The role of dimensionality and geometry in quench-induced nonequilibrium forces

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Abstract

We present an analytical formalism, supported by numerical simulations, for studying forces that act on curved walls following temperature quenches of the surrounding ideal Brownian fluid. We show that, for curved surfaces, the post-quench forces initially evolve rapidly to an extremal value, whereafter they approach their steady state value algebraically in time. In contrast to the previously-studied case of flat boundaries (lines or planes), the algebraic decay for curved geometries depends on the dimension of the system. Specifically, steady-state values of the force are approached in time as $t^{-d/2}$ in $d$-dimensional spherical (curved) geometries. For systems consisting of concentric circles or spheres, the exponent does not change for the force on the outer circle or sphere. However, the force exerted on the inner circles or sphere experiences an overshoot and, as a result, does not evolve to the steady state in a simple algebraic manner. The extremal value of the force also depends on the dimension of the system, and originates from curved boundaries and the fact that particles inside a sphere or circle are locally more confined, and diffuse less freely than particles outside the circle or sphere.

Keywords: quench, Brownian, colloids, temperature, active matter, Active Brownian particles

(Some figures may appear in colour only in the online journal)

1. Introduction

Objects immersed in fluctuating media can experience forces for a variety of reasons. The prototypical example is that of fluctuation-induced forces (FIFs), also referred to as Casimir and van der Waals forces [1–4], which can arise because the objects modify fluctuation modes of the medium in the presence of long-ranged correlations. Interestingly, these FIFs exhibit many universal properties that are independent of details of the medium and the nature of the fluctuations (e.g., quantum or thermal), and emerge in different contexts ranging from atomic and molecular physics condensed matter, and biology to material science, chemistry and biology [5–8]. In
classical systems at thermal equilibrium, FIFs generally stem from long-ranged correlations in the vicinity of critical points. In nonequilibrium situations, however, long-ranged correlations can emerge much more generally [9, 10]. In particular, the conservation of different global quantities (e.g., the number of particles) can lead to constrained nonequilibrium dynamics which subsequently give rise to long range correlations and FIFs [11–13]. For classical systems, nonequilibrium FIFs have been previously explored in the steady-states of externally driven systems, e.g., in the presence of temperature or density gradients [14–17]. Recently, the role of sudden changes (quenches) of the temperature or of other system parameters in inducing a transient nonequilibrium states and FIFs has been studied by various groups [18–24].

While nonequilibrium dynamics following quenches has attracted much attention in the context of interacting quantum systems [25], various interesting phenomena also emerge in classical quench dynamics. In particular, it has been found that aside from FIFs, post-quench dynamics of the density in classical fluids can give rise to additional forces on immersed objects or surfaces [20, 21]. Such density-induced forces (DIFs) exist (and become longer-ranged) even in non-interacting (ideal) fluids. In contrast, the fluctuation forces discussed above rely on correlations, and disappear in the absence of interactions between fluid particles.

It is well-established that geometry and dimensionality play an important role in equilibrium Casimir-type (FIF) systems [27]. However, for forces induced by sudden quenches of temperature (or of activity in active fluids), these effects have not been considered thus far. In this paper, we study the transient forces following a quench of temperature in an ideal (Brownian) fluid confined by curved surfaces or inclusions. In reference [21] such post-quench DIFs were considered for flat walls immersed in an ideal fluid, and were shown to be independent of the dimension of the system and the boundaries: the force on the walls approaches its equilibrium value algebraically in time as $r^{-1/2}$. Here we aim to understand how this picture is modified by the curvature of the boundaries. We approach the problem with a theory for diffusion in curved geometries, as well as through explicit simulations of Brownian dynamics in the given setup. While our study applies to fluids confined by spherical or curved surfaces, it may also be of interest for the dynamics and behavior of fluid droplets in a quenched medium.

In addition to temperature quenches in colloidal passive systems or granular matter [28], our findings may be relevant for active non-interacting (or weakly-interacting) systems which, to some extent, can be modeled by Brownian motion with an effective temperature [20]. In such a setting, a sudden change in the activity of the active system (e.g., for active Janus particles [29]) confined to curved walls can be described by our formalism in the appropriate coarse-grained regime [30]. While the latter may be of interest especially for living active matter and biological systems, this would require a separate study on the importance of interactions and the role of solvent effects. Another possible experimental realisation of this problem could be to apply a fluctuating electric field to a system of charged Brownian particles in order to mimic the role of the temperature. One could then quench the ‘temperature’ by tuning the strength of the electric field [31–33].

The paper is organized as follows: we first introduce the model system and simulation details in section 2. Using a coarse-grained description, we determine the analytical solutions for the post-quench dynamics of the density field of Brownian particles, from which the forces on curved walls can be also obtained. Then, in sections 3 and 4, numerical and analytical results are presented for the pressure and the force exerted on circular and spherical boundaries, respectively. Finally, in section 5, we close the paper with a summarising discussion and conclusion.

2. Model and simulation details

We study dynamics of a non-interacting Brownian fluid, inside and outside of a sphere in $d$ dimensions, after a quench in temperature. In particular, we consider the pressure and force exerted on the confining spherical walls in terms of time dependence and their scaling behavior. To study the evolution of the system after the quench, we use a coarse-grained analytical approach and show that this model is in agreement with microscopic simulations based on Langevin dynamics.

2.1. Microscopic model for numerical simulations

In simulations, we model the walls of the sphere by a repulsive quadratic potential. Dynamics of the system is described by over-damped Langevin equations:

$$\frac{dr_i}{dt} = -\mu \frac{dV_{\text{wall}}}{dr_i} r_i + \eta_i(t),$$  
(1)

Here, $r_i$ is the position of the $i$th particle ($i = 1, 2, \ldots, N$), $\mu$ the mobility of the particles, and $V_{\text{wall}}$ represents the confining wall potential. When we simulate the interior of the $d$-dimensional sphere, we use

$$V_{\text{wall}}(r) = \frac{\lambda}{2} [\Theta(r - R_d)(r - R_0)^2].$$  
(2)

For simulations of the the exterior region we substitute $\Theta(R_d - r)$ by $\Theta(r - R_0)$. Interior and exterior regions are simulated separately, so that the corresponding pressures are independent [20, 21]. In equation (2), $R_d$ is the radius of the $d$-dimensional confining spherical boundaries. From now on and for the sake of clarity, we denote the radius in the case of only one sphere (circle) with $r_0$, whereas in the cases of two spheres (circles), the two radii are denoted by $r_1$ and $r_2$ with a difference $\Delta r$ (see figure 1). Also, $\Theta(x)$ represents Heaviside step function, which is equal to one for $x > 0$ and vanishes otherwise. The strength of the quadratic potential is given by $\lambda$, and is chosen such that the effective range of the wall potential (and correspondingly the thickness of the adsorbed particle layer at the surface) is significantly smaller than the confinement length scale. In equation (1), $\eta_i$ is Gaussian white noise obeying

$$\langle \eta_{i}(t) \eta_{j}(t') \rangle = 2D \delta(t - t') \delta_{ij} \delta_{\alpha\beta},$$  
(3)

where $D$ is the diffusion constant of the particles, which is related to the mobility by fluctuation–dissipation theorem.
according to $D = \mu k_BT$, and $\eta_i$ is the $i$th component of the noise exerted on particle $i$.

