Quantifying active diffusion in an agitated fluid

– Electronic Supplementary Information (ESI) –

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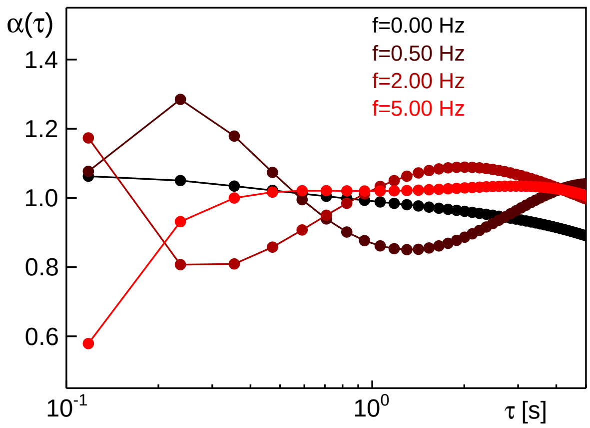

FIG. S1: Representative examples for the temporally varying apparent scaling parameter of ensemble-averaged TA-MSDs, $\alpha(\tau)$, as defined in the main text. While only minor deviations from the general scaling exponent $\alpha_e \approx 1$ are observed without stirring ($f = 0$ Hz), marked local fluctuations are seen for stirring frequencies in the range $f \sim 1$ Hz (cf. color-coded data at different frequencies). These fluctuations are closely associated with the transient bump observed in the associated MSDs (cf. Fig. 2 in the main text).

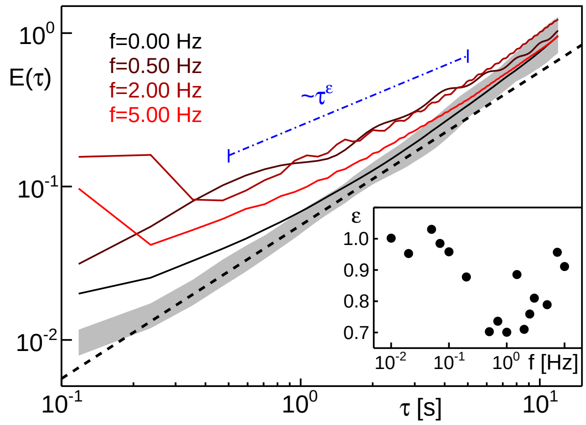

FIG. S2: Representative examples for the temporally varying ergodicity breaking parameter, $E(\tau)$, as defined in the main text. The analytical result for normal Brownian motion, $E(\tau) = 4\tau/(3N\Delta t)$, is shown as black dashed line. The grey-shaded region reflects the range of $E(\tau)$ observed for simulated random walks (20 ensembles of 100 trajectories of length N). Minor deviations from this prediction for $f = 0$ Hz at small lag times τ are attributed to a residual dynamic localization error. For stirred systems, marked deviations are seen over the entire range of lag times. Quantifying these empirically by a power law of the form $E(\tau) \sim \tau^{\varepsilon}$ (cf. blue dash-dotted line) highlights consistently lower values of ε for intermediate stirring frequencies (see inset). Therefore, stirring at intermediate frequencies induced a markedly higher spreading of individual TA-MSDs with respect to their ensemble average.

SIMULATIONS:

To probe the influence of varying diffusion coefficients on the amplitude of $G(\tau)$ we simulated ensembles of $M = 5000$ two-dimensional random walk trajectories, each having a length of $N = 500$ positions. Random step increments were calculated from Gaussian random numbers having zero mean and a variance $2D\Delta t$, with D being updated in every time step: For each trajectory, we first drew N random numbers ξ_i from a standard normal distribution. These were successively summed up to yield $S_k = \sum_{i=1}^{k} \xi_i$ with $k = 1, \ldots, N$. This eventually determined the varying diffusion constant, $D_k = D_0(1 + \varphi \frac{S_k - \min(S)}{\max(S - \min(S))})$ with the prefactor φ setting the maximum change of the mobility. In experiments, the enhancement factor $\max(D/D_0) = 1 + \varphi$ may depend on the stirring frequency as shown in Fig. 3b of the main text. To obtain a dichotomously switching mobility, D_k was replaced by $(1 + \varphi)D_0$ for all $D_k > \langle D \rangle$, and by D_0 otherwise. Examples for a continuously and a dichotomously varying diffusion coefficient are shown in Fig. S3, inset. For simplicity, we have set D_0, Δt to unity.

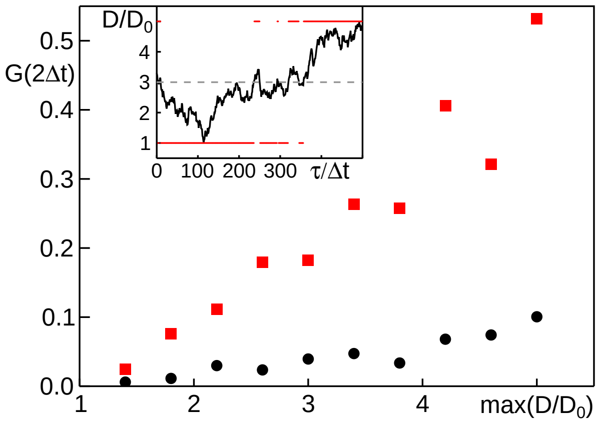

FIG. S3: The amplitude of the autocorrelation function $G(\tau)$ (defined via Eq. (4) in the main text), i.e. its first nontrivial value $G(2\Delta t)$, increases almost linearly with the enhancement factor $\max(D/D_0) = 1 + \varphi$ imposed in the simulations. While fairly large amplitudes are obtained for a dichotomous mobility change (red squares), considerably smaller values that are consistent with the experimentally observed ones (Fig. 4b in the main text) are seen for a continuously varying diffusion coefficient (black circles). Inset: Example for a continuously varying and a dichotomously varying diffusion coefficient (black and red lines, respectively).

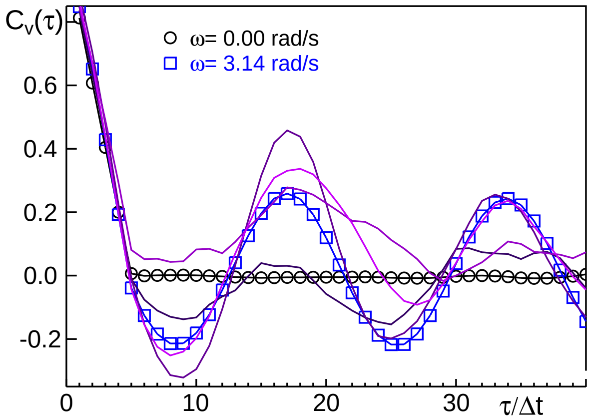

FIG. S4: Same plot as Fig. 6 in the main text with representative VACFs of individual trajectories superimposed (full colored lines). While the ensemble mean features very regular oscillations, individual VACFs show considerable fluctuations, highlighting that particles do not just show a deterministic periodic rocking motion due to the stirring.