

Broadband molecular dynamics simulation of fluid inertial effects in confined Brownian motion

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Hydrodynamic memory governs Brownian motion over a broad range of timescales, from acoustic wave propagation at short times to diffusive relaxation at long times. While confinement-induced corrections to Brownian diffusion are well established, how confinement modifies the full hydrodynamic response remains less explored. In this Letter, we use molecular-dynamics simulations of a neutrally buoyant colloidal particle in an explicit solvent to resolve the velocity autocorrelation function across a broad hydrodynamic spectrum. In the bulk, the simulations recover compressibility, added mass, the hydrodynamic long-time tail, and Stokes-Einstein diffusion without adjustable parameters. Near a rigid wall, the velocity correlations become anisotropic, their algebraic tails are modified, and the diffusion coefficients are reduced. Most importantly, the short-time dynamics reveals a pronounced enhancement of the effective added mass as the wall is approached. As such, the velocity autocorrelation function appears as a central quantity to bridge the zero-frequency mobility and the high-frequency inertial behaviour of a confined Brownian particle.

Brownian motion is one of the paradigmatic manifestations of thermal fluctuations and has played a central role in the development of statistical physics since the pioneering works of Einstein, Smoluchowski, and Langevin [1, 2]. The classical Langevin description assumes a memoryless process related to the instantaneous viscous response of the surrounding fluid allowing the dissipation of the particle momentum. This approximation leads to the Einstein relation, $D = k_B T / \gamma$ where γ is the friction coefficient. However, Brownian motion is not purely diffusive. Momentum transferred from a particle to the surrounding liquid is transported through the fluid and subsequently feeds back onto the particle motion. This hydrodynamic memory gives rise to algebraic long-time tails in the velocity autocorrelation function (VACF), as first revealed by Alder and Wainwright [3] and later described by fluctuating and unsteady hydrodynamics [4–8]. Modern experiments have directly observed this memory and resolved the crossover between inertial and diffusive regimes [10–12].

Boundaries enrich this physics by reorganizing the flow responsible for memory. The mobility of a Brownian particle near a rigid wall is known to be anisotropic, with diffusion coefficients parallel and normal to the interface that differ from their bulk values [9, 13–18]. However, diffusion probes only the zero-frequency limit of the response. The high-frequency limit, where the inertia of the accelerated fluid appears as an added mass and triggers memory effects, is harder to access [19–22] because it requires resolving the particle motion at short times. In this Letter, we show that molecular dynamics (MD) allow to resolve the full time window from the same observable. The VACF yields the diffusion coefficient by integration,

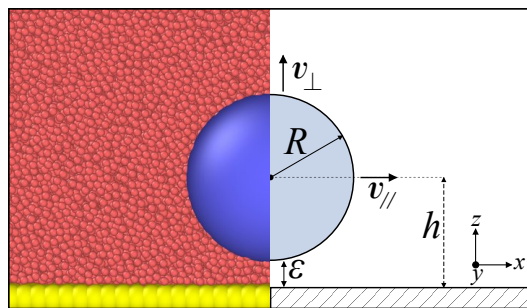


FIG. 1. Schematic of the system. Left: instantaneous snapshot of the MD simulation, showing the colloidal particle (blue, radius R) immersed in a dense WCA solvent (red) and confined near a planar wall (yellow). Right: notations are as follows; the surface-to-surface gap ϵ , the center-to-wall distance $h = \epsilon + R$, and the components v_{\perp} and v_{\parallel} of the colloid velocity, normal and parallel to the wall.

while its early-time structure gives the effective inertia of the particle-fluid system. From such an observable, we find that confinement changes not only the mobility, but the full hydrodynamic memory from the acoustic to the diffusive regimes.