Using equation (1) and (2), we simulated the dynamics of the system of passive Brownian particles inside a spherical geometry in two or three dimensions, following the temperature quench. In all simulations, we average over $N = 10^5$ particles to obtain the pressure on the walls. In particular, we define a length-scale $\ell = \sqrt{\pi k_BT/(2\lambda)}$ associated with the wall potential. We also consider the characteristic timescale $\tau_0 = \ell^2/D = 1$ for diffusion, and choose the system size to be large enough so that it can be compared with the results of the coarse-grained model. In both circular and spherical cases, we perform simulations for $r_1/\ell = 100, 200$ and $\Delta r/\ell = 100$. A forward Euler method is employed to integrate equation (1) in order to study the post-quench dynamics of each particle. In simulations, we instantaneously change the temperature by changing the diffusion coefficient of the particles in equation (1). The overall scalar force exerted on the wall by the interior particles, from which we obtain the pressure, is then defined as the sum of the radial components of all single-particle forces as

$$F = \sum_{i=1}^{N} F_i \cdot \hat{r}_i = \sum_{i=1}^{N} \frac{dV_{wall}}{dr}|_{r_i}.$$  

(4)

To obtain the pressure, we simply divide the total force $F$ by the area of corresponding wall.

2.2. Coarse-grained model

We also provide a coarse-grained theoretical description based on the Smoluchowski equation, and study the evolution of the density of the non-interacting Brownian particles following the quench. The particle density is defined as $\rho(r,t) = \sum_{i=1}^{N} \delta(r - r_i(t))$. The Langevin equation (1) leads to a spherically symmetric coarse-grained dynamics given by

$$\partial_i \rho(r_i, t) = \frac{D}{\ell^d} \frac{\partial}{\partial r_i} \left[ \rho^{-1} \partial_i \rho(r_i, t) \right],$$  

(5)

in $d$ dimensions. In this coarse-grained picture, walls are assumed to impose no-flux boundary conditions at $r = R_d$:

$$\partial_i \rho(r_i, t)|_{r=R_d} = 0.$$  

(6)

Before the quench, the system is in a steady state described by the canonical Boltzmann weight $\rho_0 \propto \exp[-V_{wall}(r)/(k_BT)]$, where $k_B$ is Boltzmann’s constant. At time $t = 0$, we change the temperature to $T = T_F$ instantaneously. After this quench, the system evolves to a new equilibrium state which is described by $\rho_F \propto \exp[-V_{wall}(r)/(k_BT_F)]$. As discussed in reference [21], the quench modifies the boundary layer of particles close to the walls, because the penetration depth of the particles into the wall potential is temperature dependent. We can therefore write the post-quench particle density (for $t > 0$), as the sum of a homogeneous contribution, representing the region outside of the wall potential, and a time- and position-dependent excess density,

$$\rho(r_i, t) = \rho_0 + \Delta \rho(r_i, t).$$  

(7)

We consider the homogeneous part of the density $\rho_0$ to be the same inside and outside the inclusions. In the coarse-grained description, we consider strongly repulsive walls, so that the initial excess density after the quench is approximated as a $\delta$-function at the boundary,

$$\Delta \rho(r, t = 0^+) = \alpha_d \rho_0 \delta(r - R_d).$$  

(8)

Here $\rho_0$ and $\alpha_d$ scale with length as $[\rho_0] = 1/\ell^d$ and $[\alpha_d] = \ell$, respectively. Indeed, $\alpha_d$ corresponds to the change of the width of the boundary layer induced by the quench, and can be computed by integrating $\rho_1$ and $\rho_F$ over the relevant volume and enforcing conservation of the particle number. We calculate this parameter analytically in appendix A. We note that the density immediately after the quench is exactly the same as before the quench. However, for a sudden quench to a higher temperature, the wall becomes statistically more penetrable for the particles which then possess a higher average energy. Therefore, at $t = 0^+$, the wall accommodates less particles than the equilibrium state corresponding to the new temperature and we have desorption of particles on the wall. This gives rise to an effective desorption of some particles on the wall, which can be thought of as the emergence of a very thin layer of empty space at the wall. Conversely, if the temperature is suddenly lowered at $t = 0$, immediately after the quench additional particles accumulate on the wall (compared to the equilibrium state corresponding to the new temperature). The density contribution of the particles adsorbed or desorbed at the wall due to the nonequilibrium quench is represented by $\Delta \rho(r, t = 0^-)$ in equation (8). The excess density therefore has a negative (positive) value when the post-quench temperature $T_F$ is higher (lower) than $T_1$. After the quench, the adsorbed (or desorbed, depending on whether the quench is to a higher or lower temperature) layer of particles at the boundary diffuses...
into the system. This gives rise to dynamics of post-quench forces and pressures acting on the boundaries. It turns out that \( \alpha_k \) is independent of the system’s dimension; we therefore use the notation \( \alpha \) throughout instead.

Using separation of variables, we can solve equation (5) and find the evolution of the excess density in time. The ideal gas law then provides the corresponding instantaneous pressure exerted on Brownian particles on the \( d \)-dimensional spherical wall after the quench,

\[
P(r = R_d, t) = k_B T_f [\rho_0 + \Delta \rho(r = R_d, t)].
\] (9)

Solving equations (5), (6) and (8) for the region outside of the sphere, we can analogously find the evolution of the density of Brownian particles on the exterior of the sphere after the quench (similar no-flux boundary conditions apply at the wall). Subtracting the outside from the inside pressure exerted on the sphere, one obtains the force exerted per area \( A_d \) on the sphere following the quench:

\[
F(t)_{A_d} = k_B T_f [\Delta \rho_{out}(R_d, t) - \Delta \rho_{in}(R_d, t)].
\] (10)

In equation (10), \( \Delta \rho_{in/out} \) represents the excess density inside/outside the sphere. For time, pressure, and force, we define the dimensionless variables

\[
i = \frac{D t}{R_d^2}, \quad \bar{P}(t) = \frac{P(t)R_d}{\alpha_d \rho_0 k_B T_f}, \quad \bar{F}(t) = \frac{F(t)R_d}{\alpha_d \rho_0 k_B T_f A_d}.
\] (11)

Here \( R_d \) takes the values \( r_0, r_1, \) or \( r_2 \), depending on the considered geometry and the wall for which we calculate the pressure and force.

3. Quench in circular geometries (\( d = 2 \))

In this section we discuss the post-quench pressure and force acting on a single circular wall or on two concentric circles of different radii.

