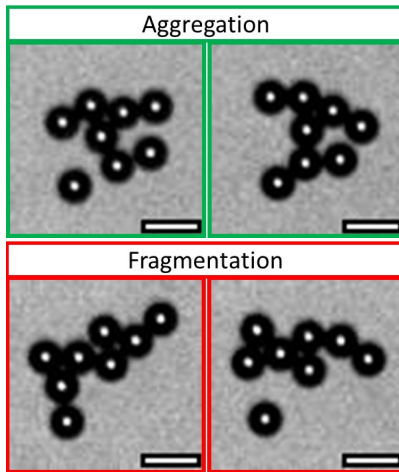


Figure 3. The bright-field images showing aggregation event (top) and fragmentation event (bottom).

The model consists of an infinite set of coupled non-linear first-order differential equations, making it impossible to solve analytically in general. However, solutions exist for specific systems under certain assumptions. To solve the rate equations, various analytical and numerical methods are employed. Analytical solutions are typically available only for simplified cases. Numerical methods, such as Monte Carlo simulations or discretized versions of the rate equations, are often used to study more complex systems. The above equation get simplified with the monomer assumption. The binary event with monomer approximation can be represented as:



The kinetic equation becomes:

$$\dot{C}_n(t) = A_{n-1}C_1C_{n-1} - A_nC_1C_n + F_{n+1}C_{n+1} - F_nC_n \quad (5)$$

By imposing the condition of detailed balance, with forward and backward rates in the Eq. 5, $F_{n+1}C_{n+1} = A_nC_1C_n$, are equivalent. We can get the steady state solution

$$C_n = \left[\prod_{m=1}^n \frac{A_{m-1}}{F_m} C_1^n \right] \quad (6)$$

with C_1 if fixed by normalization. Eq. 6 applies whenever A_n and F_n depends on the cluster size. For the particular case mentioned in [7, 8] where the exchange rates have a power law dependency with size: $A_n \sim n^\alpha$ and $F_n \sim n^{\alpha'}$, the cluster size distribution simplifies:

$$C_n \sim \frac{[(n-1)!]^{\alpha-\alpha'}}{n^{\alpha'}} C_1^n \quad (7)$$

In particular case, with $\alpha = \alpha'$ it further simplifies,

$$C_n \sim \frac{e^{-n/n_c}}{n^\alpha} \quad (8)$$

with $n_c = -1/\ln(c_1)$. The assumption $\alpha = \alpha'$ recovers the well appreciated cluster size distribution behaviour with power law and exponential cutoff at large n .

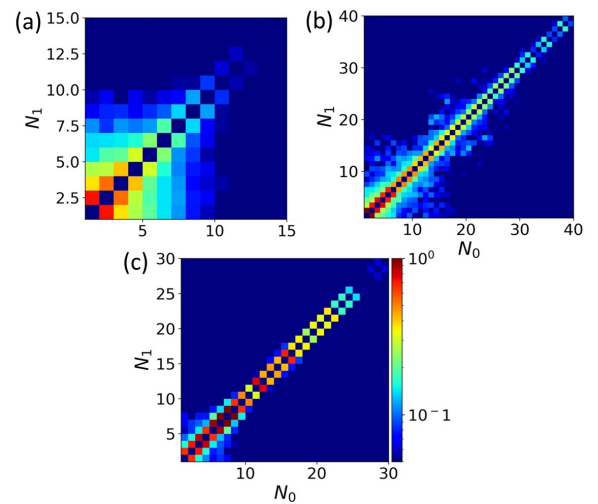


Figure 4. Transition matrix for the aggregation and fragmentation events (a) For particle size $10\mu\text{m}$ (b) For particle size $15\mu\text{m}$ (c) For particle size $31\mu\text{m}$ at the bacterial density $c_b = 10b_0$ with area fraction $\phi \sim 0.15$. The color bar is showing the logarithmic value of probability $P(N_0|N_1, \tau)$.

4 Results on cluster aggregation and fragmentation

In our system of bacteria and colloid mixture, colloids form dynamic clusters that continuously break and reform. We identified the clusters using a cutoff of 1.2σ . The bright field image of aggregation event and fragmentation event is shown in Fig. 3.

We calculated the transition matrix for cluster aggregation and fragmentation over a lag time of $\tau = 0.2$ sec, where N_0 represents the cluster size at time t_0 and N_1 represents the cluster size at time $t_0 + \tau$. The values in the

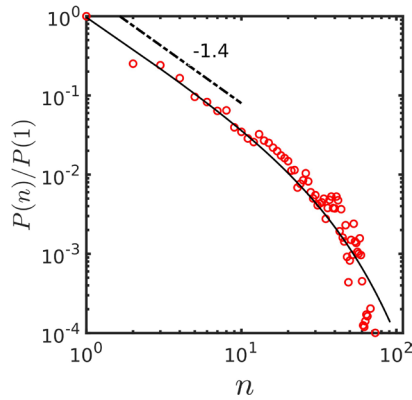


Figure 5. The cluster size distribution $P(n)/P(1)$ vs n the black curve is fitted with Eq. 8 with slope $\alpha \sim 1.4$.

transition matrix indicate the probability of a cluster transitioning from size N_0 to N_1 , denoted as $P(N_0|N_1, \tau)$, is shown in the Fig. 4. We have shown the transition matrix for various particle sizes $10 \mu\text{m}$, $15 \mu\text{m}$ and $31 \mu\text{m}$ at bacterial density $c_b = 10b_0$ with area fraction of colloids $\phi \sim 0.15$. The figure clearly shows that as the relative size of the colloids increases, the probability of single-particle aggregation and fragmentation events, which lie near the diagonal of the matrix, also increases. This observation strongly supports the monomer approximation, for which the master equation is exactly solvable. Although the transition matrix clearly indicates the presence of monomer approximation, it's important to note that our clusters are not solid structures; they are continuously restructuring and changing shape while rotating. The aggregation process does not occur with single particles merging directly into large clusters. Instead, due to the rotation of the clusters, particles are aggregating and detaching from the periphery, contributing to the dynamic nature of the system. Given that these results are preliminary, we have not yet calculated the aggregation and fragmentation rates. This work is still ongoing. However, we have determined the cluster size distribution and fitted it to Eq. 8. In Fig. 5, the cluster size distribution for particles with a size of $31 \mu\text{m}$ is shown. The black curve represents the fit to Eq. 8, with a slope of $\alpha = 1.4$. These results are reminiscent of cluster size distribution found in earlier studies of nonequilibrium systems undergoing aggregation, adsorption, and dissociation [9].

5 Conclusion

In summary, we investigated the dynamics of single particles suspended in an active bath, focusing on the mean squared displacement (MSD) and displacement distribution of colloids. Our findings reveal that the diffusive

behavior of colloids is strongly influenced by their size. For relatively smaller particles, the displacement distribution indicates Fickian diffusion, with Gaussian displacement statistics observed over longer time scales. Whereas, for larger particles where thermal effects are negligible, the dynamics are predominantly driven by the activity of the surrounding liquid. These particles still exhibit Fickian diffusion over the long duration, but their displacement distribution becomes non-Gaussian, characterized by a Laplacian core, especially at lower bacterial densities and with larger colloids. This phenomenon can be interpreted through the concept of superstatistics. Additionally, we analyzed the cluster dynamics of colloids in the steady state within the active bath using an aggregation and fragmentation kinetic model. The transition matrix provides clear evidence of monomer approximation in our system, allowing the kinetic equations to be exactly solvable under the detailed balance condition.

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