We simulate a neutrally buoyant colloidal particle of radius R immersed in a dense Weeks-Chandler-Andersen solvent using a Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [23], in reduced Lennard-Jones units and fully periodic three-dimensional geometry. The solvent particles have mass $m = 1$ and diameter $\sigma = 0.6$, and the temperature and density are chosen to form a dense liquid ($T = 750, \rho_f = 3.3$). The colloid interacts with the solvent through a purely repulsive potential of range $R + \sigma/2$, and its mass scales as R^3 so as

to keep neutral buoyancy. The solvent density ρ_f , viscosity η , and sound velocity c_s are measured independently, fixing the compressibility time $\tau_c = R/c_s$, the viscous time $\tau_f = \rho_f R^2/\eta$, and the bulk added mass without adjustable parameters. The molecular model, wall preparation, sampling protocol, and uncertainty analysis are detailed in the Supplementary Material [24].

The starting point is the unsteady force exerted on a sphere in an incompressible fluid. In the Boussinesq-Basset-Oseen description, it reads:

$$\mathbf{F}_h(t) = -6\pi\eta R \mathbf{v}(t) - \frac{1}{2}M_f \dot{\mathbf{v}}(t) - 6R^2 \sqrt{\pi\eta\rho_f} \int_0^t \frac{\dot{\mathbf{v}}(\tau)}{\sqrt{t-\tau}} d\tau. \quad (1)$$

The three terms are from left to right : the Stokes drag, the added-mass contribution, and the Basset history force associated with vorticity diffusion. Equation (1) makes it explicit why the VACF is a natural observable: it contains dissipation, inertia, and memory in a single time-dependent quantity.

Figure 2(a) shows the bulk VACF computed from MD simulations. At very short times, the fluid is compressible and the acoustic time is directly measured as $\tau_c = R/c_s$ (Fig. 2(b)). This short-time regime is usually inaccessible in micron-scale colloidal experiments because the sound-crossing time is extremely small; in MD, it is resolved directly and provides a stringent test of the microscopic solvent dynamics. For $t \gg \tau_c$, the fluid behaves effectively as incompressible and the particle dynamics is governed by the effective mass $M^* = M_p + M_a$. The simulations recover the classical prediction, $M_a = M_f/2 = 2\pi\rho_f R^3/3$. Operationally, M^* is obtained from the early post-compressible VACF normalization and is independently checked from the change of slope of the mean-square displacement in the ballistic regime (Fig. 2(c)).

At long times, the same bulk data follows the expected asymptotic expression:

$$C_v(t) \simeq \frac{k_B T}{12\rho_f(\pi\nu)^{3/2}} t^{-3/2}, \quad \nu = \eta/\rho_f. \quad (2)$$

Green-Kubo integration gives the Stokes-Einstein diffusion coefficient over the range of particle radii investigated (Fig. 2(d)). The bulk simulations therefore validate the MD explicit solvent approach over the full hierarchy of hydrodynamic timescales, from sound propagation to vorticity diffusion, before confinement is introduced.

We now place the particle at a surface-to-surface distance ϵ from a rigid wall and decompose the velocity into components parallel and perpendicular to the wall (Fig. 1). For parallel motion, the particle height is held fixed while the in-plane motion remains free. For perpendicular motion, the particle is first equilibrated at the target height and then released, and the normal VACF is measured over a time window short enough that the gap

remains essentially unchanged. This protocol separates the two directional memory responses while avoiding an external trapping potential in the measured direction.

Confinement leaves the early acoustic response almost unchanged, but it strongly modifies the subsequent hydrodynamic relaxation. Once momentum has diffused over distances comparable to the wall separation, the flow is reflected and reorganized by the boundary. For a particle at center-wall distance $h = R + \epsilon$, Felderhof's asymptotic theory predicts [25]:

$$C_{\parallel}(t) \simeq \frac{k_B T(3h^2 - R^2)}{24\rho_f\pi^{3/2}} (\nu t)^{-5/2}, \quad (3)$$

for motion parallel to the wall, and:

$$C_{\perp}(t) \simeq \frac{k_B T}{8\rho_f\pi^{3/2}} \left[-\frac{R^2}{3} (\nu t)^{-5/2} + \frac{h^4}{4} (\nu t)^{-7/2} \right], \quad (4)$$

for normal motion. Figure 3 shows that the measured VACFs indeed display this wall-induced anisotropy. The central result is not merely that diffusion changes near a wall, but that the temporal structure of the memory kernel is reshaped: long-lived hydrodynamic correlations decay faster and become direction-dependent.