3.1. Inside the circle

Setting \( d = 2 \) in equation (5), we have

\[
\partial_t \rho(r, t) = \frac{D}{r} \partial_r [r \partial_r \rho(r, t)].
\] (12)

We then use separation of variables to write density as \( \Delta \rho(r, t) = X(r)T(t) \). Putting this density back to equation (12) we have

\[
\frac{\partial_t T}{T} = \frac{D}{Xr} \left( \partial_r X + r \partial_r^2 X \right) = -\zeta^2.
\] (13)

Solving for \( X \) and \( T \) we find

\[
T(t) = e^{-\zeta^2 t}, \quad X(r) = c_1 J_0 \left( \frac{\zeta r}{\sqrt{D}} \right) + c_2 Y_0 \left( \frac{\zeta r}{\sqrt{D}} \right),
\] (14)

where \( J_n \) and \( Y_n \) are \( n \)th order Bessel functions of the first and second kind, respectively [34]. The solution found in equation (14) should be finite inside the circle, so \( c_2 = 0 \). The no-flux boundary condition at the wall translates into

\[
\partial_r X(r)|_{r=r_0} = 0,
\] (15)

which implies that the parameter \( \zeta \) takes on discrete values. Defining \( \beta = \zeta r_0 / \sqrt{D} \), the allowed values for \( \beta \) are the positive roots of the Bessel function,

\[
J_1(\beta_n) = 0, \quad n = 1, 2, 3, \ldots
\] (16)

We can then write the time-dependent evolution of the density in the form of an infinite series,

\[
\Delta \rho_{in}(r, t) = \sum_{n=1}^{\infty} c_n e^{-\beta_n^2 t} J_0 \left( \frac{\beta_n r}{r_0} \right) + c_0,
\] (17)

using dimensionless time \( t' \) as defined in equation (11) with \( R_d = r_0 \). The appropriate initial condition for this problem (which involves the excess density at the wall) takes the form \( \Delta \rho_{in}(r, t = 0) = \alpha \rho_0 \delta(r - r_0) \), and the constant \( c_0 \) can be found by integrating over the disk and enforcing particle conservation:

\[
\int_0^{r_0} r \, dr \left[ \sum_{n=1}^{\infty} c_n J_0 \left( \frac{\beta_n r}{r_0} \right) + c_0 \right] = \int_0^{r_0} r \, dr \alpha \rho_0 \delta(r - r_0),
\] (18)

which gives \( c_0 = 2 \rho_0 \alpha / r_0 \). To find the remaining coefficients \( c_n \), we use the following orthogonality relation for the Bessel function [34]:

\[
\int_0^{r_0} x \, dx J_0(\beta_n x) J_0(\beta_m x) = \delta_{nm} \frac{[J_0(\beta_n)]^2}{2},
\] (19)

with \( \beta_n \)’s given by equation (16). This yields

\[
c_n = \frac{\alpha \rho_0 r_0 J_0(\beta_m)}{\int_0^{r_0} r \, dr [J_0(\beta_m r)]^2} = \frac{2 \alpha \rho_0}{r_0 J_0(\beta_m)}.
\] (20)

Putting everything together we find the density on the circle as

\[
\Delta \rho_{in}(r_0, t) = \left[ 1 + \sum_{n=1}^{\infty} e^{-\beta_n^2 t} \right] \frac{2 \alpha \rho_0}{r_0}.
\] (21)

3.2. Outside the circle

For the evolution of density outside the circle we similarly use equations (14) and (15), from which we obtain the following result:

\[
\Delta \rho_{out}(r, t) = \int_0^\infty d \gamma \, c(\gamma) G \left( \gamma, \frac{r}{r_0} \right) e^{-\gamma^2 t},
\]

\[
G(\gamma, x) = Y_1(\gamma) J_0(\gamma x) - J_1(\gamma) Y_0(\gamma x).
\] (22)

To find the coefficients \( c(\gamma) \), we insert the above result in the initial condition written in the form

\[
\int_{r_0}^{\infty} r \, dr \Delta \rho_{out}(r, t = 0) \bar{G} \left( \gamma', \frac{r}{r_0} \right) = \alpha \rho_0 \int_{r_0}^{\infty} r \, dr \delta(r - r_0) \bar{G} \left( \gamma', \frac{r}{r_0} \right),
\] (23)
and evaluate the integrals by invoking the orthogonality condition
\[
\int_1^\infty \ dx \delta(\gamma - \gamma') \frac{|J_1(\gamma)|^2 + |Y_1(\gamma)|^2}{\gamma} = 2 \alpha \rho_0 \frac{\pi r_0}{J_1(\gamma) + Y_1(\gamma)}.
\] (24)

This finally leads to
\[
c(\gamma) = \frac{-2\alpha \rho_0}{\pi r_0 [J_1(\gamma) + Y_1(\gamma)]}.
\] (25)

Then, the density on the boundary of the circle reads
\[
\Delta \rho_{\text{out}}(r_0, t) = \int_0^\infty \frac{4 \alpha \rho_0 e^{-\epsilon/t}}{\gamma^2 \pi r_0 [J_1(\gamma) + Y_1(\gamma)]} \, d\gamma.
\] (26)

In the long time limit, this excess density decays as
\[
\Delta \rho_{\text{out}}(r_0, t \to \infty) \approx \frac{\alpha \rho_0}{2 \epsilon r_0} \bar{t}^{-1}.
\] (27)

At long times after the quench, the excess pressure exerted on the circle from outside therefore approaches zero as \(\bar{t}^{-1}\).

Having computed the density of the particles inside and outside the circle, we can use equation (10) to find the force exerted on the circle after the quench. Figure 2 shows that the force approaches its new steady-state as \(\bar{t}^{-1}\) in time. As can be seen in the inset of the figure, the force exerted on the circle starts from the value \(F_{2D} = 1\) at very short times. A similar behavior is also observed for the force exerted on a sphere. Nevertheless, immediately after the quench one would expect the quench-induced force on the wall to be zero. Indeed, we show in appendix B (via an appropriate treatment of the summation in equation (21) and the initial condition) that at \(t = 0^+\) the quench-induced force vanishes. This means that at very short times there is an ‘abrupt’ transition from zero to the initial finite force seen in the inset of figure 2. This offset value for the force at very short times is a consequence of the curvature of the geometry of the circle and a clear manifestation of the nonequilibrium character of the quench-induced dynamics. The curvature of the wall implies that diffusion of the excess particles inside the circle occurs in a confined environment, in contrast to the outside region. As a result, and assuming initial excess densities which are entirely localized at the boundaries, this finite force is reached immediately (and discontinuously in time) after the quench. However, this observation is a consequence of the coarse-graining assumptions: if we attribute a finite small width \(\epsilon\) to the initial excess density (rather than the delta function), the force becomes finite after a very short time \(t \geq (\epsilon / r_0)^2\) (see appendix B for more details).

Here we focus on the dynamics of the quench-induced force, and we calculate the value of the total force in subsection 4.4.

### 3.3. Quench effect on the medium between two circles

In this part, we consider a system of non-interacting Brownian particles confined between two circles with radii \(r_1\) and \(r_2\), as shown in figure 1(c). The details of obtaining analytic solutions for the time evolution of the excess densities between the two circular boundaries can be found in appendix C. The results for the post-quench dynamics of the pressure and forces for two concentric circles are represented in figure 3. Both analytical and simulation results, shown in figure 3(a), indicate that the pressures exerted on the small and large circles by the confined particles, scale as \(\bar{t}^{-1/2}\) at short times after the quench. This behavior stems from the fact that at very short times the excess particle density is still very localized near the walls, and its evolution is effectively described by a one-dimensional diffusion along the direction locally normal to the wall. As will become clear in the next section, the \(\bar{t}^{-1/2}\) behavior of the pressure also occurs in a spherical geometry and can be considered as a universal short-term characteristic of the pressure in all dimensions and geometries.

We now turn to the force exerted on each circle. As shown in figure 3(b), the force exerted on the large circle approaches its steady-state as \(\bar{t}^{-1}\) at long times. This behavior is the same as for the force on a single circle (figure 2), because the late-time behavior is dominated by the density relaxation in the infinite outside medium. The early time behavior of the force on the smaller circle is also similar to the case of a single circle, and as figure 3(c) shows, the force starts of a finite value \(F_1 = 1\) after the quench which is similar to the case of a single circular wall. However, the late time behavior differs significantly: after an initial overshoot, which becomes stronger and takes place at earlier times for larger \(r_1\), a steady-state behavior is approached. To understand the non-monotonic behavior of the force exerted on the smaller circle, we note that due to the curvature of the walls, at small times after the quench, the particles from inside the circle move toward the walls more than those particles between the circles. Correspondingly, the force exerted on the smaller circle is positive. At later times, the particles which were initially located on the exterior circle (at \(t = r_2\)) find time to diffuse and reach the smaller circle and, as a result, the force exerted on the smaller circle decreases. This explains the overshoot seen in figure 3(c). Upon varying \(r_1\) while keeping \(\Delta r\) fixed, the steady-state force also changes and even undergoes a sign change for large \(r_1\) depending on the
balance between the particles inside/outside the small circle. In contrast to \( F_1 \), the force on the outer circle is always positive (toward outside) and monotonically increases, as shown in figure 3(d).

4. Quench in spherical geometries \( (d = 3) \)

In this section we investigate the phenomena considered in section 3, but in three spatial dimensions.

4.1. Inside the sphere

Dynamics of a diffusive system inside a sphere \( (d = 3) \) can be written as

\[
\partial_t \rho(r,t) = \frac{D}{r^2} \partial_r \left[ r^2 \partial_r \rho(r,t) \right].
\]  

Again we use separation of variables for the excess density, \( \Delta \rho(r,t) = R(r)T(t) \). Inserting this into equation \((28)\) we find

\[
\frac{\partial_t T(t)}{T} = \frac{D}{R r^2} \left( 2 r \partial_r R + r^2 \partial_r^2 R \right) = -\zeta^2.
\]  

Solving for \( R \) and \( T \) yields

\[
T(t) = e^{-\zeta^2 t}, \quad R(r) = \frac{A}{r} \cos \left( \frac{\zeta r}{r_0} \right) + \frac{B}{r} \sin \left( \frac{\zeta r}{r_0} \right),
\]  

where \( \zeta = \zeta r_0 / \sqrt{D} \). For the interior of a sphere, the first term in the solution for \( R \) in equation \((30)\) diverges at \( r = 0 \), so that one must put \( A = 0 \). The no-flux boundary condition on the interior surface of the sphere gives

\[
\partial_r R(r)|_{r=r_0} = 0.
\]  

Similar to the 2D case, we find discrete values for the parameter \( \beta \), which are now the positive roots of

\[
\beta_n = \tan \beta_n, \quad n = 1, 2, 3, \ldots
\]  

The time-dependent solution for the excess density is then

\[
\Delta \rho_m(r,t) = \sum_{n=1}^{\infty} c_n \sin(\beta_n r / r_0) e^{-\beta_n^2 t} + c_0.
\]  

In equation \((33)\), the summation is over all positive solutions of \( \beta \) in equation \((32)\). The initial condition \( \Delta \rho_m(r,t = 0) = \alpha \rho_0 \delta(r - r_0) \) allows us to find the constant \( c_0 \) in analogy to the 2D case by calculating

\[
\int_0^{r_0} r^2 \, dr \Delta \rho_m(r,t = 0) = \int_0^{r_0} r^2 \, dr \alpha \rho_0 \delta(r - r_0),
\]  

which gives \( c_0 = 3 \alpha \rho_0 / r_0 \). The coefficients \( c_n \) are obtained by applying the orthogonality condition

\[
\int_0^1 dx \sin(\beta_n x) \sin(\beta_m x) = \delta_{mn} \frac{\sin^2 \beta_n}{2}
\]  

in the following relation:

\[
\int_0^{r_0} r \, dr \Delta \rho_m(r,t = 0) \sin(\beta_n r / r_0) = \alpha \rho_0 \int_0^{r_0} r \, dr \delta(r - r_0) \sin(\beta_m r / r_0).
\]  

One finds

\[
c_m = \frac{2 \alpha \rho_0}{\sin \beta_m}.
\]
toward its steady state value as (and the boundary layer) is still very localized on the wall, fact that, at short times after the quench, the excess density of the system. induced pressure, and is independent of the dimensionality characteristic for the short-term characteristic of quench-aries. As discussed in the previous section, this is a universal characteristic for the short-term scaling of the excess density and pressure measured with respect to their steady state is insensitive to the curvature of the bound- 
aries. Therefore, the short-timescaling effect one-dimensional. Therefore, the short-timescaling so that diffusion of the particles away from the boundary is effectively one-dimensional. Therefore, the short-timescaling

Finally the density on the interior surface of the sphere is obtained as

\[ \Delta \rho_{\text{in}}(r = r_0, t) = \left( 3 + 2 \sum_{n=1}^{\infty} e^{-\beta r_0^2} \right) \frac{\alpha \rho_0}{r_0}. \]  

(38)

The summation is over all positive solutions of \( \beta \) in equation (32). Using equations (21) and (38), we can now calculate the pressure exerted from inside on the circle and the sphere, respectively. The scaling behavior is shown in figure 4(a): at small times after the quench, the pressure decays toward its steady state value as \( t^{-1/2} \), similar to the case of a circular wall (\( d = 2 \)). This behavior originates from the fact that, at short times after the quench, the excess density (and the boundary layer) is still very localized on the wall, so that diffusion of the particles away from the boundary is effectively one-dimensional. Therefore, the short-time scaling of the excess density and pressure measured with respect to their steady state is insensitive to the curvature of the bound- 
aries. As discussed in the previous section, this is a universal characteristic for the short-term characteristic of quench-induced pressure, and is independent of the dimensionality of the system.