Having established that the wall reshapes the memory kernel itself, we now exploit the fact that a single VACF encodes two opposite limits of the same response. The low-frequency mobility is obtained from the diffusion coefficients, as:

$$D_i = \int_0^{\infty} C_i(t) dt, \quad i = \parallel, \perp, \quad (5)$$

and the high-frequency inertial response is obtained from the early post-compressible normalization $C_i \simeq k_B T/M_i^*$. These two limits are extracted from the same VACF, allowing confinement effects to be tested at low and high frequencies simultaneously. Figure 4(a) shows the normalized diffusion coefficients. Both components decrease monotonically with decreasing ϵ/R and agree with classical hydrodynamic mobility corrections, including Brenner's normal-drag result and near-wall expressions for parallel motion [2, 13, 26]. For normal motion, the small-gap limit recovers the lubrication scaling $D_{\perp}/D_0 \simeq \epsilon/R$, while the parallel component displays the weaker logarithmic reduction expected for shear-dominated flow. This confirms that the explicit molecular solvent reproduces the zero-frequency confined response.

The high-frequency response behaves in the opposite way. Instead of decreasing, the effective inertial contribution increases as the particle approaches the wall. We quantify this by the relative added-mass deviation:

$$\Delta M_i^*(\epsilon) = \frac{M_i^*(\epsilon) - M_i^*(\infty)}{M_i^*(\infty)}, \quad i = \parallel, \perp. \quad (6)$$

This counterintuitive enhancement reveals that confinement increases in fact the amount of fluid that is dynamically accelerated with the particle, despite the apparent

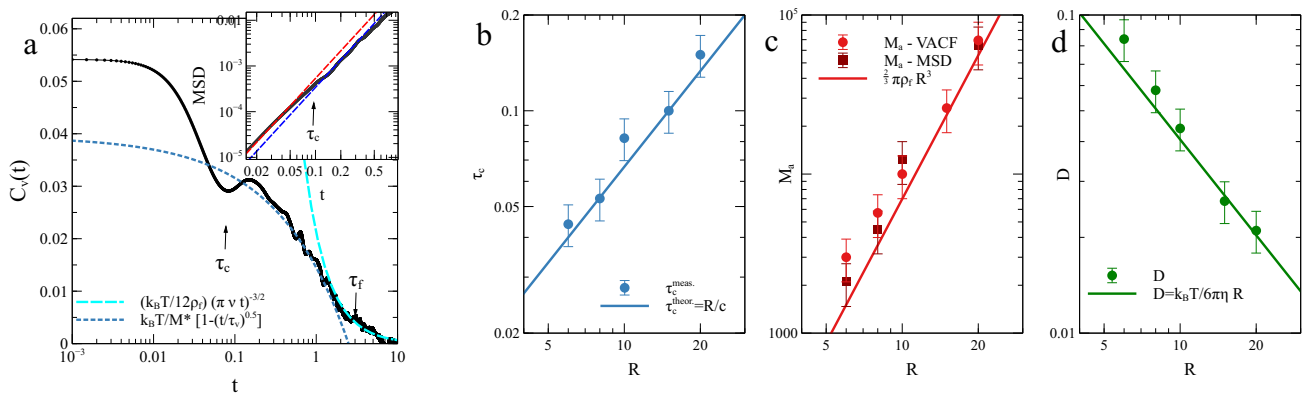


FIG. 2. Diffusion in the bulk. (a) VACF, C_v , of a colloidal particle of radius $R = 10$ immersed in a WCA fluid as a function of time t . Inset: MSD as a function of time in the ballistic regime showing the added mass transition. (b,c,d) Evolution with the colloid radius of the compressibility time τ_c , the added mass M_a estimated from the VACF and the MSD, and the diffusion coefficient D . The solid lines correspond to the theoretical predictions as indicated.