4.2. Outside the sphere

Using equations (30) and (31), the time-dependent excess density outside the sphere can be found:

\[
\Delta \rho_{\text{out}}(r, t) = \int_{r_0}^{\infty} d\gamma c(\gamma) \mathcal{K} \left( \frac{\gamma}{r_0} \right) e^{-\gamma^2_0}, \\
\mathcal{K}(\gamma, x) = \frac{\gamma}{x} \cos[\gamma(x - 1)] + \sin[\gamma(x - 1)].
\]  

(39)

The unknown coefficient \( c(\gamma) \) is fixed by the initial condition along with equation (39). To this end we need to evaluate the integral relation

\[
\int_{r_0}^{\infty} r^2 d\gamma \Delta \rho_{\text{out}}(r, t = 0) \mathcal{K} \left( \frac{\gamma}{r_0} \right) = \int_{r_0}^{\infty} d\gamma \alpha \rho_0 \delta(r - r_0) \mathcal{K} \left( \frac{\gamma}{r_0} \right).
\]  

(40)

Using the orthogonality relation

\[
\int_{r_0}^{\infty} dx \gamma^2 \mathcal{K}(\gamma, x) \mathcal{K}(\gamma', x) = \frac{\pi}{2} \delta(\gamma - \gamma')(1 + \gamma^2),
\]  

(41)

we find

\[
c(\gamma) = \frac{2 \alpha \rho_0}{\pi r_0} \frac{\gamma}{1 + \gamma^2}.
\]  

(42)

The density on the exterior of the sphere takes the form

\[
\Delta \rho_{\text{out}}(r = r_0, t) = \frac{2 \alpha \rho_0}{\pi r_0} \int_{r_0}^{\infty} d\gamma \frac{\gamma^2}{1 + \gamma^2} e^{-\gamma^2_0} \]

\[ = \frac{\alpha \rho_0}{r_0} \left[ \frac{1}{\sqrt{\pi t}} - e^{-t} \operatorname{erfc}(\sqrt{t}) \right],
\]  

(43)

where \( \operatorname{erfc}(x) \) is the complementary error function [34]. Using equation (43), we can expand the density of the outside at long times, and obtain the long-time behavior of the pressure which reads

\[
\lim_{t \to \infty} \mathcal{P}_{3D}(t) = \frac{r_0^2}{2 \sqrt{\pi}}
\]  

(44)

As for the 2D case, the time-dependent density inside and outside of the sphere can now be used to compute the force exerted on the sphere after the quench. This force is shown in figure 4(b) as a function of time. Figures 5(c) and (d) show that the force exerted on the spheres at very short times has a finite offset value \( F_i = 2 \). Similar to the 2D case and as discussed in appendix B, the quench-induced force immediately after the quench is zero, and approaches a finite value after very short times. Moreover, as discussed in the previous section, this sudden increase is an interesting non-equilibrium effect associated with the curvature of the walls. A comparison to figure 2 shows that the late time decay of the force toward its steady-state value follows \( r^{-1} \) in 2D, but \( r^{-3/2} \) in 3D. Accordingly, we can generalize these results for \( d \)-dimensional spher-
The distance between the surfaces of the two spheres is defined as \( \Delta r = r_2 - r_1 \). We note that in the case of flat boundaries, the scaling does not depend on the dimension, and the force approaches the steady state as \( F(t) \rightarrow F(t \rightarrow \infty) \propto \bar{t}^{-d/2} \) for large \( \bar{t} \). However, for spherical boundaries, the force as \( \bar{t} \rightarrow \infty \) becomes \( \propto \bar{t}^{-\frac{3}{4}} \). The force per unit length for a sphere approaches its steady-state value as \( \propto \bar{t}^{-\frac{1}{2}} \). For large spheres, the force per unit length decreases as \( \propto \bar{t}^{-\frac{3}{4}} \). The force per unit area for spheres approaches its steady-state value as \( \propto \bar{t}^{-\frac{1}{2}} \). For large spheres, the force per unit volume decreases as \( \propto \bar{t}^{-1} \). The force per unit volume for spheres approaches its steady-state value as \( \propto \bar{t}^{-1} \).

4.3. Quench of medium between two spheres

In the last part of this section, we consider a system of non-interacting Brownian particles confined between two spheres with radii \( r_1 \) and \( r_2 \), respectively; see figure 1(d). The distance between the surfaces of the two spheres is defined as \( \Delta r = r_2 - r_1 \). The solution of the post-quench dynamics of the excess density between two spheres is presented with details in appendix D, from which we obtain the pressure and forces on the two spheres as shown in figure 5. As it can be seen in figure 5(a), the force exerted on the small and large sphere decay as \( \bar{t}^{-1/2} \) at small times, for different values of \( r_1 \) and \( r_2 \). The same scaling was observed for the 2D case and also for the single sphere as shown in figures 3(a) and 4(a), respectively. Figure 5(b) shows that the force exerted on the large sphere approaches its steady-state value as \( \bar{t}^{-\frac{3}{4}} \) at long times. Finally, figures 5(c) and (d) represent the time evolution of the forces acting on the two concentric spheres; these are qualitatively very similar to those of the two circles shown in figure 3. In particular, the forces suddenly increase from zero to a finite value \( (F_1, 2 = 2) \) at very short times and the force exerted on the small sphere exhibits an overshoot for large enough values of \( r_1 \). The overshoot becomes more pronounced when \( r_1 \) is increased for a fixed \( \Delta r \).

4.4. Net force exerted on spherical boundaries

So far we have studied the dynamics of the quench-induced contribution to the pressure and force exerted on the boundaries. In particular, the results shown in all the figures have been obtained by setting the initial temperature equal to zero \( (T_1 = 0) \) in the simulations and the numerical evaluations of analytical results. As long as we are interested in the quench-induced force, these results remain unchanged even for \( T_1 \neq 0 \), because this contribution is proportional to the parameter \( \alpha \propto \sqrt{T_1} - \sqrt{T_F} \) as given by equation (A1). The net force exerted on the walls at non-zero temperatures can be found by adding the pre-quench force to the dynamical force that we found in the previous sections. The pre-quench force is related to the surface tension and arises from the inhomogeneity of the density due to the soft wall potential, as well as the curvature of the walls [35]. Such a surface tension is indeed present before and after the quench. The contribution of the quench-induced variations in the density and resulting forces has been taken into account in previous sections. Therefore, to find the net force acting on the walls, we only need to add the contribution of the surface tension before the quench. To this end, we calculate the pressure difference between inside and outside, \( \Delta P_{\text{in}}(t < 0) = P^\text{in}(t < 0) - P^\text{out}(t < 0), \) at
the pre-quench temperature $T_1$, for a $d$-dimensional spherical wall, which reads
\[
\Delta P_d = \left[ \langle F_{d}^{\text{in}} \rangle + \langle F_{d}^{\text{out}} \rangle \right] / A_d
\]
\[
= \frac{\rho_0 k_B T_1}{r_0} \left( d - 1 \right) \sqrt{\frac{2\pi k_B T_1}{\lambda}},
\]
(46)

where $\langle F_{d}^{\text{inout}} \rangle = (dV^{\text{inout}} / dr)$ denotes the average force exerted by the interior/exterior particles on the $d$-dimensional sphere with an area $A_d$. It should be noted that the bounds of the integral for finding the averages $\langle F_{d}^{\text{in}} \rangle$ and $\langle F_{d}^{\text{out}} \rangle$ are $(r_0, \infty)$ and $(0, r_0)$, respectively, which then sum up to give equation (45). Since the integrand above becomes negative for the range $(0, r_0)$, $F_{d}^{\text{out}}$ has a negative value as it acts in the direction of $-\mathbf{r}$, and therefore $F_{d}^{\text{out}} \propto -\langle F_{d}^{\text{out}} \rangle$. The total force acting on a spherical boundary following the quench can then be found by adding the initial force $F_d(t = 0^-) = A_d \Delta P_d$ to the dynamical forces introduced in the previous sections.

Using the definition of the Laplace pressure for a $d$-dimensional sphere, $\Delta P_d = \gamma (d - 1) / r_0$, we can use equation (46) to find the surface tension as
\[
\gamma = \frac{\rho_0 k_B T_1}{r_0} \sqrt{\frac{2\pi k_B T_1}{\lambda}}.
\]
(47)

Note that the surface tension $\gamma$ in $d > 1$ dimension has a dimensionality $[\gamma] = [k_B T] \ell^{-d+1}$.

5. Conclusions

We have studied systems of Brownian particles confined by surfaces in spherical geometries for 2 and 3 spatial dimensions, following a quench in temperature. For all cases considered, the analytical results were shown to be in quantitative agreement with our explicit Brownian dynamics simulations.

In particular, we calculated the time-evolution of the density analytically. This allowed us to find the dynamics of pressures and net forces acting on the boundaries. Short-time scaling of pressures on curved boundaries was shown to be insensitive to the dimension of the system, and universally decays as $t^{-1/2}$ dictated by effective a one-dimensional diffusion. The observations made for forces, which are obtained as the difference of inside and outside pressures on a given boundary, are different. Unlike what is observed for flat boundaries [21], the long-time scaling of the forces exerted on curved boundaries was shown to depend on the dimension of the system as $t^{-d/2}$.

We further note that in our system, the geometry of the curved walls determines these scalings, while for the case of 2D or 3D flat boundaries the forces behave the same as the 1D case studied in reference [21]. Additionally, we show in appendix E that for very large radii the present results for curved geometries recover those obtained for planar geometries, as expected.

Furthermore, we showed that the curvature of the boundary differentiates diffusion of particles on the two sides of the boundary after the quench, i.e., particles close to the boundary and inside a sphere have less space to diffuse than particles close to the outside boundary. As a result, the post-quench force exerted on the curved boundaries, increases to a constant value very rapidly; the constant depends on the dimension of the system.

We have therefore demonstrated that the curvature of confining boundaries has an important (dimension-dependent) effect on non-equilibrium dynamics of an ideal fluid. Our results could, in principle, be used to compute forces acting on boundaries of droplets or bubbles in an ideal fluid. In particular, we expect our model to describe the behavior of colloidal suspensions confined by curved boundaries [36]. In the appropriate coarse-graining and density regimes, our results are also relevant for dynamics of forces exerted on curved membranes due to a quench in activity of active non- or weakly-interacting systems. Indeed, a gas of active particles can be modeled by Brownian motion with an effective temperature, and a quench of the effective temperature of an active medium has been shown to give rise to qualitatively similar effects to a temperature quench in a passive medium [21]. In experiments on passive Brownian particles, sudden changes in the temperature can be achieved by laser. On the other hand, in an active system, employing tunable activity in an experimental set-up similar to that used in reference [29] can lead to an effective temperature quench. However, the role of solvent effects would have to be considered with care. Additionally, the theoretical framework established here sets the basis for computing curvature corrections for forces on planar surfaces. Another interesting case to be studied would be that of non-concentric circles (spheres), where we expect short-time and late-time behaviors of the force to be similar to those in the concentric cases. However, the transient dynamics could be different depending on the distance between the two circles (spheres) centers. It would also be interesting to study the role played by correlations arising from quenches of spherically confined systems in terms of a field-theoretical approach. These questions will be addressed in future work.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
Appendix A. Boundary layer thickness \(\alpha_d\)

In this appendix, we compute the thickness of the nonequilibrium boundary layer formed immediately after the quench which serves as the initial condition for the quench dynamics. Before the quench \((t < 0)\) the system is at equilibrium, which leads to a constant density everywhere but at the wall, where a position-dependent distribution \(\rho(r) \propto \exp[-V_{\text{wall}}(r)/kT]\) is found. In the case of harmonic wall potentials, the density profile in the walls is indeed half of a Gaussian function. Assuming the coarse-grained regime of steep wall potentials, the Gaussian profile becomes very localized and can be approximated by a Dirac delta function. Similarly, at very long times after the quench when another equilibrium is reached, the density becomes constant away from the wall, while in the wall we have \(\rho(r) \propto \exp[-V_{\text{wall}}(r)/kT]\), which can be again approximated by a delta function. Based on superposition and particle conservation, we only need to consider the evolution of the nonequilibrium or quench-induced part of the boundary layer which accounts for the extra adsorption/desorption of particles at the wall immediately after the quench. The thickness of this nonequilibrium boundary layer is, therefore, given by the difference of the pre-quench and the long-term steady values of the density as first discussed in reference [21].

For the circle we have

\[
\alpha_2 = \frac{1}{r_0} \int_0^\infty r^2 dr [e^{-V_{\text{wall}}(r)/kT_I} - e^{-V_{\text{wall}}(r)/kT_F}] 
\approx \sqrt{\frac{\pi k_B}{2\lambda}} (\sqrt{T_I} - \sqrt{T_F}), \tag{A1}
\]

where in the final step we have used the approximation \(\sqrt{\frac{2k_B}{3}} \ll \sqrt{\frac{2k_B}{2\lambda}} \ll r_0\) which means we have considered system sizes much larger than the characteristic width of the boundary layer. To calculate the coefficient \(\alpha_3\), we need to integrate the initial and final density in 3D. We use the same approximation for the system size and find:

\[
\alpha_3 = \frac{1}{r_0} \int_0^\infty r^2 dr [e^{-V_{\text{wall}}(r)/kT_I} - e^{-V_{\text{wall}}(r)/kT_F}] 
\approx \sqrt{\frac{\pi k_B}{2\lambda}} (\sqrt{T_I} - \sqrt{T_F}), \tag{A2}
\]

which is equal to the coefficient \(\alpha_2\). For this reason we have used \(\alpha\) instead of \(\alpha_d\) in the main text.