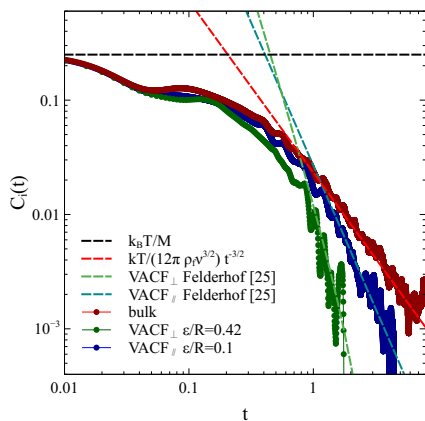


FIG. 3. VACFs C_i as a function of time t , measured near a rigid wall. Also shown are the bulk long-time tail, $t^{-3/2}$ given by Eq. 2, and the confined long-time tails $C_{\parallel} \sim t^{-5/2}$ and $C_{\perp} \sim t^{-7/2}$. Theoretical curves, Eqs. 3 and 4, are added [25].

reduction of fluid matter in the lubrication gap as compared to the bulk.

For normal motion, Yang's compact expression for the wall-induced mass amplification reads [27, 28]:

$$\Delta M_{\perp}^*(h^*) = 3 \sum_{n=1}^{\infty} \left[q^{-n/2} \sum_{k=0}^n q^k \right]^{-3}, \quad (7)$$

with:

$$q = \frac{h^* - \sqrt{h^{*2} - 1}}{h^* + \sqrt{h^{*2} - 1}}, \quad h^* = \frac{h}{R} = 1 + \frac{\epsilon}{R}. \quad (8)$$

Approximate expressions for parallel and perpendicular added masses near a wall give comparable trends and limits [29]. The agreement of the predictions with simulation data (see Fig. 4(b)) demonstrates that thermal

fluctuations in MD can probe an inertial quantity usually derived from deterministic potential-flow problems.

The coexistence of mobility suppression and inertia enhancement highlights a fundamental aspect of confined hydrodynamics. The diffusion coefficient measures $k_B T$ times the zero-frequency mobility, $D_i = k_B T \mu_i(0)$, while the added mass is reminiscent of the high-frequency inertial part of the same response. In a generalized Langevin description, for a spatial direction indexed by i , the Fourier transform of the VACF reads:

$$\tilde{C}_i(\omega) = \frac{k_B T}{-i\omega M_p + \tilde{\Gamma}_i(\omega)}, \quad (9)$$

where $\tilde{\Gamma}_i(\omega)$ is the frequency-dependent friction kernel. Thus D_i is controlled by $\tilde{\Gamma}_i(0)$, whereas M_i^* is encoded in the large- ω imaginary response. Confinement simultaneously suppresses mobility and enhances inertia as a result of different limiting manifestations of the same wall-modified hydrodynamic kernel. The low-frequency response is sensitive to no-slip boundary conditions and lubrication. The high-frequency response, by contrast, is governed by an effectively inviscid flow constrained by the same boundary. Agreement with both steady mobility corrections and potential-flow added-mass theories therefore indicates that the MD simulations with explicit solvent captures the boundary effects over a wide dynamical range. Deviations are expected only when the gap approaches the molecular diameter, where continuum hydrodynamics, the definition of the particle surface, and possible slip become ambiguous.

The above findings reveal why MD is particularly well suited to the present problem. Optical trapping and atomic force microscopy experiments measure confined Brownian motion with accuracy, but the external trap modifies the low-frequency response and the compressible regime is often beyond temporal resolution. By contrast,