Appendix B. Explicit derivation of the initial offset values of the quench-induced force

In sections 3 and 4, we have seen that the forces acting on the walls always have an initial offset of \(F = 1, 2\) in 2D and 3D, respectively. There, we have argued that, in fact, the quench-induced force at \(t = 0^+\) should be zero and therefore the apparent offset values are the result of the Dirac delta approximation for the initial excess densities. In practice, starting from initial excess densities with a finite but small width, one can see that the force very rapidly (but not immediately) approaches these offset values. Indeed, it is natural to consider the coarse-grained model primarily at time scales beyond the dynamical processes occurring directly at the wall immediately after the quench.

Here, we show explicitly that the force exerted on a sphere is indeed equal to zero at \(t = 0^+\), but very rapidly reaches a value \(F_{3D} = 2\), thereafter it evolves toward its steady state value. We also discuss the reason for this rapid increase, which is absent in a system with flat boundaries. We consider the initial condition for the excess density as

\[
\Delta \rho(r, t = 0) = \begin{cases} \frac{\alpha \rho_0}{r_0} & r - r_0 < \frac{\epsilon}{2} \\ 0 & \text{otherwise} \end{cases}
\] \tag{B1}

We take \(\epsilon\) to be very small (so that the expression approximates a \(\delta\) function), and use this initial condition to find the unknown coefficients in the density expansion in equations (33) and (39). The density on the interior of the sphere at \(t = 0\) is then

\[
\Delta \rho_{\text{in}} = \frac{\alpha \rho_0}{r_0} \left[ 3 + 2 \sum_{m=1}^{\infty} \Lambda_m - \beta_m \epsilon \cos(\beta_m \epsilon) \right], \tag{B2}
\]

where the coefficients \(\beta_m\) are the positive solutions of equation (32). The summation in equation (B2) can be approximated with an integral based on the Euler–Maclaurin formula and the fact that \(\epsilon \ll 1\) [37]. To this end, we first add \(\epsilon - \epsilon^2 + \epsilon^3/3\), which can be thought of as the \(m = 0\) term of the summation (corresponding to \(\beta_0 \rightarrow 0\)). Then, noticing that the distance between successive roots \(\beta_m\) of equation (32) is very close to \(\pi\) for large \(m\), we use the conversion \(\sum_{m=0}^{\infty} \rightarrow \int d\beta/\pi\) which yields

\[
\Delta \rho_{\text{in}} = \frac{\alpha \rho_0}{r_0} \left[ 3 + \frac{2}{\epsilon} \int_0^\pi \frac{d\beta}{\pi} \omega(\beta) - \beta \epsilon \cos(\beta \epsilon) \right] - \frac{2}{\epsilon} \left( \epsilon - \epsilon^2 + \frac{\epsilon^3}{3} \right) + O(\epsilon), \tag{B3}
\]

and eventually

\[
\Delta \rho_{\text{in}} = \frac{\alpha \rho_0}{r_0} \frac{1}{\epsilon} + O(\epsilon). \tag{B4}
\]

Similarly and from equation (39), the density exterior to the sphere at \(t = 0\) is found to be

\[
\Delta \rho_{\text{out}} = \frac{\alpha \rho_0}{r_0} \left[ 2 \int_0^\pi \frac{d\beta}{\pi} \frac{\Omega(\beta) - \beta \epsilon \cos(\beta \epsilon)}{\beta (1 + \beta^2)} \right], \tag{B5}
\]

\[
\Omega(\beta) = \sin \beta \epsilon[1 + \beta^2(1 + \epsilon)],
\]

which, after evaluating the integral, leads to

\[
\Delta \rho_{\text{out}} = \frac{\alpha \rho_0}{r_0} \frac{1}{\epsilon}. \tag{B6}
\]

Therefore the force exerted on the sphere immediately following the quench is proportional to \(\Delta \rho_{\text{in}} - \Delta \rho_{\text{out}} \propto \epsilon\) which means the force is equal to zero at \(t = 0\) for \(\epsilon \rightarrow 0\).
As time evolves from zero, the force increases very rapidly to the value \( F = 2 \), after which the approach to steady state continues as discussed in the main text. Assuming \( (e/r_0)^2 \ll 1 \), we first take the limit of \( e \to 0 \) and then the limit of \( t \to 0 \) in equations (38) and (39) from which we obtain

\[
\Delta \rho_{out}(r = r_0, t \to 0) = \frac{\alpha \rho_0}{r_0} \left[ 3 + \sum_{m=1}^{\infty} 2 \right], \tag{B7}
\]

\[
\Delta \rho_{out}(r = r_0, t \to 0) = \frac{2\alpha \rho_0}{r_0} \int_0^{\infty} \frac{d\beta}{\pi} \frac{1}{1+\beta^2}. \tag{B8}
\]

Converting the summation in internal density to an integral in the same way explained above, we can then calculate the force exerted at the sphere for finite and small values of the time as:

\[
F = \frac{r_0}{\alpha \rho_0} [\Delta \rho_{in}(r = r_0, t \to 0) - \Delta \rho_{out}(r = r_0, t \to 0)]
\]

\[
= 1 + \frac{2}{\pi} \left( \int_0^{\infty} d\beta - \int_0^{\infty} \frac{d\beta}{1+\beta^2} \right) = 2, \tag{B9}
\]

which matches very well with the inset of figure 4(b). This rapid change in the force acting on the curved surface of the sphere is absent for flat boundaries. In fact, due to the curvature of the boundary, particles on the inner surface of the sphere are more confined compared to the particles on the external surface. As a result, at a finite time, the interior particles diffuse into the wall more rapidly. This causes a rapid increase in the force acting on the boundary. A similar process occurs for the force acting on the circle in 2D.

**Appendix C. Solution for the region between two circles**

Using the solution found for the density in equation (14), and imposing no-flux boundary conditions on both circles, \( \partial_r X(r)|_{r=r_1,r_2} = 0 \), we can find discrete values for the parameter \( \beta' \) from the following equation:

\[
J_1(\beta'_n r_1) Y_1(\beta'_n r_2) = J_0(\beta'_n r_1) Y_0(\beta'_n r_2), \quad n = 1, 2, 3, \ldots \tag{C1}
\]

The time-dependent solution for the density can be written as

\[
\Delta \rho(r, t) = \sum_{n=1}^{\infty} c_n f_n(r) e^{-\beta'_n^2 r_0 t} + c_0, \tag{C2}
\]

\[
f_n(r) = J_0(\beta'_n r_1) Y_1(\beta'_n r_2) - J_1(\beta'_n r_1) Y_0(\beta'_n r_2),
\]

where the summation runs over all positive solutions of \( \beta' \) in equation (C1). The initial condition, which corresponds to excess particle layers at both surfaces, has the form of \( \Delta \rho(r, t = 0) = \alpha \rho_0 [\delta(r - r_1) + \delta(r - r_2)] \), and the constant \( c \) can be found by calculating the following integrals related to conservation of the particle number:

\[
\int_{r_1}^{r_2} r \, dr [c_n f_n(r) + c_0] = \alpha \int_{r_1}^{r_2} r \, dr [\delta(r - r_1) + \delta(r - r_2)]. \tag{C3}
\]

This gives

\[
c_0 = \frac{2\alpha \rho_0}{r_2 - r_1}. \tag{C4}
\]

For calculating coefficients \( c_n \) we need to calculate below integrals:

\[
\int_{r_1}^{r_2} r \, dr \alpha \rho_0 [\delta(r - r_1) + \delta(r - r_2)] f_m(r)
\]

\[
= \int_{r_1}^{r_2} r \, dr \sum_{n=1}^{\infty} c_n f_n(r) + c_0 \right] f_m(r), \tag{C5}
\]

which gives

\[
c_n = \frac{2\alpha \rho_0}{r_2 f_m(r_2) - r_1 f_m(r_1)}. \tag{C6}
\]

In the last step above we have used the orthogonality relation

\[
\int_{r_1}^{r_2} r \, dr f_m(r) f_n(r) = \delta_{mn} \left[ r_2^2 \left( \frac{Y_2(\beta'_n r_2)}{\beta'_n} - \frac{Y_1(\beta'_n r_2)}{\beta'_n^2} \right) - r_1^2 \left( \frac{Y_2(\beta'_n r_1)}{\beta'_n} - \frac{Y_1(\beta'_n r_1)}{\beta'_n^2} \right) \right], \tag{C7}
\]

to find the expression (C6) for the coefficients \( c_n \).

**Appendix D. Solution for the region between two spheres**

Using the solution found for the density in equation (30), subject to no-flux boundary conditions on the surface of two spheres, \( \partial_r X(r)|_{r=r_1,r_2} = 0 \), we can find discrete values for the parameter \( \beta' \) from the following equation:

\[
\frac{\Delta r \beta'_n}{\beta'_n r_1 r_2} + 1 = \tan(\Delta r \beta'_n), \quad n = 1, 2, 3, \ldots \tag{D1}
\]

The time-dependent solution for the density follows

\[
\Delta \rho(r, t) = \sum_{n=1}^{\infty} c_n e^{-\beta'_n^2 r_0 t} f_n(r) + c_0, \tag{D2}
\]

\[
f_n(r) = \sin(\beta'_n r) + a_n \cos(\beta'_n r), \tag{D3}
\]

with

\[
a_n = \frac{\beta'_n r_1 - \tan(\beta'_n r_1)}{1 + \beta'_n^2 r_1 \tan(\beta'_n r_1)} = \frac{\beta'_n r_2 - \tan(\beta'_n r_2)}{1 + \beta'_n^2 r_2 \tan(\beta'_n r_2)}. \tag{D4}
\]

The initial condition for this problem has the form \( \Delta \rho(r, t = 0) = \alpha \rho_0 [\delta(r - r_1) + \delta(r - r_2)] \), where \( c_0 \) is fixed by calculating

\[
\int_{r_1}^{r_2} r \, dr [c_n f_n(r) + c_0]
\]

\[
= \int_{r_1}^{r_2} r \, dr \alpha \rho_0 [\delta(r - r_1) + \delta(r - r_2)]. \tag{D5}
\]
One finds
\[ c_0 = \frac{3\alpha\rho_0 (r_1^2 + r_2^2)}{r_2^2 - r_1^2}. \] (D6)

For the coefficients \( c_m \), we need to compute
\[ \int r_1^{r_2} r \, dr \rho_0 \left[ \delta(r - r_1) + \delta(r - r_2) \right] f_m(r) \]
\[ = \int r_1^{r_2} r \, dr \rho(r, t = 0) f_m(r), \] (D7)

which gives
\[ c_m = 2\alpha\rho_0 \frac{r_2 f_m(r_2) + r_1 f_m(r_1)}{\int r_1^{r_2} r f_m^2(r) \, dr}, \] (D8)

To find the coefficients \( c_m \), we have used the orthogonality condition
\[ \int r_1^{r_2} \, dr f_m(r) f_m(r) = \delta_{m,0}, \] (D9)

\section*{Appendix E. Asymptotic convergence to the case of flat surfaces}

In this section, we explicitly show that our results asymptotically approach the flat geometry results of the previous study \cite{21}, by taking the limit of large radii \((R_d \to \infty)\). We first consider the circular case where we find that the density follows equation (26). Recalling that the dimensionless time for a single circle is \( t = D\tau/\tau_0 \), and changing the integration parameter to \( \tau = \sqrt{D\tau_0/\rho_0} \), we can re-write equation (26) as
\[ \Delta \rho_{\text{out}}(r_0, t) = \frac{4\alpha\rho_0}{\pi^2 r_0} \int_0^\infty d\gamma \frac{e^{-\gamma^2}}{\gamma \sqrt{1 + \frac{\gamma^2}{\tau_0} + \frac{\gamma^2}{\tau_0}}} \] (E1)

Taking the limit of \( r_0 \to \infty \), we can replace Bessel functions with their asymptotic forms
\[ J_1(\gamma) \sim \sqrt{\frac{2}{\pi \gamma}} \sin(\gamma - \pi/4), \] (E2)
\[ Y_1(\gamma) \sim -\sqrt{\frac{2}{\pi \gamma}} \cos(\gamma - \pi/4), \] (E3)

which results in
\[ \Delta \rho_{\text{out}}(r_0 \to \infty, t) = \frac{4\alpha\rho_0}{\pi^2 r_0} \frac{\rho_0}{2\sqrt{D}t_0} \int_0^\infty d\gamma e^{-\gamma^2} = \frac{\alpha\rho_0}{\pi\sqrt{D}t_0}, \] (E4)

In a similar manner, we can find the asymptotic behavior for the spherical case given by equation (43). Using the new integration variable \( \gamma \), the density becomes
\[ \Delta \rho_{\text{out}}(r = r_0, t) = \frac{2\alpha\rho_0}{\pi\sqrt{D}t_0} \int_0^\infty d\gamma e^{-\gamma^2} \frac{r_2^2 - r_1^2}{1 + r_0^2 \gamma^2}, \] (E5)

which, by taking the limit of \( r_0 \to \infty \), again reduces to \( \alpha\rho_0/\sqrt{\pi D}t \). These results are therefore consistent with the direct calculations for the case of planar surfaces.

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