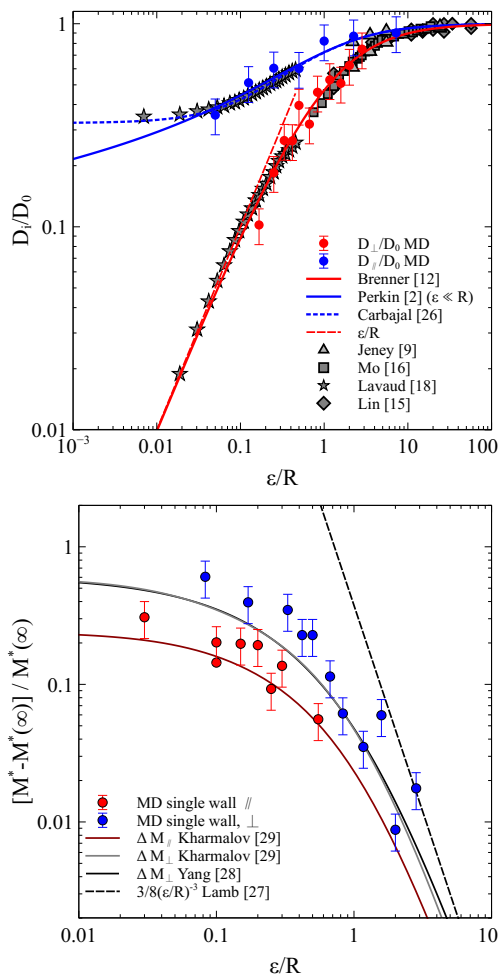


FIG. 4. (a) Normalized diffusion coefficients $D_{||}/D_0$ and D_{\perp}/D_0 as functions of the normalized distance to the wall ϵ/R . Also shown are theoretical predictions as well as experimental results from the literature, as indicated. (b) Relative added-mass deviation, $[M^*(\epsilon) - M^*(\infty)]/M^*(\infty)$, as a function of the normalized distance to the wall ϵ/R . Also shown are theoretical predictions from the literature.

the MD trajectories provide direct access to free thermal motion over the acoustic, inertial, hydrodynamic-memory, and diffusive regimes. Additional checks, including the dependence on particle radius, the robustness of mass extraction, and the two-wall Fauchoux-Libchaber geometry, are reported in the Supplementary Material [24].

A natural next step is to use Eq. (9) over the full frequency range. In practice, this requires a controlled transform of a noisy, finite-time VACF and careful treatment of the acoustic oscillations at the shortest times and algebraic tails at the longest times. The present data already contains this piece of information. Once regularized, the resulting kernel would allow to identify the crossover frequencies between compressibility, added mass, wall-modified vorticity diffusion, and steady mo-

bility. This perspective shows why the VACF is a more complete observable than the mean-square displacement: it retains the temporal ordering of hydrodynamic processes that are integrated out in the diffusion coefficient.

The resulting picture can be summarized in terms of two hydrodynamic limits. In the low-frequency limit, the particle explores a quasi-steady Stokes flow distorted by the presence of the wall. The relevant quantity is then the mobility tensor, and the details of the VACF have been integrated out. In the high-frequency limit, the particle accelerates before vorticity has diffused over the gap. The relevant quantity is then the kinetic energy of the surrounding fluid, which is altered by the presence of the wall. The same boundary condition therefore produces two opposite signatures: a reduced mobility and an increased inertia. This contrast is the central physical result of the present work, and the use of an explicit molecular solvent is essential for connecting the two extreme limits without changing models.

The approach also provides a practical way to assess the range of validity of continuum hydrodynamics. When the gap is large compared with the solvent diameter, the measured VACFs can be interpreted using continuum predictions for the algebraic tails, steady drag, and added mass. As the gap approaches molecular dimensions, several notions become less sharp: the position of the hydrodynamic wall, the surface of the colloid, and the effective slip length. To keep this comparison free of adjustable parameters, we restrict the present simulations to gaps ϵ large compared to the solvent molecular size. In this regime, the continuum predictions for the algebraic tails, the steady drag, and the added mass apply without fitting the position of the hydrodynamic wall, the surface of the colloid, or the effective slip length. At smaller gaps, these notions become less sharp, and outside the continuum description that is the focus of the present Letter. The statistical convergence and the sensitivity of the extracted mass are documented in the Supplementary Material [24].

Finally, the present formulation suggests extensions beyond a single rigid wall. A two-wall geometry mainly changes the low-frequency part of the response and is therefore included as an additional validation in the Supplementary Material [24]. More complex situations, such as patterned walls, soft interfaces, liquid-liquid boundaries, or viscoelastic media, should affect both the real and imaginary parts of $\tilde{\Gamma}_i(\omega)$ in a geometry-specific manner. The same VACF-based analysis could then distinguish whether an observed confinement effect is dominated by steady drag, elastic storage, inertial coupling, or acoustic propagation. In this sense, the present work is not only a numerical validation of known wall corrections, but also a demonstration of fluctuation-based hydrodynamic spectroscopy under confinement.

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